quantitatively. Thus, the concentration of elements in the third region 103 and the second region 102 present at a depth of approximately 5 nm from the surface can be analyzed quantitatively.

[0142]

When the surface of the positive electrode active material 100 is subjected to the XPS analysis and the concentration of the first transition metal is defined as 1, a relative value of the concentration of the second transition metal is preferably greater than or equal to 0.05 and less than or equal to 0.4, further preferably greater than or equal to 0.1 and less than or equal to 0.3. In addition, a relative value of the concentration of the representative element is preferably greater than or equal to 0.4 and less than or equal to 1.5, further preferably greater than or equal to 0.45 and less than or equal to 1.00. Furthermore, a relative value of the concentration of fluorine is preferably greater than or equal to 0.05 and less than or equal to 1.5, further preferably greater than or equal to 0.3 and less than or equal to 1.00.

[0143]

Note that, as described above, elements contained in the first region 101, the second region 102, and the third region 103 may each have a concentration gradient; thus, the first region 101 may contain the element in the second region 102 or the third region 103, such as fluorine. Similarly, the third region 103 may contain the element in the first region 101 or the second region 102. In addition, the first region 101, the second region 102, and the third region 103 may each contain another element such as carbon, sulfur, silicon, sodium, calcium, chlorine, or zirconium.

[0144]

[Particle diameter]

If the particle diameter of the positive electrode active material 100 is too large, diffusion of lithium is difficult, whereas if the particle diameter is too small, it is difficult to maintain a crystal structure described later. Thus, D50 (also referred to as a median diameter) is preferably 5 μm or more and 100 μm or less, and further preferably 10 μm or more and 70 μm or less. In the case where the coating film is formed on the surface of the positive electrode active material 100 by a spray dry apparatus in a later step, it is preferable that the nozzle diameter and the maximum particle diameter of the positive electrode active material 100 be substantially the same. When the particle diameter is less than 5 μm and a spray dry apparatus having a nozzle diameter of 20 μm is used, secondary particles are covered collectively, which leads to a decrease in coverage.

[0145]

To increase the density of the positive electrode active material layer, it is effective to mix large particles (the longest portion is approximately 20 μm or more and 40 μm or less) and small particles (the longest portion is approximately 1 μm) and embed spaces between the large particles

with the small particles. Thus, there may be two peaks of particle size distribution.

[0146]

The particle size of the positive electrode active material is influenced not only by the particle sizes of starting materials but also by a ratio between lithium and the first transition metal (hereinafter expressed as a ratio of Li to the first transition metal) which are contained in the starting material.

[0147]

In the case where the particle size of the starting material is small, the grain growth needs to be performed at the time of baking so that the grain size of the positive electrode active material is in the above-described preferred range.

[0148]

To promote the grain growth at the time of baking, it is effective to make the ratio of Li to the first transition metal of the starting material larger than 1, that is, to make the amount of lithium a little larger. For example, when the ratio of Li to the first transition metal is approximately 1.06, a positive electrode active material in which D50 is larger than or equal to 15 μm is easily obtained. Note that, as described later, lithium may be lost to the outside of a system in the formation process of the positive electrode active material; thus, the ratio between lithium and the first transition metal of the obtained positive electrode active material does not agree with the ratio between lithium and the first transition metal of the starting material in some cases.

[0149]

However, if the amount of lithium is too large to make the particle size be in the preferred range, the capacity retention rate of a secondary battery containing the positive electrode active material might be decreased.

[0150]

Then, the present inventors found that with the second region 102 containing the second transition metal in the superficial portion, the particle size can be in the preferred range by control of the ratio of Li to the first transition metal and a positive electrode active material having high capacity retention rate can be formed.

[0151]

In the positive electrode active material of one embodiment of the present invention including a region containing the second transition metal in the superficial portion, the ratio of Li to the first transition metal in the starting material is preferably greater than or equal to 1.00 and less than or equal to 1.07, further preferably greater than or equal to 1.03 and less than or equal to 1.06.

[0152]

[Formation of second region]

The second region 102 can be formed by coating particles of the composite oxide of lithium and the first transition metal with a material containing the second transition metal.

[0153]

As the coating method of the material containing the second transition metal, a liquid phase method such as a sol-gel method, a solid phase method, a sputtering method, an evaporation method, a chemical vapor deposition (CVD) method, a pulsed laser deposition (PLD) method, or the like can be used. In this embodiment, the case where the sol-gel method which can be performed with a uniform coverage under an atmospheric pressure is used is described.

[0154]

<Sol-gel method>

A method for forming a material containing the second transition metal using a sol-gel method is described with reference to FIGS. 4A-1, 4A-2, 4A-3, 4B, 4C, 4D-1, and 4D-2.

[0155]

First, an alkoxide of the second transition metal is dissolved in alcohol.

[0156]

FIG. 4A-1 shows a general formula of the alkoxide of the second transition metal. In the formula of FIG. 4A-1, M2 indicates the alkoxide of the second transition metal. R represents an alkyl group having 1 to 18 carbon atoms or a substituted or unsubstituted aryl group having 6 to 13 carbon atoms. Although FIG. 4A-1 shows the general formula in which the second transition metal has a valence of 4, one embodiment of the present invention is not limited thereto. The second transition metal may have a valence of 2, a valence of 3, a valence of 5, a valence of 6, or a valence of 7. In this case, the alkoxide of the second transition metal includes an alkoxy group corresponding to the valence of the second transition metal.

[0157]

FIG. 4A-2 shows a general formula of the titanium alkoxide used when titanium is used as the second transition metal. R in FIG. 4A-2 represents an alkyl group having 1 to 18 carbon atoms or a substituted or unsubstituted aryl group having 6 to 13 carbon atoms.

[0158]

As the titanium alkoxide, tetramethoxytitanium, tetraethoxytitanium, tetra-*n*-propoxytitanium, tetra-*i*-propoxytitanium (also referred to as tetraisopropyl orthotitanate, titanium (IV) isopropoxide, titanium tetraisopropoxide (IV), TTIP, and the like), tetra-*n*-butoxytitanium, tetra-*i*-butoxytitanium, tetra-*sec*-butoxytitanium, tetra-*t*-butoxytitanium, or the like can be used.

[0159]

FIG. 4A-3 shows a chemical formula of titanium (IV) isopropoxide (TTIP) described in

a formation method below, which is a kind of titanium alkoxide.

[0160]

As a solvent in which the alkoxide of the second transition metal is dissolved, an alcohol such as methanol, ethanol, propanol, 2-propanol, butanol, or 2-butanol is preferably used.

[0161]

Next, particles of composite oxide of lithium, a transition metal, magnesium, and fluorine are mixed into the alcohol solution of the alkoxide of the second transition metal and stirred in an atmosphere containing water vapor.

[0162]

When the solution is put in an atmosphere containing H_2O, hydrolysis of water and an alkoxide of the second transition metal occurs as in FIG. 4B. Then, as in FIG. 4C, dehydration condensation occurs between the products of FIG. 4B. When the hydrolysis of FIG. 4B and the condensation reaction of FIG. 4C occur repeatedly, a sol of an oxide of the second transition metal is generated. This reaction also occurs on a particle 110 of the composite oxide as in FIGS. 4D-1 and 4D-2, and a layer containing the second transition metal is formed on the surface of the particle 110.

[0163]

After that, the particle 110 is collected, and the alcohol is vaporized. The details of the formation method are described later.

[0164]

Note that in this embodiment, an example in which the particles of the composite oxide of lithium, the first transition metal, the representative element, and fluorine are coated with the material containing the second transition metal before the particles are applied to a positive electrode current collector is described; however, one embodiment of the present invention is not limited thereto. After the positive electrode active material layer including the particles of the composite oxide of lithium, the first transition metal, the representative element, and fluorine is formed on the positive electrode current collector, the positive electrode current collector and the positive electrode active material layer may be both soaked into an alkoxide solution of the second transition metal to be coated with the material containing the second transition metal.

[0165]

[Segregation of third region]

The third region 103 can be formed by a sputtering method, a solid phase method, a liquid phase method such as a sol-gel method, or the like. However, the present inventors found that when a source of a representative element such as magnesium and a source of fluorine are mixed with a material of the first region 101 and then the mixture is heated, the representative element is

segregated on a superficial portion of the positive electrode active material particle to form the third region 103. In addition, they found that with the third region 103 formed in this manner, the positive electrode active material 100 has excellent cycle characteristics.

[0166]

In the case where the third region 103 is formed through the heating as described above, the heating is preferably performed after the particle of the composite oxide is coated with the material containing the second transition metal. This is because even after the particle is coated with the material containing the second transition metal, the representative element such as magnesium is unexpectedly segregated on the surface of the particle when the heating is performed.

[0167]

Segregation models of the representative element are described with reference to FIGS. 5A to 5C and FIGS. 6A to 6D. It is probable that the segregation model of the representative element such as magnesium is slightly different in accordance with the ratio between lithium and the first transition metal contained in a starting material. Then, a segregation model in which the ratio of Li to the first transition metal in the starting material is less than 1.03, that is, the amount of lithium is small, is described with reference to FIGS. 5A to 5C. In addition, a segregation model in which the ratio of Li to the first transition metal in the starting material is greater than or equal to 1.03, that is, the amount of lithium is large, is described with reference to FIGS. 6A to 6D. In these segregation models in FIGS. 5A to 5C and FIGS. 6A to 6D, the first transition metal is cobalt, the second transition metal is titanium, and the representative element is magnesium.

[0168]

FIG. 5A illustrates a model diagram of the vicinity of the surface of the particle 110 of the composite oxide containing lithium, cobalt, magnesium, and fluorine, which is formed at a ratio of Li to Co in the starting material of less than 1.03. A region 111 in the drawings contains lithium, cobalt, magnesium, and fluorine, and lithium cobaltate ($LiCoO_2$) is a main component of the region 111. Lithium cobaltate has a layered rock-salt structure.

[0169]

It is generally known that, at the time of synthesizing particles of the composite oxide containing lithium, cobalt, magnesium, and fluorine, lithium partly moves outside a system (a particle on which lithium is formed). This is because lithium is volatilized at the time of baking, lithium is eluted to a solvent at the time of mixing the starting material, and the like. Thus, the ratio of Li to Co in the particle 110 of the composite oxide containing lithium, cobalt, magnesium, and fluorine becomes smaller than the ratio of Li to Co in the starting material in some cases.

[0170]

When the ratio of Li to Co in the starting material is less than 1.03, on the surface of the

particle 110, lithium is released from the lithium cobaltate and cobalt oxide is easily generated. Thus, as illustrated in FIG. 5A, the surface of the particle 110 of the composite oxide is covered with a cobalt oxide (CoO) layer 114 in some cases.

[0171]

The cobalt oxide has a rock-salt crystal structure. Thus, in the particle 110 in FIG. 5A, the cobalt oxide layer 114 having a rock-salt crystal structure is provided over and in contact with the region 111 containing lithium cobaltate having a layered rock-salt crystal structure in some cases.

[0172]

The particle 110 is coated with a material containing titanium by a sol-gel method or the like. FIG. 5B illustrates a state where the particle 110 is coated with a layer 112 containing titanium by a sol-gel method. At the stage of FIG. 5B, the layer 112 containing titanium is a gel of titanium oxide; thus, the crystallinity is low.

[0173]

Next, the particle 110 coated with the layer 112 containing titanium is heated. Although the details of the heating conditions are described later, for example, FIG. 5C illustrates a state where the particle 110 is heated in an oxygen atmosphere at 800 °C for two hours to form the positive electrode active material 100, which is one embodiment of the present invention. By heating, titanium in the layer 112 containing titanium is diffused into the inside of the particle 110. At the same time, magnesium and fluorine contained in the region 111 are segregated on the surface of the particle 110.

[0174]

As described above, on the surface of the particle 110, cobalt oxide having a rock-salt structure is present. In addition, magnesium oxide also has a rock-salt crystal structure. Thus, it is probable that magnesium is more stable in the state of being present as magnesium oxide on the surface of the particle 110 as compared with the state of being present inside the particle 110. That could be why magnesium is segregated on the surface of the particle 110 when the particle 110 is heated.

[0175]

Moreover, it is considered that fluorine contained in the starting material promotes the segregation of magnesium.

[0176]

Fluorine has higher electronegativity than oxygen. Thus, it is probable that even in a stable compound such as magnesium oxide, when fluorine is added, uneven charge distribution occurs and a bond between magnesium and oxygen is weakened. Furthermore, it is probable that

fluorine is substituted for oxygen in the magnesium oxide, whereby magnesium easily moves around the substituted fluorine.

[0177]

Moreover, this can also be described from a phenomenon in which a melting point of a mixture decreases. When magnesium oxide (melting point: 2852 °C) and lithium fluoride (melting point: 848 °C) are added at the same time, the melting point of the magnesium oxide is lowered. It is considered that the melting point is lowered, whereby magnesium easily moves at the time of heating, and the segregation of magnesium easily occurs.

[0178]

Lastly, the third region 103 becomes a solid solution of cobalt oxide and magnesium oxide which has a rock-salt crystal structure. Furthermore, fluorine is probably substituted for part of oxygen contained in the cobalt oxide and the magnesium oxide.

[0179]

Cobalt sites of lithium cobaltate are substituted for part of the diffused titanium and lithium titanate is substituted for another part of the diffused titanium. The second region 102 after the heating contains lithium titanate having a rock-salt crystal structure.

[0180]

The first region 101 after the heating contains lithium cobaltate having a layered rock-salt crystal structure.

[0181]

Next, the case where the ratio of Li to Co in the starting material is larger than or equal to 1.03 is described with reference to FIGS. 6A to 6D. FIG. 6A illustrates a model diagram of the vicinity of the surface of the particle 120 of the composite oxide containing lithium, cobalt, magnesium, and fluorine, which is formed at a ratio of Li to Co in the starting material of greater than or equal to 1.03. A region 121 in the drawings contains lithium, cobalt, magnesium, and fluorine.

[0182]

Since the particle 120 in FIG. 6A contains a sufficient amount of lithium, even when lithium is released from the particle 120 at the time of baking the particle 120 of the composite oxide of lithium, cobalt, magnesium, and fluorine or the like, lithium is diffused from the inside of the particle 120 to the surface thereof to compensate; as a result, a cobalt oxide layer is not easily formed on the surface.

[0183]

FIG. 6B illustrates a state where the particle 120 in FIG. 6A is coated with a layer 122 containing titanium by a sol-gel method. At the stage of FIG. 6B, the layer 122 containing

titanium is a gel of titanium oxide; thus, the crystallinity is low.

[0184]

FIG. 6C illustrates the state where the particle 120 coated with the layer 122 containing titanium in FIG. 6B starts to be heated. By heating, titanium in the layer 122 containing titanium is diffused into the inside of the particle 110. The diffused titanium is bonded with lithium contained in the region 121 to become lithium titanate, and a layer 125 containing the lithium titanate is formed.

[0185]

Since lithium is bonded with titanium to form lithium titanate, the amount of lithium is relatively insufficient at the surface of the particle 120. Thus, it is probable that, as illustrated in FIG. 6C, a cobalt oxide layer 124 is temporarily formed on the surface of the particle 120.

[0186]

FIG. 6D illustrates the state where the state of FIG. 6C is sufficiently heated to form the positive electrode active material 100, which is one embodiment of the present invention. It is considered that, since the cobalt oxide layer 124 having a rock-salt crystal structure is present on the surface, magnesium is more stable in the state of being present as magnesium oxide on the surface of the particle 120 as compared with the state of being present inside the particle 120. As in the case of FIGS. 5A to 5C, fluorine promotes the segregation of magnesium.

[0187]

Thus, as illustrated in FIG. 6D, magnesium and fluorine contained in the region 121 are segregated on the surface to be the third region 103 with the cobalt oxide.

[0188]

In this manner, the positive electrode active material 100, which includes the third region 103 containing magnesium oxide and cobalt oxide, the second region 102 containing lithium titanate, and the first region 101 containing lithium cobaltate, is formed.

[0189]

Note that in the case where the representative element is segregated by heating, when the composite oxide containing lithium and the first transition metal included in the first region 101 is a polycrystal or has crystal defects, the representative element can be segregated not only in the superficial portion but also in the vicinity of a grain boundary of the composite oxide containing lithium and the first transition metal or in the vicinity of crystal defects thereof. The representative element segregated in the vicinity of a grain boundary or in the vicinity of crystal defects can contribute to further improvement in stability of the crystal structure of the composite oxide containing lithium and the first transition metal included in the first region 101.

[0190]

When the composite oxide containing lithium and the first transition metal included in the first region 101 includes a crack portion, the representative element is also segregated in the crack portion by heating. In addition, not only the representative element but also the second transition metal can be segregated. The crack portion is in contact with the electrolyte solution like the surface of the particle. Thus, the representative element and the second transition metal are segregated in the crack portion, and the third region 103 and the second region 102 are generated, whereby a chemically stable material can be used for the region in contact with the electrolyte solution. As a result, a secondary battery having excellent cycle characteristics can be provided.

[0191]

The ratio between a representative element (T) and fluorine (F) in a starting material is preferably in a range of T:F = 1:x ($1.5 \leq x \leq 4$) (atomic ratio) because the segregation of the representative element effectively occurs. Further preferably, the ratio between T and F is approximately 1:2 (atomic ratio).

[0192]

Since the third region 103 formed by segregation is formed by epitaxial growth, orientations of crystals in the second region 102 and the third region 103 are partly and substantially aligned with each other in some cases. That is, the second region 102 and the third region 103 become topotaxy in some cases. When the orientations of crystals in the second region 102 and the third region 103 are substantially aligned with each other, these regions can serve as a more favorable coating layer.

[0193]

However, not all of the representative elements such as magnesium which is added as a starting material need not be segregated in the third region 103. For example, the first region 101 may contain a small amount of representative element such as magnesium.

[0194]

<Fourth region 104>

In addition, as illustrated in FIG. 1C, the positive electrode active material 100 may include a fourth region 104 on the third region 103. Furthermore, when the positive electrode active material 100 contains a defect such as a crack portion 106, the fourth region 104 may be present to embed the defect such as the crack portion 106.

[0195]

The fourth region 104 contains some elements contained in the second region 102 and the third region 103. For example, the fourth region 104 contains the second transition metal and the representative element.

[0196]

The fourth region 104 may have a projection, a stripe shape, or a layered shape. The fourth region 104 is formed using the second transition metal and the representative element not contained in the second region 102 or the third region 103 of the second transition metals and the representative elements contained in the starting material and the like. That is, with the fourth region 104, the amount of the second transition metal and the representative element contained in the second region 102 and the third region 103 can be kept in an appropriate range and the crystal structures of the second region 102 and the third region 103 can be stabilized in some cases. Moreover, with the fourth region 104, the defect such as the crack portion 106 included in the positive electrode active material 100 can be repaired.

[0197]

The presence of the fourth region 104 and the shape of the fourth region 104 can be observed by a scanning electron microscope (SEM) or the like. Elements contained in the fourth region 104 can be analyzed by SEM-EDX or the like.

[0198]

[Method for forming positive electrode active material]

Next, an example of a method for forming the positive electrode active material 100, which is one embodiment of the present invention, is described.

[0199]

<Step 11: Preparation of starting materials>

First, starting materials are prepared. From the starting materials prepared in this process, the first region 101 and the third region 103 are formed finally.

[0200]

As materials of lithium and the first transition metal contained in the first region 101, a source of lithium and a source of the first transition metal are prepared. In addition, as materials of the compound of the representative elements contained in the third region 103, a source of the representative element is prepared.

[0201]

In addition to these sources, a source of fluorine is preferably prepared. Fluorine used for the materials has an effect of segregating the representative elements contained in the third region 103 on the surface of the positive electrode active material 100 in a later step.

[0202]

As the source of lithium, for example, lithium carbonate and lithium fluoride can be used. As the source of the first transition metal, for example, an oxide of the first transition metal can be used. As the source of the representative element, for example, an oxide of the representative

element contained in the third region and fluoride of the representative element contained in the third region can be used.

[0203]

As the source of fluorine, for example, lithium fluoride and fluoride of the representative element contained in the third region can be used. That is, lithium fluoride can be used as either the source of lithium or the source of fluorine.

[0204]

The amount of fluorine contained in the source of fluorine is preferably 1.0 time to 4 times (atomic ratio), further preferably 1.5 times to 3 times (atomic ratio) the amount of representative element contained in the source of the representative element.

[0205]

<Step 12: Mixing of starting materials>

Next, the source of lithium, the source of the first transition metal, and the source of the representative element are mixed. In addition, the source of fluorine is preferably added. For example, a ball mill and a bead mill can be used for the mixing.

[0206]

<Step 13: First heating>

Next, the materials mixed in Step 12 are heated. In this step, the heating is referred to as baking or first heating in some cases. The heating is preferably performed at higher than or equal to 800 °C and lower than or equal to 1100 °C, further preferably at higher than or equal to 900 °C and lower than or equal to 1000 °C. The heating time is preferably greater than or equal to 2 hours and less than or equal to 20 hours. The baking is preferably performed in a dried atmosphere such as dry air. In the dried atmosphere, for example, the dew point is preferably lower than or equal to −50 °C, further preferably lower than or equal to −100 °C. In this embodiment, the heating is performed at 1000 °C for 10 hours, the temperature rising rate is 200 °C/h, and dry air whose dew point is −109 °C flows at 10 L/min. After that, the heated materials are cooled to room temperature.

[0207]

By the heating in Step 13, the composite oxide of lithium and the first transition metal having a layered rock-salt crystal structure can be synthesized. At this time, the representative element and fluorine contained in the starting materials form a solid solution in the composite oxide. However, some representative elements have been already segregated on the surface of the composite oxide in some cases.

[0208]

In addition, as the starting materials, particles of the composite oxide containing lithium,

cobalt, fluorine, and magnesium which are synthesized in advance may be used. In this case, Step 12 and Step 13 can be omitted. For example, lithium cobalt oxide particles (C-20F, produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD.) can be used as one of the starting materials. The lithium cobalt oxide particle has a diameter of approximately 20 μm and contains fluorine, magnesium, calcium, sodium, silicon, sulfur, and phosphorus in a region which can be analyzed by XPS from the surface.

[0209]

<Step 14: Coating with second transition metal>

Next, the composite oxide of lithium and the first transition metal is cooled to room temperature. Then, the surface of the composite oxide particle of lithium and the first transition metal is coated with a material containing the second transition metal. In the formation method example, a sol-gel method is used.

[0210]

First, the alkoxide of the second transition metal which is dissolved in alcohol and the composite oxide particles of lithium and the first transition metal are mixed.

[0211]

For example, in the case where titanium is used as the second transition metal, TTIP can be used as the alkoxide of the second transition metal. As alcohol, isopropanol can be used, for example.

[0212]

Next, the above mixed solution is stirred in an atmosphere containing water vapor. The stirring can be performed with a magnetic stirrer, for example. The stirring time is not limited as long as water and TTIP in an atmosphere cause hydrolysis and polycondensation reaction. For example, the stirring can be performed at 25 °C and a humidity of 90 %RH (Relative Humidity) for 4 hours.

[0213]

As described above, when water and TTIP in an atmosphere are reacted, a sol-gel reaction can proceed more slowly as compared with the case where liquid water is added. Alternatively, when titanium alkoxide and water are reacted at room temperature, a sol-gel reaction can proceed more slowly as compared with the case where heating is performed at a temperature higher than the boiling point of alcohol which is a solvent, for example. A sol-gel reaction proceeds slowly, whereby a high-quality coating layer containing titanium with a uniform thickness can be formed.

[0214]

After the above process, precipitate is collected from the mixed solution. As the collection method, filtration, centrifugation, evaporation and drying, or the like can be used. In

this embodiment, filtration is used. For the filtration, a paper filter is used, and the residue is washed by alcohol which is the same as the solvent in which titanium alkoxide is dissolved.

[0215]

Then, the collected residue is dried. In this embodiment, vacuum drying is performed at 70 °C for one hour.

[0216]

<Step 15: Second heating>

Next, the composite oxide particle coated with the material containing the second transition metal, which is formed in Step 14, is heated. This step is referred to as second heating in some cases. In the heating, the retention time within a specified temperature range is preferably shorter than or equal to 50 hours, further preferably longer than or equal to 2 hours and shorter than or equal to 10 hours, still further preferably longer than or equal to 1 hour and shorter than or equal to 3 hours. If the heating time is too short, there is concern that the segregation of the representative elements does not occur; however, if the heating time is too long, there is concern that the favorable second region 102 is not formed because diffusion of the second transition metal proceeds too much.

[0217]

The specified temperature is preferably higher than or equal to 500 °C and lower than or equal to 1200 °C, further preferably higher than or equal to 800 °C and lower than or equal to 1000 °C. If the specified temperature is too low, there is concern that the segregation of the representative elements and the second transition metal does not occur. However, if the specified temperature is too high, there is concern that the first transition metal in the composite oxide particle is reduced to decompose the composite oxide particle, that a layered structure of lithium and the first transition metal in the composite oxide particle cannot be kept, and the like.

[0218]

In this embodiment, the specified temperature is 800 °C and kept for 2 hours, the temperature rising rate is 200 °C/h, and the flow rate of dry air is 10 L/min.

[0219]

By the heating in Step 15, the composite oxide of lithium and the first transition metal and the oxide of the second transition metal covering the composite oxide become topotaxy. In other words, the first region 101 and the second region 102 become topotaxy.

[0220]

By the heating in Step 15, the representative elements which form a solid solution inside the composite oxide particle of lithium and the first transition metal are unevenly distributed on the surface to form a solid solution, that is, the representative elements are segregated, the

compound of the representative elements is formed, and the third region 103 is formed. At this time, the compound of the representative elements is formed by heteroepitaxial growth from the second region 102. That is, the second region 102 and the third region 103 become topotaxy.

[0221]

Since the second region 102 and the third region 103 contain crystals whose orientations are substantially aligned with each other and have a stable bond with the first region 101, when the positive electrode active material 100 is used for the secondary battery, a change of the crystal structure in the first region 101 which is caused by charging and discharging can be effectively suppressed. Even when lithium is released from the first region 101 due to charging, the superficial portion having a stable bond can suppress release of oxygen and the first transition metal such as cobalt from the first region 101. Furthermore, a chemically stable material can be used for a region in contact with the electrolyte solution. Thus, a secondary battery having excellent cycle characteristics can be provided.

[0222]

Note that the entire first region 101 and second region 102 does not need to become topotaxy as long as part of the first region 101 and second region 102 becomes topotaxy. Furthermore, the entire second region 102 and third region 103 does not need to become topotaxy as long as part of the second region 102 and third region 103 becomes topotaxy.

[0223]

In the case where the compound of the representative elements contained in the third region contains oxygen, the heating in Step 15 is preferably performed in an atmosphere containing oxygen. Heating in an atmosphere containing oxygen promotes the formation of the third region 103.

[0224]

Furthermore, fluorine contained in the starting materials promotes the segregation of the representative elements.

[0225]

In this manner, in the method for forming the positive electrode active material of one embodiment of the present invention, after the elements forming the second region 102 are coated, heating is performed to form the third region 103, and two kinds of regions can be formed on the surface of the positive electrode active material 100. That is, in general, two coating steps are necessary for providing two kinds of regions in a superficial portion; however, in the method for forming the positive electrode active material of one embodiment of the present invention, only one coating step (sol-gel process) is needed, which is a formation method with high productivity.

[0226]

<Step 16: Cooling>

Next, the particles heated in Step 15 are cooled to room temperature. The time of decreasing temperature is preferably long because topotaxy is easily generated. For example, the time of decreasing temperature from retention temperature to room temperature is preferably the same as the time of increasing temperature or longer, specifically longer than or equal to 10 hours and shorter than or equal to 50 hours.

[0227]

<Step 17: Collecting>

Next, the cooled particles are collected. Moreover, the particles are preferably made to pass through a sieve. Through the above process, the positive electrode active material 100 including the first region 101, the second region 102, and the third region 103 can be formed.

[0228]

This embodiment can be implemented in appropriate combination with any of the other embodiments.

[0229]

(Embodiment 2)

In this embodiment, examples of materials which can be used for a secondary battery containing the positive electrode active material 100 described in the above embodiment are described. In this embodiment, a secondary battery in which a positive electrode, a negative electrode, and an electrolyte solution are wrapped in an exterior body is described as an example.

[0230]

[Positive electrode]

The positive electrode includes a positive electrode active material layer and a positive electrode current collector.

[0231]

<Positive electrode active material layer>

The positive electrode active material layer contains at least a positive electrode active material. The positive electrode active material layer may contain, in addition to the positive electrode active material, other materials such as a coating film of the active material surface, a conductive additive, and a binder.

[0232]

As the positive electrode active material, the positive electrode active material 100 described in the above embodiment can be used. When the above-described positive electrode active material 100 is used, a secondary battery with high capacity and excellent cycle characteristics can be obtained.

[0233]

Examples of the conductive additive include a carbon material, a metal material, and a conductive ceramic material. Alternatively, a fiber material may be used as the conductive additive. The content of the conductive additive with respect to the total amount of the active material layer is preferably greater than or equal to 1 wt% and less than or equal to 10 wt%, more preferably greater than or equal to 1 wt% and less than or equal to 5 wt%.

[0234]

A network for electric conduction can be formed in the active material layer by the conductive additive. The conductive additive also allows maintaining of a path for electric conduction between the positive electrode active material particles. The addition of the conductive additive to the active material layer increases the electric conductivity of the active material layer.

[0235]

Examples of the conductive additive include natural graphite, artificial graphite such as mesocarbon microbeads, and carbon fiber. Examples of carbon fiber include mesophase pitch-based carbon fiber, isotropic pitch-based carbon fiber, carbon nanofiber, and carbon nanotube. Carbon nanotube can be formed by, for example, a vapor deposition method. Other examples of the conductive additive include carbon materials such as carbon black (e.g., acetylene black (AB)), graphite (black lead) particles, graphene, and fullerene. Alternatively, metal powder or metal fibers of copper, nickel, aluminum, silver, gold, or the like, a conductive ceramic material, or the like can be used.

[0236]

Alternatively, a graphene compound may be used as the conductive additive.

[0237]

A graphene compound has excellent electrical characteristics of high conductivity and excellent physical properties of high flexibility and high mechanical strength. Furthermore, a graphene compound has a planar shape. A graphene compound enables low-resistance surface contact. Furthermore, a graphene compound has extremely high conductivity even with a small thickness in some cases and thus allows a conductive path to be formed in an active material layer efficiently even with a small amount. For this reason, it is preferable to use a graphene compound as the conductive additive because the area where the active material and the conductive additive are in contact with each other can be increased. It is preferable to form the graphene compound serving as a conductive additive as a coating film to cover the entire surface of the active material with a spray dry apparatus, in which case the electrical resistance may be reduced. Here, it is particularly preferable to use, for example, graphene, multilayer graphene, or RGO as a graphene

compound. Note that RGO refers to a compound obtained by reducing graphene oxide (GO), for example.

[0238]

In the case where an active material with a small particle diameter (e.g., 1 μm or less) is used, the specific surface area of the active material is large and thus more conductive paths for the active material particles are needed. Thus, the amount of conductive additive tends to increase and the supported amount of active material tends to decrease relatively. When the supported amount of active material decreases, the capacity of the secondary battery also decreases. In such a case, a graphene compound that can efficiently form a conductive path even in a small amount is particularly preferably used as the conductive additive because the supported amount of active material does not decrease.

[0239]

A cross-sectional structure example of an active material layer 200 containing a graphene compound as a conductive additive is described below.

[0240]

FIG. 7A shows a longitudinal cross-sectional view of the active material layer 200. The active material layer 200 includes positive electrode active material particles 100, a graphene compound 201 serving as a conductive additive, and a binder (not illustrated). Here, graphene or multilayer graphene may be used as the graphene compound 201, for example. The graphene compound 201 preferably has a sheet-like shape. The graphene compound 201 may have a sheet-like shape formed of a plurality of sheets of multilayer graphene and/or a plurality of sheets of graphene that partly overlap with each other.

[0241]

The longitudinal cross section of the active material layer 200 in FIG. 7B shows substantially uniform dispersion of the sheet-like graphene compounds 201 in the active material layer 200. The graphene compounds 201 are schematically shown by thick lines in FIG. 7B but are actually thin films each having a thickness corresponding to the thickness of a single layer or a multi-layer of carbon molecules. The plurality of graphene compounds 201 are formed in such a way as to partly coat or adhere to the surfaces of the plurality of positive electrode active material particles 100, so that the graphene compounds 201 make surface contact with the positive electrode active material particles 100.

[0242]

Here, the plurality of graphene compounds are bonded to each other to form a net-like graphene compound sheet (hereinafter referred to as a graphene compound net or a graphene net). The graphene net covering the active material can function as a binder for bonding active materials.

The amount of a binder can thus be reduced, or the binder does not have to be used. This can increase the proportion of the active material in the electrode volume or weight. That is to say, the capacity of the secondary battery can be increased.

[0243]

Here, it is preferable to perform reduction after a layer to be the active material layer 200 is formed in such a manner that graphene oxide is used as the graphene compound 201 and mixed with an active material. When graphene oxide with extremely high dispersibility in a polar solvent is used for the formation of the graphene compounds 201, the graphene compounds 201 can be substantially uniformly dispersed in the active material layer 200. The solvent is removed by volatilization from a dispersion medium in which graphene oxide is uniformly dispersed, and the graphene oxide is reduced; hence, the graphene compounds 201 remaining in the active material layer 200 partly overlap with each other and are dispersed such that surface contact is made, thereby forming a three-dimensional conduction path. Note that graphene oxide can be reduced either by heat treatment or with the use of a reducing agent, for example.

[0244]

Unlike a conductive additive in the form of particles, such as acetylene black, which makes point contact with an active material, the graphene compound 201 is capable of making low-resistance surface contact; accordingly, the electrical conduction between the positive electrode active material particles 100 and the graphene compounds 201 can be improved with a smaller amount of the graphene compound 201 than that of a normal conductive additive. This increases the proportion of the positive electrode active material particles 100 in the active material layer 200, resulting in increased discharge capacity of the secondary battery.

[0245]

Alternatively, the graphene compound may cover the entire surface of the active material in advance with a spray dry apparatus. After that, at the time of forming the positive electrode active material layer, a graphene compound can be further added to make the conductive path between the active materials more favorable.

[0246]

As the binder, a rubber material such as styrene-butadiene rubber (SBR), styrene-isoprene-styrene rubber, acrylonitrile-butadiene rubber, butadiene rubber, or ethylene-propylene-diene copolymer can be used, for example. Alternatively, fluororubber can be used as the binder.

[0247]

For the binder, for example, water-soluble polymers are preferably used. As the water-soluble polymers, a polysaccharide and the like can be used. As the polysaccharide, a cellulose derivative such as carboxymethyl cellulose (CMC), methyl cellulose, ethyl cellulose,

hydroxypropyl cellulose, diacetyl cellulose, or regenerated cellulose, starch, or the like can be used. It is more preferred that such water-soluble polymers be used in combination with any of the above rubber materials.

[0248]

Alternatively, as the binder, a material such as polystyrene, poly(methyl acrylate), poly(methyl methacrylate) (PMMA), sodium polyacrylate, polyvinyl alcohol (PVA), polyethylene oxide (PEO), polypropylene oxide, polyimide, polyvinyl chloride, polytetrafluoroethylene, polyethylene, polypropylene, polyisobutylene, polyethylene terephthalate, nylon, polyvinylidene fluoride (PVdF), polyacrylonitrile (PAN), ethylene-propylene-diene polymer, polyvinyl acetate, or nitrocellulose is preferably used.

[0249]

A plurality of the above materials may be used in combination for the binder.

[0250]

For example, a material having a significant viscosity modifying effect and another material may be used in combination. For example, a rubber material or the like has high adhesion or high elasticity but may have difficulty in viscosity modification when mixed in a solvent. In such a case, a rubber material or the like is preferably mixed with a material having a significant viscosity modifying effect, for example. As a material having a significant viscosity modifying effect, for example, a water-soluble polymer is preferably used. An example of a water-soluble polymer having an especially significant viscosity modifying effect is the above-mentioned polysaccharide; for example, a cellulose derivative such as carboxymethyl cellulose (CMC), methyl cellulose, ethyl cellulose, hydroxypropyl cellulose, diacetyl cellulose, or regenerated cellulose, or starch can be used.

[0251]

Note that a cellulose derivative such as carboxymethyl cellulose obtains a higher solubility when converted into a salt such as a sodium salt or an ammonium salt of carboxymethyl cellulose, and accordingly, easily exerts an effect as a viscosity modifier. The high solubility can also increase the dispersibility of an active material and other components in the formation of slurry for an electrode. In this specification, cellulose and a cellulose derivative used as a binder of an electrode include salts thereof.

[0252]

The water-soluble polymers stabilize viscosity by being dissolved in water and allow stable dispersion of the active material and another material combined as a binder such as styrene-butadiene rubber in an aqueous solution. Furthermore, a water-soluble polymer is expected to be easily and stably adsorbed to an active material surface because it has a functional group. Many

cellulose derivatives such as carboxymethyl cellulose have functional groups such as a hydroxyl group and a carboxyl group. Because of functional groups, polymers are expected to interact with each other and cover an active material surface in a large area.

[0253]

In the case where the binder covering or being in contact with the active material surface forms a film, the film is expected to serve as a passivation film to suppress the decomposition of the electrolyte solution. Here, the passivation film refers to a film without electric conductivity or a film with extremely low electric conductivity, and can inhibit the decomposition of an electrolyte solution at a potential at which a battery reaction occurs in the case where the passivation film is formed on the active material surface, for example. It is preferred that the passivation film can conduct lithium ions while suppressing electric conduction.

[0254]

<Positive electrode current collector>

The positive electrode current collector can be formed using a material that has high conductivity, such as a metal like stainless steel, gold, platinum, aluminum, or titanium, or an alloy thereof. It is preferred that a material used for the positive electrode current collector not dissolve at the potential of the positive electrode. Alternatively, the positive electrode current collector can be formed using an aluminum alloy to which an element that improves heat resistance, such as silicon, titanium, neodymium, scandium, or molybdenum, is added. Still alternatively, a metal element that forms silicide by reacting with silicon can be used. Examples of the metal element that forms silicide by reacting with silicon include zirconium, titanium, hafnium, vanadium, niobium, tantalum, chromium, molybdenum, tungsten, cobalt, and nickel. The current collector can have any of various shapes including a foil-like shape, a plate-like shape (sheet-like shape), a net-like shape, a punching-metal shape, and an expanded-metal shape. The current collector preferably has a thickness of 5 μm to 30 μm.

[0255]

[Negative electrode]

The negative electrode includes a negative electrode active material layer and a negative electrode current collector. The negative electrode active material layer may contain a conductive additive and a binder.

[0256]

<Negative electrode active material>

As a negative electrode active material, for example, an alloy-based material or a carbon-based material can be used.

[0257]

For the negative electrode active material, an element which enables charge-discharge reactions by an alloying reaction and a dealloying reaction with lithium can be used. For example, a material containing at least one of silicon, tin, gallium, aluminum, germanium, lead, antimony, bismuth, silver, zinc, cadmium, indium, and the like can be used. Such elements have higher capacity than carbon. In particular, silicon has a significantly high theoretical capacity of 4200 mAh/g. For this reason, silicon is preferably used as the negative electrode active material. Alternatively, a compound containing any of the above elements may be used. Examples of the compound include SiO, Mg_2Si, Mg_2Ge, SnO, SnO_2, Mg_2Sn, SnS_2, V_2Sn_3, $FeSn_2$, $CoSn_2$, Ni_3Sn_2, Cu_6Sn_5, Ag_3Sn, Ag_3Sb, Ni_2MnSb, $CeSb_3$, $LaSn_3$, $La_3Co_2Sn_7$, $CoSb_3$, InSb, and SbSn. Here, an element that enables charge-discharge reactions by an alloying reaction and a dealloying reaction with lithium, a compound containing the element, and the like may be referred to as an alloy-based material.

[0258]

In this specification and the like, SiO refers, for example, to silicon monoxide. SiO can alternatively be expressed as SiO_x. Here, x preferably has an approximate value of 1. For example, x is preferably 0.2 or more and 1.5 or less, more preferably 0.3 or more and 1.2 or less.

[0259]

As the carbon-based material, graphite, graphitizing carbon (soft carbon), non-graphitizing carbon (hard carbon), a carbon nanotube, graphene, carbon black, and the like can be used.

[0260]

Examples of graphite include artificial graphite and natural graphite. Examples of artificial graphite include meso-carbon microbeads (MCMB), coke-based artificial graphite, and pitch-based artificial graphite. As artificial graphite, spherical graphite having a spherical shape can be used. For example, MCMB is preferably used because it may have a spherical shape. Moreover, MCMB may preferably be used because it can relatively easily have a small surface area. Examples of natural graphite include flake graphite and spherical natural graphite.

[0261]

Graphite has a low potential substantially equal to that of a lithium metal (higher than or equal to 0.05 V and lower than or equal to 0.3 V vs. Li/Li^+) when lithium ions are intercalated into the graphite (while a lithium-graphite intercalation compound is formed). For this reason, a lithium-ion secondary battery can have a high operating voltage. In addition, graphite is preferred because of its advantages such as a relatively high capacity per unit volume, relatively small volume expansion, low cost, and higher level of safety than that of a lithium metal.

[0262]

Alternatively, for the negative electrode active material, an oxide such as titanium dioxide (TiO_2), lithium titanium oxide ($Li_4Ti_5O_{12}$), lithium-graphite intercalation compound (Li_xC_6), niobium pentoxide (Nb_2O_5), tungsten oxide (WO_2), or molybdenum oxide (MoO_2) can be used.

[0263]

Still alternatively, for the negative electrode active material, $Li_{3-x}M_xN$ (M = Co, Ni, or Cu) with a Li_3N structure, which is a nitride containing lithium and a transition metal, can be used. For example, $Li_{2.6}Co_{0.4}N_3$ is preferable because of high charge and discharge capacity (900 mAh/g and 1890 mAh/cm^3).

[0264]

A nitride containing lithium and a transition metal is preferably used, in which case lithium ions are contained in the negative electrode active material and thus the negative electrode active material can be used in combination with a material for a positive electrode active material which does not contain lithium ions, such as V_2O_5 or Cr_3O_8. In the case of using a material containing lithium ions as a positive electrode active material, the nitride containing lithium and a transition metal can be used for the negative electrode active material by extracting the lithium ions contained in the positive electrode active material in advance.

[0265]

Alternatively, a material which causes a conversion reaction can be used for the negative electrode active material; for example, a transition metal oxide which does not form an alloy with lithium, such as cobalt oxide (CoO), nickel oxide (NiO), and iron oxide (FeO), may be used. Other examples of the material which causes a conversion reaction include oxides such as Fe_2O_3, CuO, Cu_2O, RuO_2, and Cr_2O_3, sulfides such as $CoS_{0.89}$, NiS, and CuS, nitrides such as Zn_3N_2, Cu_3N, and Ge_3N_4, phosphides such as NiP_2, FeP_2, and CoP_3, and fluorides such as FeF_3 and BiF_3.

[0266]

For the conductive additive and the binder that can be included in the negative electrode active material layer, materials similar to those of the conductive additive and the binder that can be included in the positive electrode active material layer can be used.

[0267]

<Negative electrode current collector>

For the negative electrode current collector, a material similar to that of the positive electrode current collector can be used. Note that a material which is not alloyed with a carrier ion such as lithium is preferably used for the negative electrode current collector.

[0268]

[Electrolyte solution]

The electrolyte solution contains a solvent and an electrolyte. As a solvent of the

electrolyte solution, an aprotic organic solvent is preferably used. For example, one of ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate, chloroethylene carbonate, vinylene carbonate, γ-butyrolactone, γ-valerolactone, dimethyl carbonate (DMC), diethyl carbonate (DEC), ethyl methyl carbonate (EMC), methyl formate, methyl acetate, ethyl acetate, methyl propionate, ethyl propionate, propyl propionate, methyl butyrate, 1,3-dioxane, 1,4-dioxane, dimethoxyethane (DME), dimethyl sulfoxide, diethyl ether, methyl diglyme, acetonitrile, benzonitrile, tetrahydrofuran, sulfolane, and sultone can be used, or two or more of these solvents can be used in an appropriate combination in an appropriate ratio.

[0269]

Alternatively, when one or more kinds of ionic liquids (room temperature molten salts) which have features of non-flammability and non-volatility is used as a solvent of the electrolyte solution, a secondary battery can be prevented from exploding or catching fire even when the secondary battery internally shorts out or the internal temperature increases owing to overcharging or the like. An ionic liquid contains a cation and an anion. The ionic liquid contains an organic cation and an anion. Examples of the organic cation used for the electrolyte solution include aliphatic onium cations such as a quaternary ammonium cation, a tertiary sulfonium cation, and a quaternary phosphonium cation, and aromatic cations such as an imidazolium cation and a pyridinium cation. Examples of the anion used for the electrolyte solution include a monovalent amide-based anion, a monovalent methide-based anion, a fluorosulfonate anion, a perfluoroalkylsulfonate anion, a tetrafluoroborate anion, a perfluoroalkylborate anion, a hexafluorophosphate anion, and a perfluoroalkylphosphate anion.

[0270]

As an electrolyte dissolved in the above-described solvent, one of lithium salts such as $LiPF_6$, $LiClO_4$, $LiAsF_6$, $LiBF_4$, $LiAlCl_4$, LiSCN, LiBr, LiI, Li_2SO_4, $Li_2B_{10}Cl_{10}$, $Li_2B_{12}Cl_{12}$, $LiCF_3SO_3$, $LiC_4F_9SO_3$, $LiC(CF_3SO_2)_3$, $LiC(C_2F_5SO_2)_3$, $LiN(CF_3SO_2)_2$, $LiN(C_4F_9SO_2)(CF_3SO_2)$, and $LiN(C_2F_5SO_2)_2$ can be used, or two or more of these lithium salts can be used in an appropriate combination in an appropriate ratio.

[0271]

The electrolyte solution used for a secondary battery is preferably highly purified and contains a small amount of dust particles and elements other than the constituent elements of the electrolyte solution (hereinafter also simply referred to as impurities). Specifically, the weight ratio of impurities to the electrolyte solution is less than or equal to 1 %, preferably less than or equal to 0.1 %, and further preferably less than or equal to 0.01 %.

[0272]

Furthermore, an additive agent such as vinylene carbonate, propane sultone (PS), *tert*-

butylbenzene (TBB), fluoroethylene carbonate (FEC), LiBOB, or a dinitrile compound such as succinonitrile or adiponitrile may be added to the electrolyte solution. The concentration of a material to be added with respect to the whole solvent is, for example, higher than or equal to 0.1 wt% and lower than or equal to 5 wt%.

[0273]

Alternatively, a gelled electrolyte obtained in such a manner that a polymer is swelled with an electrolyte solution may be used.

[0274]

When a polymer gel electrolyte is used, safety against liquid leakage and the like is improved. Furthermore, a secondary battery can be thinner and more lightweight.

[0275]

As a polymer that undergoes gelation, a silicone gel, an acrylic gel, an acrylonitrile gel, a polyethylene oxide-based gel, a polypropylene oxide-based gel, a fluorine-based polymer gel, or the like can be used.

[0276]

Examples of the polymer include a polymer having a polyalkylene oxide structure, such as polyethylene oxide (PEO); PVDF; polyacrylonitrile; and a copolymer containing any of them. For example, PVDF-HFP, which is a copolymer of PVDF and hexafluoropropylene (HFP) can be used. The formed polymer may be porous.

[0277]

Instead of the electrolyte solution, a solid electrolyte including an inorganic material such as a sulfide-based inorganic material or an oxide-based inorganic material, or a solid electrolyte including a high-molecular material such as a polyethylene oxide (PEO)-based high-molecular material may alternatively be used. When the solid electrolyte is used, a separator and a spacer are not necessary. Furthermore, since the battery can be entirely solidified, there is no possibility of liquid leakage to increase the safety of the battery dramatically.

[0278]

[Separator]

The secondary battery preferably includes a separator. As the separator, for example, paper; nonwoven fabric; glass fiber; ceramics; or synthetic fiber using nylon (polyamide), vinylon (polyvinyl alcohol-based fiber), polyester, acrylic, polyolefin, or polyurethane can be used. The separator is preferably formed to have an envelope-like shape to wrap one of the positive electrode and the negative electrode.

[0279]

The separator may have a multilayer structure. For example, an organic material film

such as polypropylene or polyethylene can be coated with a ceramic-based material, a fluorine-based material, a polyamide-based material, a mixture thereof, or the like. Examples of the ceramic-based material include aluminum oxide particles and silicon oxide particles. Examples of the fluorine-based material include PVDF and a polytetrafluoroethylene. Examples of the polyamide-based material include nylon and aramid (meta-based aramid and para-based aramid).

[0280]

Deterioration of the separator in charging and discharging at high voltage can be suppressed and thus the reliability of the secondary battery can be improved because oxidation resistance is improved when the separator is coated with the ceramic-based material. In addition, when the separator is coated with the fluorine-based material, the separator is easily brought into close contact with an electrode, resulting in high output characteristics. When the separator is coated with the polyamide-based material, in particular, aramid, the safety of the secondary battery is improved because heat resistance is improved.

[0281]

For example, both surfaces of a polypropylene film may be coated with a mixed material of aluminum oxide and aramid. Alternatively, a surface of the polypropylene film in contact with the positive electrode may be coated with the mixed material of aluminum oxide and aramid, and a surface of the polypropylene film in contact with the negative electrode may be coated with the fluorine-based material.

[0282]

With the use of a separator having a multilayer structure, the capacity of the secondary battery per volume can be increased because the safety of the secondary battery can be maintained even when the total thickness of the separator is small.

[0283]

[Exterior body]

For an exterior body included in the secondary battery, a metal material such as aluminum and a resin material can be used, for example. An exterior body in the form of a film can also be used. As the film, for example, a film having a three-layer structure in which a highly flexible metal thin film of aluminum, stainless steel, copper, nickel, or the like is provided over a film formed of a material such as polyethylene, polypropylene, polycarbonate, ionomer, or polyamide, and an insulating synthetic resin film of a polyamide-based resin, a polyester-based resin, or the like is provided as the outer surface of the exterior body over the metal thin film can be used.

[0284]

[Charging and discharging methods]

The secondary battery can be charged and discharged in the following manner, for

example.

[0285]

<<CC charge>>

First, CC charge, which is one of charging methods, is described. CC charge is a charging method in which a constant current is made to flow to a secondary battery in the whole charging period and charge is terminated when the voltage reaches a predetermined voltage. The secondary battery is assumed to be an equivalent circuit with internal resistance R and secondary battery capacitance C as illustrated in FIG. 8A. In that case, a secondary battery voltage V_B is the sum of a voltage V_R applied to the internal resistance R and a voltage V_C applied to the secondary battery capacitance C.

[0286]

While the CC charge is performed, a switch is on as illustrated in FIG. 8A, so that a constant current I flows to the secondary battery. During the period, the current I is constant; thus, according to the Ohm's law ($V_R = R \times I$), the voltage V_R applied to the internal resistance R is also constant. In contrast, the voltage V_C applied to the secondary battery capacitance C increases over time. Accordingly, the secondary battery voltage V_B increases over time.

[0287]

When the secondary battery voltage V_B reaches a predetermined voltage, e.g., 4.3 V, the charge is terminated. On termination of the CC charge, the switch is turned off as illustrated in FIG. 8B, and the current I becomes 0. Thus, the voltage V_R applied to the internal resistance R becomes 0 V. Consequently, the secondary battery voltage V_B is decreased by the lost voltage drop in the internal resistance R.

[0288]

FIG. 8C shows an example of the secondary battery voltage V_B and charging current during a period in which the CC charge is performed and after the CC charge is terminated. The secondary battery voltage V_B increases while the CC charge is performed, and slightly decreases after the CC charge is terminated.

[0289]

<<CCCV charge>>

Next, CCCV charge, which is a charging method different from the above-described method, is described. CCCV charge is a charging method in which CC charge is performed until the voltage reaches a predetermined voltage and then constant voltage (CV) charge is performed until the amount of current flow becomes small, specifically, a termination current value.

[0290]

While the CC charge is performed, a switch of a constant current power source is on and

a switch of a constant voltage power source is off as illustrated in FIG. 9A, so that the constant current I flows to the secondary battery. During the period, the current I is constant; thus, according to the Ohm's law ($V_R = R \times I$), the voltage V_R applied to the internal resistance R is also constant. In contrast, the voltage V_C applied to the secondary battery capacitance C increases over time. Accordingly, the secondary battery voltage V_B increases over time.

[0291]

When the secondary battery voltage V_B reaches a predetermined voltage, e.g., 4.3 V, switching is performed from the CC charge to the CV charge. While the CV charge is performed, the switch of the constant voltage power source is on and the switch of the constant current power source is off as illustrated in FIG. 9B; thus, the secondary battery voltage V_B is constant. In contrast, the voltage V_C applied to the secondary battery capacitance C increases over time. Since $V_B = V_R + V_C$ is satisfied, the voltage V_R applied to the internal resistance R decreases over time. As the voltage V_R applied to the internal resistance R decreases, the current I flowing to the secondary battery also decreases according to the Ohm's law ($V_R = R \times I$).

[0292]

When the current I flowing to the secondary battery becomes a predetermined current, e.g., approximately 0.01 C, charge is terminated. On termination of the CCCV charge, all the switches are turned off as illustrated in FIG. 9C, so that the current I becomes 0. Thus, the voltage V_R applied to the internal resistance R becomes 0 V. However, the voltage V_R applied to the internal resistance R becomes sufficiently small by the CV charge; thus, even when a voltage drop no longer occurs in the internal resistance R, the secondary battery voltage V_B hardly decreases.

[0293]

FIG. 9D shows an example of the secondary battery voltage V_B and charging current while the CCCV charge is performed and after the CCCV charge is terminated. Even after the CCCV charge is terminated, the secondary battery voltage V_B hardly decreases.

[0294]

<<CC discharge>>

Next, CC discharge, which is one of discharging methods, is described. CC discharge is a discharging method in which a constant current is made to flow from the secondary battery in the whole discharging period, and discharge is terminated when the secondary battery voltage V_B reaches a predetermined voltage, e.g., 2.5 V.

[0295]

FIG. 10 shows an example of the secondary battery voltage V_B and discharging current while the CC discharge is performed. As discharge proceeds, the secondary battery voltage V_B decreases.

[0296]

Next, a discharge rate and a charge rate will be described. The discharge rate refers to the relative ratio of discharging current to battery capacity and is expressed in a unit C. A current of approximately 1 C in a battery with a rated capacity X (Ah) is X A. The case where discharge is performed at a current of $2X$ A is rephrased as follows: discharge is performed at 2 C. The case where discharge is performed at a current of $X/5$ A is rephrased as follows: discharge is performed at 0.2 C. Similarly, the case where charging is performed at a current of $2X$ A is rephrased as follows: charging is performed at 2 C, and the case where charging is performed at a current of $X/5$ A is rephrased as follows: charging is performed at 0.2 C.

[0297]

(Embodiment 3)

In this embodiment, examples of a shape of a secondary battery containing the positive electrode active material 100 described in the above embodiment are described. For the materials used for the secondary battery described in this embodiment, the description of the above embodiment can be referred to.

[0298]

[Coin-type secondary battery]

First, an example of a coin-type secondary battery is described. FIG. 11A is an external view of a coin-type (single-layer flat type) secondary battery, and FIG. 11B is a cross-sectional view thereof.

[0299]

In a coin-type secondary battery 300, a positive electrode can 301 doubling as a positive electrode terminal and a negative electrode can 302 doubling as a negative electrode terminal are insulated from each other and sealed by a gasket 303 made of polypropylene or the like. A positive electrode 304 includes a positive electrode current collector 305 and a positive electrode active material layer 306 provided in contact with the positive electrode current collector 305. A negative electrode 307 includes a negative electrode current collector 308 and a negative electrode active material layer 309 provided in contact with the negative electrode current collector 308.

[0300]

Note that only one surface of each of the positive electrode 304 and the negative electrode 307 used for the coin-type secondary battery 300 is provided with an active material layer.

[0301]

For the positive electrode can 301 and the negative electrode can 302, a metal having a corrosion-resistant property to an electrolyte solution, such as nickel, aluminum, or titanium, an alloy of such a metal, or an alloy of such a metal and another metal (e.g., stainless steel) can be

used. Alternatively, the positive electrode can 301 and the negative electrode can 302 are preferably covered with nickel, aluminum, or the like in order to prevent corrosion due to the electrolyte solution. The positive electrode can 301 and the negative electrode can 302 are electrically connected to the positive electrode 304 and the negative electrode 307, respectively.

[0302]

The negative electrode 307, the positive electrode 304, and the separator 310 are immersed in the electrolyte solution. Then, as illustrated in FIG. 11B, the positive electrode 304, the separator 310, the negative electrode 307, and the negative electrode can 302 are stacked in this order with the positive electrode can 301 positioned at the bottom, and the positive electrode can 301 and the negative electrode can 302 are subjected to pressure bonding with the gasket 303 located therebetween. In such a manner, the coin-type secondary battery 300 can be manufactured.

[0303]

When the positive electrode active material described in the above embodiment is used in the positive electrode 304, the coin-type secondary battery 300 with high capacity and excellent cycle characteristics can be obtained.

[0304]

Here, a current flow in charging a secondary battery is described with reference to FIG. 11C. When a secondary battery using lithium is regarded as a closed circuit, lithium ions transfer and a current flows in the same direction. Note that in the secondary battery using lithium, an anode and a cathode change places in charge and discharge, and an oxidation reaction and a reduction reaction occur on the corresponding sides; hence, an electrode with a high reaction potential is called a positive electrode and an electrode with a low reaction potential is called a negative electrode. For this reason, in this specification, the positive electrode is referred to as a "positive electrode" or a "plus electrode" and the negative electrode is referred to as a "negative electrode" or a "minus electrode" in all the cases where charge is performed, discharge is performed, a reverse pulse current is supplied, and a charging current is supplied. The use of the terms "anode" and "cathode" related to an oxidation reaction and a reduction reaction might cause confusion because the anode and the cathode change places at the time of charging and discharging. Thus, the terms "anode" and "cathode" are not used in this specification. If the term "anode" or "cathode" is used, it should be mentioned that the anode or the cathode is which of the one at the time of charging or the one at the time of discharging and corresponds to which of a positive (plus) electrode or a negative (minus) electrode.

[0305]

Two terminals in FIG. 11C are connected to a charger, and the coin-type secondary battery

300 is charged. As the charge of the coin-type secondary battery 300 proceeds, a potential difference between electrodes increases.

[0306]

[Cylindrical secondary battery]

Next, an example of a cylindrical secondary battery will be described with reference to FIGS. 12A to 12D. A cylindrical secondary battery 600 includes, as illustrated in FIG. 12A, a positive electrode cap (battery lid) 601 on the top surface and a battery can (outer can) 602 on the side and bottom surfaces. The positive electrode cap and the battery can (outer can) 602 are insulated from each other by a gasket (insulating packing) 610.

[0307]

FIG. 12B is a schematic cross-sectional view of the cylindrical secondary battery. Inside the battery can 602 having a hollow cylindrical shape, a battery element in which a strip-like positive electrode 604 and a strip-like negative electrode 606 are wound with a strip-like separator 605 located therebetween is provided. Although not illustrated, the battery element is wound around a center pin. One end of the battery can 602 is close and the other end thereof is open. For the battery can 602, a metal having a corrosion-resistant property to an electrolyte solution, such as nickel, aluminum, or titanium, an alloy of such a metal, or an alloy of such a metal and another metal (e.g., stainless steel) can be used. Alternatively, the battery can 602 is preferably covered with nickel, aluminum, or the like in order to prevent corrosion due to the electrolyte solution. Inside the battery can 602, the battery element in which the positive electrode, the negative electrode, and the separator are wound is provided between a pair of insulating plates 608 and 609 that face each other. Furthermore, a nonaqueous electrolyte solution (not illustrated) is injected inside the battery can 602 provided with the battery element. As the nonaqueous electrolyte solution, a nonaqueous electrolyte solution that is similar to that of the coin-type secondary battery can be used.

[0308]

Since the positive electrode and the negative electrode of the cylindrical secondary battery are wound, active materials are preferably formed on both sides of the current collectors. A positive electrode terminal (positive electrode current collecting lead) 603 is connected to the positive electrode 604, and a negative electrode terminal (negative electrode current collecting lead) 607 is connected to the negative electrode 606. Both the positive electrode terminal 603 and the negative electrode terminal 607 can be formed using a metal material such as aluminum. The positive electrode terminal 603 and the negative electrode terminal 607 are resistance-welded to a safety valve mechanism 612 and the bottom of the battery can 602, respectively. The safety valve mechanism 612 is electrically connected to the positive electrode cap 601 through a positive

temperature coefficient (PTC) element 611. The safety valve mechanism 612 cuts off electrical connection between the positive electrode cap 601 and the positive electrode 604 when the internal pressure of the battery exceeds a predetermined threshold value. The PTC element 611, which serves as a thermally sensitive resistor whose resistance increases as temperature rises, limits the amount of current by increasing the resistance, in order to prevent abnormal heat generation. Barium titanate ($BaTiO_3$)-based semiconductor ceramic can be used for the PTC element.

[0309]

Alternatively, as illustrated in FIG. 12C, a plurality of cylindrical secondary batteries 600 may be sandwiched between a conductive plate 613 and a conductive plate 614 to form a module 615. The plurality of cylindrical secondary batteries 600 may be connected in parallel, connected in series, or connected in series after being connected in parallel. With the module 615 including the plurality of cylindrical secondary batteries 600, large electric power can be extracted.

[0310]

FIG. 12D is a top view of the module 615. The conductive plate 613 is shown by a dotted line for clarity of the drawing. As illustrated in FIG. 12D, the module 615 may include a wiring 616 which electrically connects the plurality of cylindrical secondary batteries 600 to each other. It is possible to provide the conductive plate 613 over the wiring 616 to overlap with each other. In addition, a temperature control device 617 may be provided between the plurality of cylindrical secondary batteries 600. When the cylindrical secondary batteries 600 are overheated, the temperature control device 617 can cool them, and when the cylindrical secondary batteries 600 are cooled too much, the temperature control device 617 can heat them. Thus, the performance of the module 615 is not easily influenced by the outside air temperature.

[0311]

When the positive electrode active material described in the above embodiment is used in the positive electrode 604, the cylindrical secondary battery 600 with high capacity and excellent cycle characteristics can be obtained.

[0312]

[Structural examples of secondary battery]

Other structural examples of secondary batteries will be described with reference to FIGS. 13A and 13B, FIGS. 14A-1, 14A-2, 14B-1, and 14B-2, FIGS. 15A and 15B, and FIG. 16.

[0313]

FIGS. 13A and 13B are external views of a secondary battery. The secondary battery includes a circuit board 900 and a secondary battery 913. A label 910 is attached to the secondary battery 913. As shown in FIG. 13B, the secondary battery further includes a terminal 951, a terminal 952, an antenna 914, and an antenna 915.

[0314]

The circuit board 900 includes terminals 911 and a circuit 912. The terminals 911 are connected to the terminals 951 and 952, the antennas 914 and 915, and the circuit 912. Note that a plurality of terminals 911 serving as a control signal input terminal, a power supply terminal, and the like may be provided.

[0315]

The circuit 912 may be provided on the rear surface of the circuit board 900. The shape of each of the antennas 914 and 915 is not limited to a coil shape and may be a linear shape or a plate shape. Furthermore, a planar antenna, an aperture antenna, a traveling-wave antenna, an EH antenna, a magnetic-field antenna, a dielectric antenna, or the like may be used. Alternatively, the antenna 914 or the antenna 915 may be a flat-plate conductor. The flat-plate conductor can serve as one of conductors for electric field coupling. That is, the antenna 914 or the antenna 915 can serve as one of two conductors of a capacitor. Thus, electric power can be transmitted and received not only by an electromagnetic field or a magnetic field but also by an electric field.

[0316]

The line width of the antenna 914 is preferably larger than that of the antenna 915. This makes it possible to increase the amount of electric power received by the antenna 914.

[0317]

The secondary battery includes a layer 916 between the secondary battery 913 and the antennas 914 and 915. The layer 916 has a function of blocking an electromagnetic field from the secondary battery 913, for example. As the layer 916, for example, a magnetic body can be used.

[0318]

Note that the structure of the secondary battery is not limited to that shown in FIGS. 13A and 13B.

[0319]

For example, as shown in FIGS. 14A-1 and 14A-2, two opposite surfaces of the secondary battery 913 in FIGS. 13A and 13B may be provided with respective antennas. FIG. 14A-1 is an external view showing one side of the opposite surfaces, and FIG. 14A-2 is an external view showing the other side of the opposite surfaces. For portions similar to those in FIGS. 13A and 13B, a description of the secondary battery illustrated in FIGS. 13A and 13B can be referred to as appropriate.

[0320]

As illustrated in FIG. 14A-1, the antenna 914 is provided on one of the opposite surfaces of the secondary battery 913 with the layer 916 located therebetween, and as illustrated in FIG.

14A-2, an antenna 918 is provided on the other of the opposite surfaces of the secondary battery 913 with a layer 917 located therebetween. The layer 917 has a function of blocking an electromagnetic field from the secondary battery 913, for example. As the layer 917, for example, a magnetic body can be used.

[0321]

With the above structure, both of the antennas 914 and 918 can be increased in size. The antenna 918 has a function of communicating data with an external device, for example. An antenna with a shape that can be applied to the antenna 914, for example, can be used as the antenna 918. As a system for communication using the antenna 918 between the secondary battery and another device, a response method that can be used between the secondary battery and another device, such as NFC, can be employed.

[0322]

Alternatively, as illustrated in FIG. 14B-1, the secondary battery 913 in FIGS. 13A and 13B may be provided with a display device 920. The display device 920 is electrically connected to the terminal 911. It is possible that the label 910 is not provided in a portion where the display device 920 is provided. For portions similar to those in FIGS. 13A and 13B, a description of the secondary battery illustrated in FIGS. 13A and 13B can be referred to as appropriate.

[0323]

The display device 920 can display, for example, an image showing whether charge is being carried out, an image showing the amount of stored power, or the like. As the display device 920, electronic paper, a liquid crystal display device, an electroluminescent (EL) display device, or the like can be used. For example, the use of electronic paper can reduce power consumption of the display device 920.

[0324]

Alternatively, as illustrated in FIG. 14B-2, the secondary battery 913 illustrated in FIGS. 13A and 13B may be provided with a sensor 921. The sensor 921 is electrically connected to the terminal 911 via a terminal 922. For portions similar to those in FIGS. 13A and 13B, a description of the secondary battery illustrated in FIGS. 13A and 13B can be referred to as appropriate.

[0325]

The sensor 921 has a function of measuring, for example, displacement, position, speed, acceleration, angular velocity, rotational frequency, distance, light, liquid, magnetism, temperature, chemical substance, sound, time, hardness, electric field, electric current, voltage, electric power, radiation, flow rate, humidity, gradient, oscillation, odor, or infrared rays. With the sensor 921, for example, data on an environment (e.g., temperature) where the secondary battery is placed can

be determined and stored in a memory inside the circuit 912.

[0326]

Furthermore, structural examples of the secondary battery 913 will be described with reference to FIGS. 15A and 15B and FIG. 16.

[0327]

The secondary battery 913 illustrated in FIG. 15A includes a wound body 950 provided with the terminals 951 and 952 inside a housing 930. The wound body 950 is soaked in an electrolyte solution inside the housing 930. The terminal 952 is in contact with the housing 930. An insulator or the like inhibits contact between the terminal 951 and the housing 930. Note that in FIG. 15A, the housing 930 divided into two pieces is illustrated for convenience; however, in the actual structure, the wound body 950 is covered with the housing 930 and the terminals 951 and 952 extend to the outside of the housing 930. For the housing 930, a metal material (such as aluminum) or a resin material can be used.

[0328]

Note that as illustrated in FIG. 15B, the housing 930 in FIG. 15A may be formed using a plurality of materials. For example, in the secondary battery 913 in FIG. 15B, a housing 930a and a housing 930b are bonded to each other, and the wound body 950 is provided in a region surrounded by the housing 930a and the housing 930b.

[0329]

For the housing 930a, an insulating material such as an organic resin can be used. In particular, when a material such as an organic resin is used for the side on which an antenna is formed, blocking of an electric field from the secondary battery 913 can be inhibited. When an electric field is not significantly blocked by the housing 930a, an antenna such as the antennas 914 and 915 may be provided inside the housing 930a. For the housing 930b, a metal material can be used, for example.

[0330]

FIG. 16 illustrates the structure of the wound body 950. The wound body 950 includes a negative electrode 931, a positive electrode 932, and separators 933. The wound body 950 is obtained by winding a sheet of a stack in which the negative electrode 931 overlaps with the positive electrode 932 with the separator 933 provided therebetween. Note that a plurality of stacks each including the negative electrode 931, the positive electrode 932, and the separator 933 may be stacked.

[0331]

The negative electrode 931 is connected to the terminal 911 in FIGS. 13A and 13B via one of the terminals 951 and 952. The positive electrode 932 is connected to the terminal 911 in

FIGS. 13A and 13B via the other of the terminals 951 and 952.

[0332]

When the positive electrode active material described in the above embodiment is used in the positive electrode 932, the secondary battery 913 with high capacity and excellent cycle characteristics can be obtained.

[0333]

[Laminated secondary battery]

Next, an example of a laminated secondary battery will be described with reference to FIGS. 17A to 17C, FIG. 18A and 18B, FIG. 19, FIG. 20, FIGS. 21A to 21C, FIGS. 22A, 22B1, 22B2, 22C, and 22D, and FIGS. 23A and 23B. When the laminated secondary battery has flexibility and is used in an electronic device at least part of which is flexible, the secondary battery can be bent as the electronic device is bent.

[0334]

A laminated secondary battery 980 is described with reference to FIGS. 17A to 17C. The laminated secondary battery 980 includes a wound body 993 illustrated in FIG. 17A. The wound body 993 includes a negative electrode 994, a positive electrode 995, and a separator 996. The wound body 993 is, like the wound body 950 illustrated in FIG. 16, obtained by winding a sheet of a stack in which the negative electrode 994 overlaps with the positive electrode 995 with the separator 996 therebetween.

[0335]

Note that the number of stacks each including the negative electrode 994, the positive electrode 995, and the separator 996 may be determined as appropriate depending on capacity and an element volume which are required. The negative electrode 994 is connected to a negative electrode current collector (not illustrated) via one of a lead electrode 997 and a lead electrode 998. The positive electrode 995 is connected to a positive electrode current collector (not illustrated) via the other of the lead electrode 997 and the lead electrode 998.

[0336]

As illustrated in FIG. 17B, the wound body 993 is packed in a space formed by bonding a film 981 and a film 982 having a depressed portion that serve as exterior bodies by thermocompression bonding or the like, whereby the laminated secondary battery 980 can be formed as illustrated in FIG. 17C. The wound body 993 includes the lead electrode 997 and the lead electrode 998, and is soaked in an electrolyte solution inside a space surrounded by the film 981 and the film 982 having a depressed portion.

[0337]

For the film 981 and the film 982 having a depressed portion, a metal material such as

aluminum or a resin material can be used, for example. With the use of a resin material for the film 981 and the film 982 having a depressed portion, the film 981 and the film 982 having a depressed portion can be changed in their forms when external force is applied; thus, a flexible secondary battery can be fabricated.

[0338]

Although FIGS. 17B and 17C illustrate an example where a space is formed by two films, the wound body 993 may be placed in a space formed by bending one film.

[0339]

When the positive electrode active material described in the above embodiment is used in the positive electrode 995, the laminated secondary battery 980 with high capacity and excellent cycle characteristics can be obtained.

[0340]

In FIGS. 17A to 17C, an example in which the laminated secondary battery 980 includes a wound body in a space formed by films serving as exterior bodies is described; however, as illustrated in FIGS. 18A and 18B, a secondary battery may include a plurality of strip-shaped positive electrodes, a plurality of strip-shaped separators, and a plurality of strip-shaped negative electrodes in a space formed by films serving as exterior bodies, for example.

[0341]

A laminated secondary battery 500 illustrated in FIG. 18A includes a positive electrode 503 including a positive electrode current collector 501 and a positive electrode active material layer 502, a negative electrode 506 including a negative electrode current collector 504 and a negative electrode active material layer 505, a separator 507, an electrolyte solution 508, and an exterior body 509. The separator 507 is provided between the positive electrode 503 and the negative electrode 506 in the exterior body 509. The exterior body 509 is filled with the electrolyte solution 508. The electrolyte solution described in Embodiment 2 can be used for the electrolyte solution 508.

[0342]

In the laminated secondary battery 500 illustrated in FIG. 18A, the positive electrode current collector 501 and the negative electrode current collector 504 also serve as terminals for an electrical contact with an external portion. For this reason, the positive electrode current collector 501 and the negative electrode current collector 504 may be arranged so as to be partly exposed to the outside of the exterior body 509. Alternatively, a lead electrode and the positive electrode current collector 501 or the negative electrode current collector 504 may be bonded to each other by ultrasonic welding, and instead of the positive electrode current collector 501 and the negative electrode current collector 504, the lead electrode may be exposed to the outside of

the exterior body 509.

[0343]

As the exterior body 509 of the laminated secondary battery 500, for example, a laminate film having a three-layer structure can be employed in which a highly flexible metal thin film of aluminum, stainless steel, copper, nickel, or the like is provided over a film formed of a material such as polyethylene, polypropylene, polycarbonate, ionomer, or polyamide, and an insulating synthetic resin film of a polyamide-based resin, a polyester-based resin, or the like is provided over the metal thin film as the outer surface of the exterior body.

[0344]

FIG. 18B illustrates an example of a cross-sectional structure of the laminated secondary battery 500. Although FIG. 18A illustrates an example including only two current collectors for simplicity, an actual battery includes a plurality of electrode layers.

[0345]

The example in FIG. 18B includes 16 electrode layers. The laminated secondary battery 500 has flexibility even though including 16 electrode layers. FIG. 18B illustrates a structure including 8 layers of negative electrode current collectors 504 and 8 layers of positive electrode current collectors 501, i.e., 16 layers in total. Note that FIG. 18B illustrates a cross section of the lead portion of the negative electrode, and the 8 negative electrode current collectors 504 are bonded to each other by ultrasonic welding. It is needless to say that the number of electrode layers is not limited to 16, and may be more than 16 or less than 16. With a large number of electrode layers, the secondary battery can have high capacity. In contrast, with a small number of electrode layers, the secondary battery can have small thickness and high flexibility.

[0346]

FIGS. 19 and 20 each illustrate an example of the external view of the laminated secondary battery 500. In FIGS. 19 and 20, the positive electrode 503, the negative electrode 506, the separator 507, the exterior body 509, a positive electrode lead electrode 510, and a negative electrode lead electrode 511 are included.

[0347]

FIG. 21A illustrates external views of the positive electrode 503 and the negative electrode 506. The positive electrode 503 includes the positive electrode current collector 501, and the positive electrode active material layer 502 is formed on a surface of the positive electrode current collector 501. The positive electrode 503 also includes a region where the positive electrode current collector 501 is partly exposed (hereinafter referred to as a tab region). The negative electrode 506 includes the negative electrode current collector 504, and the negative electrode active material layer 505 is formed on a surface of the negative electrode current collector

504. The negative electrode 506 also includes a region where the negative electrode current collector 504 is partly exposed, that is, a tab region. The areas and the shapes of the tab regions included in the positive electrode and the negative electrode are not limited to those illustrated in FIG. 21A.

[0348]

[Method for manufacturing laminated secondary battery]

Here, an example of a method for manufacturing the laminated secondary battery whose external view is illustrated in FIG. 19 will be described with reference to FIGS. 21B and 21C.

[0349]

First, the negative electrode 506, the separator 507, and the positive electrode 503 are stacked. FIG. 21B illustrates a stack including the negative electrode 506, the separator 507, and the positive electrode 503. The secondary battery described here as an example includes 5 negative electrodes and 4 positive electrodes. Next, the tab regions of the positive electrodes 503 are bonded to each other, and the tab region of the positive electrode on the outermost surface and the positive electrode lead electrode 510 are bonded to each other. The bonding can be performed by ultrasonic welding, for example. In a similar manner, the tab regions of the negative electrodes 506 are bonded to each other, and the negative electrode lead electrode 511 is bonded to the tab region of the negative electrode on the outermost surface.

[0350]

After that, the negative electrode 506, the separator 507, and the positive electrode 503 are placed over the exterior body 509.

[0351]

Subsequently, the exterior body 509 is folded along a dashed line as illustrated in FIG. 21C. Then, the outer edge of the exterior body 509 is bonded. The bonding can be performed by thermocompression bonding, for example. At this time, a part (or one side) of the exterior body 509 is left unbonded (to provide an inlet) so that the electrolyte solution 508 can be introduced later.

[0352]

Next, the electrolyte solution 508 is introduced into the exterior body 509 from the inlet of the exterior body 509. The electrolyte solution 508 is preferably introduced in a reduced pressure atmosphere or in an inert gas atmosphere. Lastly, the inlet is bonded. In the above manner, the laminated secondary battery 500 can be manufactured.

[0353]

When the positive electrode active material described in the above embodiment is used in the positive electrode 503, the laminated secondary battery 500 with high capacity and excellent

cycle characteristics can be obtained.

[0354]

[Bendable secondary battery]

Next, an example of a bendable secondary battery is described with reference to FIGS. 22A, 22B1, 22B2, 22C and 22D and FIGS. 23A and 23B.

[0355]

FIG. 22A is a schematic top view of a bendable secondary battery 50. FIGS. 22B1, 22B2, and 22C are schematic cross-sectional views taken along cutting line C1-C2, cutting line C3-C4, and cutting line A1-A2, respectively, in FIG. 22A. The battery 50 includes an exterior body 51 and a positive electrode 11a, and a negative electrode 11b held in the exterior body 51. A lead 12a electrically connected to the positive electrode 11a and a lead 12b electrically connected to the negative electrode 11b are extended to the outside of the exterior body 51. In addition to the positive electrode 11a and the negative electrode 11b, an electrolyte solution (not illustrated) is enclosed in a region surrounded by the exterior body 51.

[0356]

FIGS. 23A and 23B illustrate the positive electrode 11a and the negative electrode 11b included in the battery 50. FIG. 23A is a perspective view illustrating the stacking order of the positive electrode 11a, the negative electrode 11b, and the separator 14. FIG. 23B is a perspective view illustrating the lead 12a and the lead 12b in addition to the positive electrode 11a and the negative electrode 11b.

[0357]

As illustrated in FIG. 23A, the battery 50 includes a plurality of strip-shaped positive electrodes 11a, a plurality of strip-shaped negative electrodes 11b, and a plurality of separators 14. The positive electrode 11a and the negative electrode 11b each include a projected tab portion and a portion other than the tab. A positive electrode active material layer is formed on one surface of the positive electrode 11a other than the tab portion, and a negative electrode active material layer is formed on one surface of the negative electrode 11b other than the tab portion.

[0358]

The positive electrodes 11a and the negative electrodes 11b are stacked so that surfaces of the positive electrodes 11a on each of which the positive electrode active material layer is not formed are in contact with each other and that surfaces of the negative electrodes 11b on each of which the negative electrode active material layer is not formed are in contact with each other.

[0359]

Furthermore, the separator 14 is provided between the surface of the positive electrode 11a on which the positive electrode active material is formed and the surface of the negative

electrode 11b on which the negative electrode active material is formed. In FIG. 23A, the separator 14 is shown by a dotted line for easy viewing.

[0360]

In addition, as illustrated in FIG. 23B, the plurality of positive electrodes 11a are electrically connected to the lead 12a in a bonding portion 15a. The plurality of negative electrodes 11b are electrically connected to the lead 12b in a bonding portion 15b.

[0361]

Next, the exterior body 51 is described with reference to FIGS. 22B1, 22B2, 22C, and 22D.

[0362]

The exterior body 51 has a film-like shape and is folded in half with the positive electrodes 11a and the negative electrodes 11b between facing portions of the exterior body 51. The exterior body 51 includes a folded portion 61, a pair of seal portions 62, and a seal portion 63. The pair of seal portions 62 is provided with the positive electrodes 11a and the negative electrodes 11b positioned therebetween and thus can also be referred to as side seals. The seal portion 63 has portions overlapping with the lead 12a and the lead 12b and can also be referred to as a top seal.

[0363]

Part of the exterior body 51 that overlaps with the positive electrodes 11a and the negative electrodes 11b preferably has a wave shape in which crest lines 71 and trough lines 72 are alternately arranged. The seal portions 62 and the seal portion 63 of the exterior body 51 are preferably flat.

[0364]

FIG. 22B1 shows a cross section cut along the part overlapping with the crest line 71. FIG. 22B2 shows a cross section cut along the part overlapping with the trough line 72. FIGS. 22B1 and 22B2 correspond to cross sections of the battery 50, the positive electrodes 11a, and the negative electrodes 11b in the width direction.

[0365]

The distance between an end portion of the negative electrode 11b in the width direction and the seal portion 62 is referred to as a distance La. When the battery 50 changes in shape, for example, is bent, the positive electrode 11a and the negative electrode 11b change in shape such that the positions thereof are shifted from each other in the length direction as described later. At the time, if the distance La is too short, the exterior body 51 and the positive electrode 11a and the negative electrode 11b are rubbed hard against each other, so that the exterior body 51 is damaged in some cases. In particular, when a metal film of the exterior body 51 is exposed, there is concern that the metal film is corroded by the electrolyte solution. Thus, the distance La is

preferably set as long as possible. However, if the distance La is too long, the volume of the battery 50 is increased.

[0366]

The distance La between the end portion of the negative electrode 11b and the seal portion 62 is preferably increased as the total thickness of the stacked positive electrodes 11a and negative electrodes 11b is increased.

[0367]

Specifically, when the total thickness of the stacked positive electrodes 11a and negative electrodes 11b and the separators 214 ~~(not illustrated)~~ is referred to as a thickness t, the distance La is preferably 0.8 times or more and 3.0 times or less, further preferably 0.9 times or more and 2.5 times or less, still further preferably 1.0 times or more and 2.0 times or less as large as the thickness t. When the distance La is in the above-described range, a compact battery which is highly reliable for bending can be obtained.

[0368]

Furthermore, when a distance between the pair of seal portions 62 is referred to as a distance Lb, it is preferable that the distance Lb be sufficiently longer than a width Wb of the negative electrode 11b. In this case, even when the positive electrode 11a and the negative electrode 11b come into contact with the exterior body 51 by change in the shape of the battery 50 such as repeated bending, the position of part of the positive electrode 11a and the negative electrode 11b can be shifted in the width direction; thus, the positive and negative electrodes 11a and 11b and the exterior body 51 can be effectively prevented from being rubbed against each other.

[0369]

For example, the difference between the distance Lb (i.e., the distance between the pair of seal portions 62) and the width Wb of the negative electrode 11b is preferably 1.6 times or more and 6.0 times or less, further preferably 1.8 times or more and 5.0 times or less, still further preferably 2.0 times or more and 4.0 times or less as large as the total thickness t of the positive electrode 11a and the negative electrode 11b.

[0370]

In other words, the distance Lb, the width Wb, and the thickness t preferably satisfy the relation of the following Formula 2.

[0371]

[Formula 2]

$$\frac{Lb - Wb}{2t} \geq a \qquad \text{(Formula 2)}$$

[0372]

In the formula, a is 0.8 or more and 3.0 or less, preferably 0.9 or more and 2.5 or less, further preferably 1.0 or more and 2.0 or less.

[0373]

FIG. 22C illustrates a cross section including the lead 12a and corresponds to a cross section of the battery 50, the positive electrode 11a, and the negative electrode 11b in the length direction. As illustrated in FIG. 22C, a space 73 is preferably provided between end portions of the positive electrode 11a and the negative electrode 11b in the length direction and the exterior body 51 in the folded portion 61.

[0374]

FIG. 22D is a schematic cross-sectional view of the battery 50 in a state of being bent. FIG. 22D corresponds to a cross section along cutting line B1-B2 in FIG. 22A.

[0375]

When the battery 50 is bent, a part of the exterior body 51 positioned on the outer side in bending is unbent and the other part positioned on the inner side changes its shape as it shrinks. More specifically, the part of the exterior body 51 positioned on the outer side in bending changes its shape such that the wave amplitude becomes smaller and the length of the wave period becomes larger. In contrast, the part of the exterior body 51 positioned on the inner side in bending changes its shape such that the wave amplitude becomes larger and the length of the wave period becomes smaller. When the exterior body 51 changes its shape in this manner, stress applied to the exterior body 51 due to bending is relieved, so that a material itself that forms the exterior body 51 does not need to expand and contract. As a result, the battery 50 can be bent with weak force without damage to the exterior body 51.

[0376]

Furthermore, as illustrated in FIG. 22D, when the battery 50 is bent, the positions of the positive electrode 11a and the negative electrode 11b are shifted relatively. At this time, ends of the stacked positive electrodes 11a and negative electrodes 11b on the seal portion 63 side are fixed by the fixing member 17. Thus, the plurality of positive electrodes 11a and the plurality of negative electrodes 11b are more shifted at a position closer to the folded portion 61. Therefore, stress applied to the positive electrode 11a and the negative electrode 11b is relieved, and the positive electrode 11a and the negative electrode 11b themselves do not need to expand and contract. As a result, the battery 50 can be bent without damage to the positive electrode 11a and the negative electrode 11b.

[0377]

Furthermore, the space 73 is provided between the end portions of the positive and negative electrodes 11a and 11b and the exterior body 51, whereby the relative positions of the positive electrode 11a and the negative electrode 11b can be shifted while the end portions of the positive electrode 11a and the negative electrode 11b located on an inner side when the battery 50 is bent do not contact the exterior body 51.

[0378]

In the battery 50 illustrated in FIGS. 22A, 22B1, 22B2, 22C and 22D and FIGS. 23A and 23B, the exterior body, the positive electrode 11a, and the negative electrode 11b are less likely to be damaged and the battery characteristics are less likely to deteriorate even when the battery 50 is repeatedly bent and unbent. When the positive electrode active material described in the above embodiment is used for the positive electrode 11a included in the battery 50, a battery with more excellent cycle characteristics can be obtained.

[0379]

(Embodiment 4)

In this embodiment, examples of electronic devices including the secondary battery of one embodiment of the present invention are described.

[0380]

First, FIGS. 24A to 24G show examples of electronic devices including the bendable secondary battery described in Embodiment 3. Examples of an electronic device including a flexible secondary battery include television sets (also referred to as televisions or television receivers), monitors of computers or the like, digital cameras or digital video cameras, digital photo frames, mobile phones (also referred to as cellular phones or mobile phone devices), portable game machines, portable information terminals, audio reproducing devices, and large game machines such as pachinko machines.

[0381]

In addition, a flexible secondary battery can be incorporated along a curved inside/outside wall surface of a house or a building or a curved interior/exterior surface of an automobile.

[0382]

FIG. 24A illustrates an example of a mobile phone. A mobile phone 7400 is provided with a display portion 7402 incorporated in a housing 7401, an operation button 7403, an external connection port 7404, a speaker 7405, a microphone 7406, and the like. Note that the mobile phone 7400 includes a secondary battery 7407. When the secondary battery of one embodiment of the present invention is used as the secondary battery 7407, a lightweight mobile phone with a long lifetime can be provided.

[0383]

FIG. 24B illustrates the mobile phone 7400 that is bent. When the whole mobile phone 7400 is curved by external force, the secondary battery 7407 included in the mobile phone 7400 is also curved. FIG. 24C illustrates the curved secondary battery 7407. The secondary battery 7407 is a thin storage battery. The secondary battery 7407 is curved and fixed. Note that the secondary battery 7407 includes a lead electrode 7408 electrically connected to a current collector 7409. The current collector 7409 is, for example, copper foil, and partly alloyed with gallium; thus, adhesion between the current collector 7409 and an active material layer in contact with the current collector 7409 is improved and the secondary battery 7407 can have high reliability even in a state of being bent.

[0384]

FIG. 24D illustrates an example of a bangle display device. A portable display device 7100 includes a housing 7101, a display portion 7102, an operation button 7103, and a secondary battery 7104. FIG. 24E illustrates the bent secondary battery 7104. When the curved secondary battery 7104 is on a user's arm, the housing changes its form and the curvature of a part or the whole of the secondary battery 7104 is changed. Note that the radius of curvature of a curve at a point refers to the radius of the circular arc that best approximates the curve at that point. The reciprocal of the radius of curvature is curvature. Specifically, part or the whole of the housing or the main surface of the secondary battery 7104 is changed in the range of radius of curvature from 40 mm to 150 mm. When the radius of curvature at the main surface of the secondary battery 7104 is greater than or equal to 40 mm and less than or equal to 150 mm, the reliability can be kept high. When the secondary battery of one embodiment of the present invention is used as the secondary battery 7104, a lightweight portable display device with a long lifetime can be provided.

[0385]

FIG. 24F illustrates an example of a watch-type portable information terminal. A portable information terminal 7200 includes a housing 7201, a display portion 7202, a band 7203, a buckle 7204, an operation button 7205, an input output terminal 7206, and the like.

[0386]

The portable information terminal 7200 is capable of executing a variety of applications such as mobile phone calls, e-mailing, viewing and editing texts, music reproduction, Internet communication, and a computer game.

[0387]

The display surface of the display portion 7202 is curved, and images can be displayed on the curved display surface. In addition, the display portion 7202 includes a touch sensor, and operation can be performed by touching the screen with a finger, a stylus, or the like. For example,

by touching an icon 7207 displayed on the display portion 7202, application can be started.

[0388]

With the operation button 7205, a variety of functions such as time setting, power on/off, on/off of wireless communication, setting and cancellation of a silent mode, and setting and cancellation of a power saving mode can be performed. For example, the functions of the operation button 7205 can be set freely by setting the operation system incorporated in the portable information terminal 7200.

[0389]

The portable information terminal 7200 can employ near field communication that is a communication method based on an existing communication standard. For example, mutual communication between the portable information terminal 7200 and a headset capable of wireless communication can be performed, and thus hands-free calling is possible.

[0390]

Moreover, the portable information terminal 7200 includes the input output terminal 7206, and data can be directly transmitted to and received from another information terminal via a connector. In addition, charging via the input output terminal 7206 is possible. Note that the charging operation may be performed by wireless power feeding without using the input output terminal 7206.

[0391]

The display portion 7202 of the portable information terminal 7200 includes the secondary battery of one embodiment of the present invention. When the secondary battery of one embodiment of the present invention is used, a lightweight portable information terminal with a long lifetime can be provided. For example, the secondary battery 7104 illustrated in FIG. 24E that is in the state of being curved can be provided in the housing 7201. Alternatively, the secondary battery 7104 illustrated in FIG. 24E can be provided in the band 7203 such that it can be curved.

[0392]

A portable information terminal 7200 preferably includes a sensor. As the sensor, for example a human body sensor such as a fingerprint sensor, a pulse sensor, or a temperature sensor, a touch sensor, a pressure sensitive sensor, an acceleration sensor, or the like is preferably mounted.

[0393]

FIG. 24G illustrates an example of an armband display device. A display device 7300 includes a display portion 7304 and the secondary battery of one embodiment of the present invention. The display device 7300 can include a touch sensor in the display portion 7304 and can serve as a portable information terminal.

[0394]

The display surface of the display portion 7304 is bent, and images can be displayed on the bent display surface. A display state of the display device 7300 can be changed by, for example, near field communication, which is a communication method based on an existing communication standard.

[0395]

The display device 7300 includes an input output terminal, and data can be directly transmitted to and received from another information terminal via a connector. In addition, charging via the input output terminal is possible. Note that the charging operation may be performed by wireless power feeding without using the input output terminal.

[0396]

When the secondary battery of one embodiment of the present invention is used as the secondary battery included in the display device 7300, a lightweight display device with a long lifetime can be provided.

[0397]

In addition, FIG. 24H, FIGS. 25A to 25C, and FIG. 26 show examples of electronic devices including the secondary battery with excellent cycle characteristics described in the above embodiment.

[0398]

When the secondary battery of one embodiment of the present invention is used as a secondary battery of a daily electronic device, a lightweight product with a long lifetime can be provided. As the daily electronic devices, an electric toothbrush, an electric shaver, electric beauty equipment, and the like are given. As secondary batteries of these products, in consideration of handling ease for users, small and lightweight stick type secondary batteries with high capacity are desired.

[0399]

FIG. 24H is a perspective view of a device which is called a vaporizer. In FIG. 24H, a vaporizer 7500 includes an atomizer 7501 including a heating element, a secondary battery 7504 supplying power to the atomizer, and a cartridge 7502 including a liquid supply bottle, a sensor, and the like. To improve safety, a protection circuit which prevents overcharge and overdischarge of the secondary battery 7504 may be electrically connected to the secondary battery 7504. The secondary battery 7504 in FIG. 24H includes an output terminal for connecting to a charger. When the vaporizer 7500 is held by a user, the secondary battery 7504 becomes a tip portion; thus, it is preferable that the secondary battery 7504 have a short total length and be lightweight. With the secondary battery of one embodiment of the present invention which has high capacity and

excellent cycle characteristics, the small and lightweight vaporizer 7500 which can be used for a long time for a long period can be provided.

[0400]

Next, FIGS. 25A and 25B illustrate an example of a foldable tablet terminal. A tablet terminal 9600 illustrated in FIGS. 25A and 25B includes a housing 9630a, a housing 9630b, a movable portion 9640 connecting the housings 9630a and 9630b, a display portion 9631 including a display portion 9631a and a display portion 9631b, a display mode changing switch 9626, a power switch 9627, a power saving mode changing switch 9625, a fastener 9629, and an operation switch 9628. A flexible panel is used for the display portion 9631, whereby a tablet terminal with a larger display portion can be provided. FIG. 25A illustrates the tablet terminal 9600 that is opened, and FIG. 25B illustrates the tablet terminal 9600 that is closed.

[0401]

The tablet terminal 9600 includes a power storage unit 9635 inside the housings 9630a and 9630b. The power storage unit 9635 is provided across the housings 9630a and 9630b, passing through the movable portion 9640.

[0402]

Part of the display portion 9631a can be a touch panel region ~~9632a~~ and data can be input when a displayed operation key ~~9638~~ is touched. Although a structure in which a half region in the display portion 9631a has only a display function and the other half region has a touch panel function is shown as an example, the display portion 9631a is not limited to the structure. The whole region in the display portion 9631a may have a touch panel function. For example, the display portion 9631a can display keyboard buttons in the whole region to be a touch panel, and the display portion 9631b can be used as a display screen.

[0403]

Like the display portion 9631a, part of the display portion 9631b can be a touch panel region ~~9632b~~. A switching button ~~9639~~ for showing/hiding a keyboard of the touch panel is touched with a finger, a stylus, or the like, so that keyboard buttons can be displayed on the display portion 9631b.

[0404]

Touch input can be performed in the touch panel region ~~9632a~~ and the touch panel region ~~9632b~~ at the same time.

[0405]

The display mode switch 9626 can switch the display between a portrait mode and a landscape mode, and between monochrome display and color display, for example. The power saving mode changing switch 9625 can control display luminance in accordance with the amount

of external light in use of the tablet terminal 9600, which is measured with an optical sensor incorporated in the tablet terminal 9600. Another detection device including a sensor for detecting inclination, such as a gyroscope sensor or an acceleration sensor, may be incorporated in the tablet terminal, in addition to the optical sensor.

[0406]

Although the display portion 9631a and the display portion 9631b have the same area in FIG. 25A, one embodiment of the present invention is not limited to this example. The display portion 9631a and the display portion 9631b may have different areas or different display quality. For example, one display panel may be capable of higher-definition display than the other display panel.

[0407]

The tablet terminal is closed in FIG. 25B. The tablet terminal includes the housing 9630, a solar cell 9633, and a charge and discharge control circuit 9634 including a DCDC converter 9636. The power storage unit of one embodiment of the present invention is used as the power storage unit 9635.

[0408]

The tablet terminal 9600 can be folded such that the housings 9630a and 9630b overlap with each other when not in use. Thus, the display portions 9631a and 9631b can be protected, which increases the durability of the tablet terminal 9600. With the power storage unit 9635 including the secondary battery of one embodiment of the present invention which has high capacity and excellent cycle characteristics, the tablet terminal 9600 which can be used for a long time for a long period can be provided.

[0409]

The tablet terminal illustrated in FIGS. 25A and 25B can also have a function of displaying various kinds of data (e.g., a still image, a moving image, and a text image), a function of displaying a calendar, a date, or the time on the display portion, a touch-input function of operating or editing data displayed on the display portion by touch input, a function of controlling processing by various kinds of software (programs), and the like.

[0410]

The solar cell 9633, which is attached on the surface of the tablet terminal, supplies electric power to a touch panel, a display portion, an image signal processor, and the like. Note that the solar cell 9633 can be provided on one or both surfaces of the housing 9630 and the power storage unit 9635 can be charged efficiently. The use of a lithium-ion battery as the power storage unit 9635 brings an advantage such as reduction in size.

[0411]

The structure and operation of the charge and discharge control circuit 9634 illustrated in FIG. 25B will be described with reference to a block diagram in FIG. 25C. The solar cell 9633, the power storage unit 9635, the DC-DC converter 9636, a converter 9637, switches SW1 to SW3, and the display portion 9631 are illustrated in FIG. 25C, and the power storage unit 9635, the DC-DC converter 9636, the converter 9637, and the switches SW1 to SW3 correspond to the charge and discharge control circuit 9634 in FIG. 25B.

[0412]

First, an example of the operation in the case where power is generated by the solar cell 9633 using external light is described. The voltage of electric power generated by the solar cell is raised or lowered by the DCDC converter 9636 to a voltage for charging the power storage unit 9635. When the power from the solar cell 9633 is used for the operation of the display portion 9631, the switch SW1 is turned on and the voltage of the power is raised or lowered by the converter 9637 to a voltage needed for operating the display portion 9631. When display on the display portion 9631 is not performed, the switch SW1 is turned off and the switch SW2 is turned on, so that the power storage unit 9635 can be charged.

[0413]

Note that the solar cell 9633 is described as an example of a power generation means; however, one embodiment of the present invention is not limited to this example. The power storage unit 9635 may be charged using another power generation means such as a piezoelectric element or a thermoelectric conversion element (Peltier element). For example, the power storage unit 9635 may be charged with a non-contact power transmission module that transmits and receives power wirelessly (without contact) to charge the battery or with a combination of other charging means.

[0414]

FIG. 26 illustrates other examples of electronic devices. In FIG. 26, a display device 8000 is an example of an electronic device including a secondary battery 8004 of one embodiment of the present invention. Specifically, the display device 8000 corresponds to a display device for TV broadcast reception and includes a housing 8001, a display portion 8002, speaker portions 8003, the secondary battery 8004, and the like. The secondary battery 8004 of one embodiment of the present invention is provided in the housing 8001. The display device 8000 can receive electric power from a commercial power supply. Alternatively, the display device 8000 can use electric power stored in the secondary battery 8004. Thus, the display device 8000 can operate with the use of the secondary battery 8004 of one embodiment of the present invention as an uninterruptible power supply even when electric power cannot be supplied from a commercial power supply due to power failure or the like.

[0415]

A semiconductor display device such as a liquid crystal display device, a light-emitting device in which a light-emitting element such as an organic EL element is provided in each pixel, an electrophoretic display device, a digital micromirror device (DMD), a plasma display panel (PDP), or a field emission display (FED) can be used for the display portion 8002.

[0416]

Note that the display device includes, in its category, all of information display devices for personal computers, advertisement displays, and the like other than TV broadcast reception.

[0417]

In FIG. 26, an installation lighting device 8100 is an example of an electronic device using a secondary battery 8103 of one embodiment of the present invention. Specifically, the lighting device 8100 includes a housing 8101, a light source 8102, the secondary battery 8103, and the like. Although FIG. 26 illustrates the case where the secondary battery 8103 is provided in a ceiling 8104 on which the housing 8101 and the light source 8102 are installed, the secondary battery 8103 may be provided in the housing 8101. The lighting device 8100 can receive electric power from a commercial power supply. Alternatively, the lighting device 8100 can use electric power stored in the secondary battery 8103. Thus, the lighting device 8100 can operate with the use of the secondary battery 8103 of one embodiment of the present invention as an uninterruptible power supply even when electric power cannot be supplied from a commercial power supply due to power failure or the like.

[0418]

Note that although the installation lighting device 8100 provided in the ceiling 8104 is illustrated in FIG. 26 as an example, the secondary battery of one embodiment of the present invention can be used as an installation lighting device provided in, for example, a wall 8105, a floor 8106, a window 8107, or the like other than the ceiling 8104. Alternatively, the secondary battery can be used in a tabletop lighting device or the like.

[0419]

As the light source 8102, an artificial light source which emits light artificially by using power can be used. Specifically, an incandescent lamp, a discharge lamp such as a fluorescent lamp, and a light-emitting element such as an LED or an organic EL element are given as examples of the artificial light source.

[0420]

In FIG. 26, an air conditioner including an indoor unit 8200 and an outdoor unit 8204 is an example of an electronic device including a secondary battery 8203 of one embodiment of the present invention. Specifically, the indoor unit 8200 includes a housing 8201, an air outlet 8202,

the secondary battery 8203, and the like. Although FIG. 26 illustrates the case where the secondary battery 8203 is provided in the indoor unit 8200, the secondary battery 8203 may be provided in the outdoor unit 8204. Alternatively, the secondary batteries 8203 may be provided in both the indoor unit 8200 and the outdoor unit 8204. The air conditioner can receive electric power from a commercial power supply. Alternatively, the air conditioner can use electric power stored in the secondary battery 8203. Particularly in the case where the secondary batteries 8203 are provided in both the indoor unit 8200 and the outdoor unit 8204, the air conditioner can operate with the use of the secondary battery 8203 of one embodiment of the present invention as an uninterruptible power supply even when electric power cannot be supplied from a commercial power supply due to power failure or the like.

[0421]

Note that although the split-type air conditioner including the indoor unit and the outdoor unit is illustrated in FIG. 26 as an example, the secondary battery of one embodiment of the present invention can be used in an air conditioner in which the functions of an indoor unit and an outdoor unit are integrated in one housing.

[0422]

In FIG. 26, an electric refrigerator-freezer 8300 is an example of an electronic device using a secondary battery 8304 of one embodiment of the present invention. Specifically, the electric refrigerator-freezer 8300 includes a housing 8301, a refrigerator door 8302, a freezer door 8303, the secondary battery 8304, and the like. The secondary battery 8304 is provided in the housing 8301 in FIG. 26. The electric refrigerator-freezer 8300 can receive electric power from a commercial power supply. Alternatively, the electric refrigerator-freezer 8300 can use electric power stored in the secondary battery 8304. Thus, the electric refrigerator-freezer 8300 can operate with the use of the secondary battery 8304 of one embodiment of the present invention as an uninterruptible power supply even when electric power cannot be supplied from a commercial power supply due to power failure or the like.

[0423]

Note that among the electronic devices described above, a high-frequency heating apparatus such as a microwave oven and an electronic device such as an electric rice cooker require high power in a short time. The tripping of a circuit breaker of a commercial power source in use of electronic devices can be prevented by using the secondary battery of one embodiment of the present invention as an auxiliary power source for supplying power which cannot be supplied enough by a commercial power source.

[0424]

In addition, in a time period when electronic devices are not used, particularly when the

proportion of the amount of power which is actually used to the total amount of power which can be supplied from a commercial power source (such a proportion referred to as a usage rate of power) is low, power can be stored in the secondary battery, whereby the usage rate of power can be reduced in a time period when the electronic devices are used. For example, in the case of the electric refrigerator-freezer 8300, power can be stored in the secondary battery 8304 in night time when the temperature is low and the refrigerator door 8302 and the freezer door 8303 are not often opened and closed. On the other hand, in daytime when the temperature is high and the refrigerator door 8302 and the freezer door 8303 are frequently opened and closed, the secondary battery 8304 is used as an auxiliary power source; thus, the usage rate of power in daytime can be reduced.

[0425]

According to one embodiment of the present invention, the secondary battery can have excellent cycle characteristics and improve reliability. Furthermore, in accordance with one embodiment of the present invention, a secondary battery with high capacity can be obtained; thus, the secondary battery itself can be made more compact and lightweight as a result of improved characteristics of the secondary battery. Thus, the secondary battery of one embodiment of the present invention is used in the electronic device described in this embodiment, whereby a more lightweight electronic device with a longer lifetime can be obtained. This embodiment can be implemented in appropriate combination with any of the other embodiments.

[0426]

(Embodiment 5)

In this embodiment, examples of vehicles including the secondary battery of one embodiment of the present invention are described.

[0427]

The use of secondary batteries in vehicles enables production of next-generation clean energy vehicles such as hybrid electric vehicles (HEVs), electric vehicles (EVs), and plug-in hybrid electric vehicles (PHEVs).

[0428]

FIGS. 27A to 27C each illustrate an example of a vehicle using the secondary battery of one embodiment of the present invention. An automobile 8400 illustrated in FIG. 27A is an electric vehicle that runs on the power of an electric motor. Alternatively, the automobile 8400 is a hybrid electric vehicle capable of driving appropriately using either an electric motor or an engine. One embodiment of the present invention can provide a high-mileage vehicle. The automobile 8400 includes the secondary battery. As the secondary battery, the modules of the secondary batteries illustrated in FIGS. 12C and 12D may be arranged to be used in a floor portion

in the automobile. Alternatively, a battery pack in which a plurality of secondary batteries each of which is illustrated in FIGS. 17A to 17C are combined may be placed in a floor portion in the automobile. The secondary battery is used not only for driving an electric motor 8406, but also for supplying electric power to a light-emitting device such as a headlight 8401 or a room light (not illustrated).

[0429]

The secondary battery can also supply electric power to a display device of a speedometer, a tachometer, or the like included in the automobile 8400. Furthermore, the secondary battery can supply electric power to a semiconductor device included in the automobile 8400, such as a navigation system.

[0430]

FIG. 27B illustrates an automobile 8500 including the secondary battery. The automobile 8500 can be charged when the secondary battery is supplied with electric power through external charging equipment by a plug-in system, a contactless power feeding system, or the like. In FIG. 27B, [[a]] secondary ~~battery~~ batteries 8024 and 8025 included in the automobile 8500 is charged with the use of a ground-based charging apparatus 8021 through a cable 8022. In charging, a given method such as CHAdeMO (registered trademark) or Combined Charging System may be employed as a charging method, the standard of a connector, or the like as appropriate. The ground-based charging apparatus 8021 may be a charging station provided in a commerce facility or a power source in a house. With the use of a plug-in technique, the secondary battery 8024 included in the automobile 8500 can be charged by being supplied with electric power from the outside, for example. The charging can be performed by converting AC electric power into DC electric power through a converter such as an AC-DC converter.

[0431]

Furthermore, although not illustrated, the vehicle may include a power receiving device so that it can be charged by being supplied with electric power from an above-ground power transmitting device in a contactless manner. In the case of the contactless power feeding system, by fitting a power transmitting device in a road or an exterior wall, charging can be performed not only when the electric vehicle is stopped but also when driven. In addition, the contactless power feeding system may be utilized to perform transmission and reception of electric power between vehicles. A solar cell may be provided in the exterior of the automobile to charge the secondary battery when the automobile stops or moves. To supply electric power in such a contactless manner, an electromagnetic induction method or a magnetic resonance method can be used.

[0432]

FIG. 27C shows an example of a motorcycle using the secondary battery of one

embodiment of the present invention. A motor scooter 8600 illustrated in FIG. 27C includes a secondary battery 8602, side mirrors 8601, and indicators 8603. The secondary battery 8602 can supply electric power to the indicators 8603.

[0433]

Furthermore, in the motor scooter 8600 illustrated in FIG. 27C, the secondary battery 8602 can be held in a storage unit under seat 8604. The secondary battery 8602 can be held in the storage unit under seat 8604 even with a small size. The secondary battery 8602 is detachable; thus, the secondary battery 8602 is carried indoors when it is charged, and is stored before the motorcycle is driven.

[0434]

In accordance with one embodiment of the present invention, the secondary battery can have improved cycle characteristics and the capacity of the secondary battery can be increased. Thus, the secondary battery itself can be made more compact and lightweight. The compact and lightweight secondary battery contributes to a reduction in the weight of a vehicle, and thus increases the driving radius. Furthermore, the secondary battery included in the vehicle can be used as a power source for supplying electric power to products other than the vehicle. In such a case, the use of a commercial power source can be avoided at peak time of electric power demand, for example. If the use of a commercial power source can be avoided at peak time of electric power demand, the avoidance can contribute to energy saving and a reduction in carbon dioxide emissions. Moreover, if the cycle characteristics are excellent, the secondary battery can be used for a long period; thus, the use amount of rare metals such as cobalt can be reduced.

[0435]

This embodiment can be implemented in appropriate combination with the other embodiments.

[Example 1]

[0436]

In this example, the positive electrode active materials which are embodiments of the present invention are formed, and the observation results of the positive electrode active materials by STEM, the results of TEM images subjected to fast Fourier transform, and the analysis results obtained by energy dispersive X-ray spectroscopy (EDX) are described. In addition, the evaluation results of characteristics of secondary batteries containing the positive electrode active materials are described.

[0437]

[Formation of positive electrode active material]

<<Sample 01>>

In this example, a positive electrode active material of Sample 01, which contains lithium cobaltate as a composite oxide of lithium and a first transition metal contained in a first region, lithium titanate as an oxide of a second transition metal contained in a second region, and magnesium oxide as an oxide of a representative element contained in a third region, was formed.

[0438]

In this example, lithium cobalt oxide particles (C-20F, produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD.) were used as a starting material. Thus, in this example, Step 12 and Step 13 described in Embodiment 1 were omitted. Note that the above-described lithium cobalt oxide particles each have a particle diameter of approximately 20 μm, and contain fluorine, magnesium, calcium, sodium, silicon, sulfur, and phosphorus in a region which can be analyzed by XPS.

[0439]

Next, as Step 14, the lithium cobalt oxide particles containing magnesium and fluorine were coated with a material containing titanium by a sol-gel method. Specifically, TTIP was dissolved in isopropanol, and an isopropanol solution of TTIP was formed. Then, the lithium cobalt oxide particles were mixed into the solution so that TTIP to lithium cobalt oxide containing magnesium and fluorine was 0.01 ml/g.

[0440]

The above mixed solution was stirred with a magnetic stirrer for four hours, at 25 °C, and at a humidity of 90 %RH. Through the process, water in an atmosphere and TTIP caused hydrolysis and polycondensation reaction, and a layer containing titanium was formed on the surface of the lithium cobalt oxide particle containing magnesium and fluorine.

[0441]

The mixed solution which had been subjected to the above process was filtered to collect the residue. As a filter for filtration, Kiriyama filter paper (No. 4) was used.

[0442]

The collected residue was dried in a vacuum at 70 °C for one hour.

[0443]

Next, the lithium cobalt oxide particles coated with the material containing titanium was heated. With a muffle furnace, the heating was performed under the following conditions: the flow rate of dry air was 10 L/min; the temperature was 800 °C (the temperature rising rate was 200 °C/h); and the retention time was two hours. The dew point of the dry air is preferably lower than or equal to −109 °C.

[0444]

Then, the heated particles were cooled to room temperature. The time of decreasing

temperature from the retention temperature to room temperature was 10 hours to 15 hours. After that, crushing treatment was performed. In the crushing treatment, the particles were made to pass through a sieve. The sieve has an aperture width of 53 μm.

[0445]

Lastly, the cooled particles were collected, and the positive electrode active material of Sample 01 was obtained.

[0446]

<<Sample 02>>

Sample 02 was formed as a comparative example by heating lithium cobalt oxide particles containing magnesium and fluorine without being coated with a material containing titanium.

[0447]

Lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-20F) were used as the lithium cobalt oxide particles containing magnesium and fluorine.

[0448]

The lithium cobalt oxide particles containing magnesium and fluorine were heated. The heating was performed under the following conditions: the temperature was 800 °C (the temperature rising rate was 200 °C/h); the retention time was two hours; and the flow rate of oxygen was 10 L/min.

[0449]

The heated particles were cooled and made to pass through a sieve like Sample 01 to obtain a positive electrode active material of Sample 02.

[0450]

It is probable that Sample 02 is a positive electrode active material which contains lithium cobalt oxide inside and includes a region containing magnesium in a superficial portion.

[0451]

<<Sample 03>>

Sample 03 was formed as a comparative example in the following manner: a region containing titanium was formed in lithium cobalt oxide particles which do not contain magnesium by a sol-gel method and then the lithium cobalt oxide particles were heated.

[0452]

Lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-10N) were used. In the lithium cobalt oxide particles, magnesium is not detected and fluorine is detected at approximately 1 atomic% by XPS.

[0453]

A region containing titanium was formed by a sol-gel method in the lithium cobalt oxide particles, and the lithium cobalt oxide particles were dried, heated, cooled, and made to pass through a sieve, like Sample 01. The obtained lithium cobalt oxide particles were used as a positive electrode active material of Sample 03.

[0454]

It is probable that Sample 03 is a positive electrode active material which contains lithium cobalt oxide inside and includes a region containing titanium in a superficial portion.

[0455]

<<Sample 04>>

For Sample 04, as a comparative example, lithium cobalt oxide particles were used as it is without being heated.

[0456]

Lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-10N) were used.

[0457]

Sample 04 is a positive electrode active material which does not have a coating layer.

[0458]

<<Sample 05>>

For Sample 05, as a comparative example, lithium cobalt oxide particles containing magnesium and fluorine were used as it is without being heated.

[0459]

Lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-20F) were used as the lithium cobalt oxide particles containing magnesium and fluorine. That is, Sample 05 was used as the same as the starting material of Sample 01.

[0460]

Table 1 shows the conditions of Sample 01 to Sample 05.

[0461]

[Table 1]

	Conditions
Sample 01	$LiCoO_2$+Mg+F, coated with a material containing Ti, heated
Sample 02	$LiCoO_2$+Mg+F, heated
Sample 03	$LiCoO_2$, coated with a material containing Ti, heated
Sample 04	$LiCoO_2$, not heated
Sample 05	$LiCoO_2$+Mg+F, not heated

[0462]

[STEM]

The obtained positive electrode active material of Sample 01 was observed by an electron microscope (JEM-ARM200F, manufactured by JEOL Ltd.) under the condition where the acceleration voltage was 200 kV. FIG. 28 shows the obtained electron microscope image. As shown in FIG. 28, the positive electrode active material probably includes three different regions: the first region 101; the second region 102; and the third region 103. The third region 103 is observed as a region brighter than the first region 101 and the second region 102. Furthermore, crystal orientations of the first region 101 and the second region 102 are partly aligned with each other, and crystal orientations of the second region 102 and the third region 103 are partly aligned with each other.

[0463]

[STEM-FFT]

FIG. 29A1 shows a fast Fourier transform (FFT) image of a region 103FFT in the STEM image of FIG. 28. In FIG. 29A2, a center point O of FIG. 29A1 is shown by a cross, and bright points A, B, and C are each surrounded by a circle. Similarly, FIG. 29B1 shows an FFT image of a region 102FFT. In FIG. 29B2, a center point O of FIG. 29B1 is shown by a cross, and bright points A, B, and C are each surrounded by a circle. In addition, FIG. 29C1 shows an FFT image of a region 101FFT. In FIG. 29C2, a center point O of FIG. 29C1 is shown by a cross, and bright points A, B, and C are each surrounded by a circle.

[0464]

In FIG. 29A2, a distance *d* between the bright point A and the center point O is 0.256 nm, a distance *d* between the bright point B and the center point O is 0.241 nm, and a distance *d* between the bright point C and the center point O is 0.209 nm. In addition, ∠COA is 121°, ∠COB is 52°, and ∠AOB is 69°. From these results, the region 103FFT probably contains magnesium oxide (MgO, cubic crystal).

[0465]

Similarly, in FIG. 29B2, the distance *d* between the bright point A and the center point O is 0.238 nm, the distance *d* between the bright point B and the center point O is 0.225 nm, and the distance *d* between the bright point C and the center point O is 0.198 nm. In addition, ∠COA is 123°, ∠COB is 52°, and ∠AOB is 71°. From these results, the region 102FFT probably contains lithium titanate ($LiTiO_2$, cubic crystal).

[0466]

In FIG. 29C2, the distance *d* between the bright point A and the center point O is 0.240 nm, the distance *d* between the bright point B and the center point O is 0.235 nm, and the distance

d between the bright point C and the center point O is 0.196 nm. In addition, ∠COA is 126°, ∠COB is 52°, and ∠AOB is 74°. From these results, the region 101FFT probably contains lithium cobaltate ($LiCoO_2$, rhombohedral).

[0467]

[EDX]

FIGS. 30A1, 30A2, 30B1, 30B2, 30C1, and 30C2 show a high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) image and element mapping images with EDX of the positive electrode active material of Sample 01. FIG. 30A1 shows a HAADF-STEM image, FIG. 30A2 shows a mapping image of oxygen atoms, FIG. 30B1 shows a mapping image of cobalt atoms, FIG. 30B2 shows a mapping image of fluorine atoms, FIG. 30C1 shows a mapping image of titanium atoms, and FIG. 30C2 shows a mapping image of magnesium atoms. Note that in EDX element mapping images in FIGS. 30A2, 30B1, 30B2, 30C1, and 30C2 and FIGS. 31A2, 31B1, 31B2, 31C1, and 31C2, a region where the number of elements is less than or equal to a lower limit of the detection is indicated in white, and as the number of elements is increased, the white region becomes black.

[0468]

As shown in FIGS. 30A2 and 30B1, it is found that the oxygen atoms and the cobalt atoms are distributed in the whole of the positive electrode active material particle. In contrast, as shown in FIGS. 30B2, 30C1, and 30C2, it is found that the fluorine atoms, the titanium atoms, and the magnesium atoms are unevenly distributed in a region close to the surface of the positive electrode active material.

[0469]

Next, FIGS. 31A1, 31A2, 31B1, 31B2, 31C1, and 31C2 show a HAADF-STEM image and element mapping images with EDX of the positive electrode active material of Sample 05, which is a comparative example. FIG. 31A1 shows a HAADF-STEM image, FIG. 31A2 shows a mapping image of oxygen atoms, FIG. 31B1 shows a mapping image of cobalt atoms, FIG. 31B2 shows a mapping image of fluorine atoms, FIG. 31C1 shows a mapping image of titanium atoms, and FIG. 31C2 shows a mapping image of magnesium atoms.

[0470]

As shown in FIGS. 31B2 and 31C2, it is found that, even in Sample 05 which is not heated, a certain amount of magnesium and fluorine is unevenly distributed in the vicinity of the surface.

[0471]

[EDX line analysis]

FIG. 32 shows results of line analysis with TEM-EDX performed on a cross section of the vicinity of the surface of the positive electrode active material of Sample 01. FIG. 32 is a

graph showing data detected on a line connecting the outside of the positive electrode active material of Sample 01 to the inside of the positive electrode active material, and a distance of 0 nm indicates the outside of the positive electrode active material and a distance of 14 nm indicates the inside of the particle. With EDX, the analysis region tends to be large, so that elements not only at a center of an electron beam irradiation region but also in a region around the center may be detected.

[0472]

As shown in FIG. 32, it is found that there are peaks of magnesium and titanium in the vicinity of the surface of the positive electrode active material of Sample 01, the distribution of magnesium is closer to the surface than the distribution of titanium is. It is also found that the peak of magnesium is closer to the surface than the peak of titanium is. In addition, it is probable that cobalt and oxygen are present from the outermost surface of the positive electrode active material particle.

[0473]

As shown in FIG. 32, fluorine is hardly detected. This is probably because fluorine, which is a light element, is difficult to detect with EDX.

[0474]

From the above STEM images, FFT images, element mapping images with EDX, and EDX line analysis, it is found that Sample 01 is a positive electrode active material of one embodiment of the present invention, which includes the first region containing lithium cobaltate, the second region containing lithium, titanium, cobalt, and oxygen, and the third region containing magnesium and oxygen. It is found that, in Sample 01, part of the second region and part of the third region overlap.

[0475]

In the graph of FIG. 32, the amount of detected oxygen is stable at a distance of 4 nm or more. Thus, in this example, the average value O_{ave} of the amount of detected oxygen in the stable region is obtained, and a distance x of the measurement point at which the measurement value closest to 0.5 O_{ave}, the value of 50 % of the average value O_{ave}, is obtained is assumed to be the surface of the positive electrode active material particle.

[0476]

In this example, the average value O_{ave} of the amount of detected oxygen in a range from a distance of 4 nm to a distance of 14 nm is 674.2. The x axis of the measurement point at which the measurement value closest to 337.1, which is 50 % of 674.2, is obtained indicates a distance of 1.71 nm. Thus, in this example, a distance of 1.71 nm in the graph of FIG. 32 is assumed to be the surface of the positive electrode active material particle.

[0477]

When the surface of the positive electrode active material particle is set at a distance of 1.71 nm in FIG. 32, the peak of magnesium and the peak of titanium are present at 0.72 nm and 1.00 nm, respectively, from the surface of the positive electrode active material particle.

[0478]

The concentration of magnesium is higher than or equal to 1/5 of the peak from the surface of the positive electrode active material particle to a distance of 4.42 nm, that is, to a region at 2.71 nm from the surface. The measurement value of magnesium is less than 1/5 of the peak at a distance of 4.57 nm or more, that is, at a depth of 2.86 nm or more from the surface of the positive electrode active material particle. Thus, it is found that in Sample 01, a region from the surface to a depth of 2.71 nm is the third region.

[0479]

Furthermore, the concentration of titanium is higher than or equal to 1/2 of the peak from a distance of 2.14 nm to a distance of 3.42 nm. Thus, it is found that a region from a depth of 0.43 nm to a depth of 1.71 nm from the surface of the positive electrode active material particle is the second region.

[0480]

Next, evaluation results of charge and discharge characteristics of secondary batteries which are fabricated using the positive electrode active materials of Sample 01 to Sample 05 formed in the above manner are described.

[0481]

[Fabrication of secondary batteries]

CR2032 coin-type secondary batteries (with a diameter of 20 mm and a height of 3.2 mm) were fabricated using the positive electrode active materials of Sample 01 to Sample 05 formed in the above manner.

[0482]

A positive electrode formed by applying slurry in which a positive electrode active material (LCO), acetylene black (AB), and polyvinylidene fluoride (PVDF) were mixed at a weight ratio of 95:2.5:2.5 to a current collector was used.

[0483]

A lithium metal was used for the counter electrode.

[0484]

As an electrolyte contained in an electrolyte solution, 1 mol/L lithium hexafluorophosphate ($LiPF_6$) was used, and as the electrolyte solution, a solution in which ethylene carbonate (EC) and diethyl carbonate (DEC) at a volume ratio of 3:7 and vinylene carbonate (VC)

at a 2 weight% were mixed was used.

[0485]

A positive electrode can and a negative electrode can were formed of stainless steel (SUS).

[0486]

[Evaluation of charge and discharge characteristics]

Next, charge and discharge characteristics of the secondary batteries of Sample 01 and Sample 05 formed in the above manner were evaluated. The measurement temperature was 25 °C. Twenty cycles of charging and discharging were performed at 4.6 V (CCCV, 0.5 C, a cutoff current of 0.01 C) and 2.5 V (CC, 0.5 C), respectively. Here, 1C was set to 137 mA/g, which was a current value per weight of the positive electrode active material.

[0487]

FIG. 33 is a graph showing charge and discharge characteristics of the secondary battery using the positive electrode active material of Sample 01. FIG. 33 shows excellent charge and discharge characteristics with a wide plateau. In addition, results of 20 cycles of charging and discharging almost overlap, which means that the cycle characteristics are excellent.

[0488]

FIG. 34 is a graph showing charge and discharge characteristics of the secondary battery of Sample 05, which is a comparative example. In the initial cycles, excellent charge and discharge characteristics are shown; however, as indicated by arrows in FIG. 34, charge capacity and discharge capacity decrease with an increase in cycles.

[0489]

[Evaluation of cycle characteristics]

<<Charging at 4.4 V>>

The cycle characteristics of the secondary batteries of Sample 01 and Sample 05 charged at 4.4 V were evaluated. The measurement temperature was 25 °C. The charging was performed at 4.4 V (CCCV, 0.5 C, a cutoff current of 0.01 C), and the discharging was performed at 2.5 V (CC, 0.5 C).

[0490]

FIG. 35 is a graph showing the cycle characteristics of the secondary batteries charged at 4.4 V. In FIG. 35, a solid line and a dotted line indicate secondary batteries containing the positive electrode active materials of Sample 01 and Sample 05, respectively. As shown in FIG. 35, in the secondary battery containing Sample 01, an energy density retention rate is 99.5 % even after 50 cycles of charging and discharging were performed, which shows extremely excellent cycle characteristics. In the secondary battery containing Sample 05, an energy density retention rate is 94.3 % after 50 cycles were performed.

[0491]

<<Charging at 4.6 V>>

The cycle characteristics of the secondary batteries of Sample 01 to Sample 04 charged at 4.6 V were evaluated. The measurement temperature was 25 °C. The charging was performed to 4.6 V (CCCV, 0.5 C, a cutoff current of 0.01 C), and the discharging was performed to 2.5 V (CC, 0.5 C).

[0492]

FIG. 36 is a graph showing the cycle characteristics charged at 4.6 V. As shown in FIG. 36, in the secondary battery containing Sample 01, which is the positive electrode active material of one embodiment of the present invention, an energy density retention rate is 94.1 % even after 50 cycles of charging and discharging were performed at a high voltage of 4.6 V, which shows extremely excellent cycle characteristics. On the other hand, the secondary batteries containing the positive electrode active materials of Sample 02, Sample 03, and Sample 04, which are comparative examples, are inferior to that of Sample 01, and in Sample 04, for example, energy density retention rate is 33.2 % after 50 cycles were performed.

[0493]

As described above, it is found that the positive electrode active material with the structure of one embodiment of the present invention can achieve an advantageous effect when charging and discharging is performed at a voltage higher than 4.4 V.

[Example 2]

[0494]

In this example, the positive electrode active materials which are embodiments of the present invention are formed, and results of analysis which is different from that in Example 1 are described. In addition, evaluation results of characteristics of secondary batteries containing the positive electrode active materials under conditions different from those in Example 1 are described.

[0495]

In this example, a positive electrode active material which contains lithium cobaltate as a composite oxide of lithium and a first transition metal contained in a first region, lithium titanate as an oxide of a second transition metal contained in a second region, and magnesium oxide as an oxide of a representative element contained in a third region, was formed.

[0496]

[Formation of positive electrode active material and fabrication of secondary battery]

<<Sample 06 and Sample 07>>

In this example, lithium cobalt oxide particles (C-20F, produced by NIPPON

CHEMICAL INDUSTRIAL CO., LTD.) were used as a starting material.

[0497]

Next, as Step 14, the lithium cobalt oxide particles were coated with titanium oxide by a sol-gel method and dried. Step 14 was performed in a manner similar to that in Example 1 except that mixture was performed so that TTIP to lithium cobalt oxide was 0.004 ml/g. The lithium cobalt oxide particles which are coated with the titanium oxide and are not heated yet are referred to as Sample 06.

[0498]

Next, Sample 06, which is the lithium cobalt oxide particles coated with the titanium oxide, was heated. With a muffle furnace, the heating was performed at 800 °C in an oxygen atmosphere under the following conditions: the retention time was two hours; and the flow rate of oxygen was 10 L/min.

[0499]

Then, as in Example 1, the particles were cooled and collected to obtain the positive electrode active material. The heated positive electrode active material is referred to as Sample 07.

[0500]

[TEM-EDX]

Sample 06 and Sample 07, in particular, cracks generated in the particles and the vicinity of the cracks were subjected to analysis with TEM-EDX.

[0501]

First, results of TEM-EDX plane analysis of titanium are shown in FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2 and FIGS. 38A, 38B1, 38B2, 38C1, 38C2, 38D1, 38D2, 38E1, and 38E2.

[0502]

FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2 show TEM-EDX analysis results of Sample 06 before heating. FIG. 37A is a cross-sectional TEM image showing the surfaces of the particles and the crack portions. FIG. 37B1 and FIG. 37B2 show a HAADF-STEM image and a Ti mapping image of a region including the surface of the particle that is denoted by a circle marked with "1" in FIG. 37A, respectively. Similarly, FIG. 37C1 and FIG. 37C2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 20 nm from the surface in the crack portion that is denoted by a circle marked with "2" in FIG. 37A, respectively. FIG. 37D1 and FIG. 37D2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 500 nm from the surface in the crack portion that is denoted by a circle marked with "3" in FIG. 37A, respectively. FIG. 37E1 and

FIG. 37E2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 1000 nm from the surface in the crack portion that is denoted by a circle marked with "4" in FIG. 37A, respectively. Note that in EDX element mapping images in FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2, FIGS. 38A, 38B1, 38B2, 38C1, 38C2, 38D1, 38D2, 38E1, and 38E2, FIGS. 39A, 39B1, 39B2, 39C1, 39C2, 39D1, 39D2, 39E1, and 39E2, and FIGS. 40A, 40B1, 40B2, 40C1, 40C2, 40D1, 40D2, 40E1, and 40E2, a region where the number of elements is less than or equal to a lower limit of the detection is indicated in white, and as the number of elements is increased, the white region becomes black.

[0503]

FIGS. 38A, 38B1, 38B2, 38C1, 38C2, 38D1, 38D2, 38E1, and 38E2 show TEM-EDX analysis results of Sample 07 after heating. FIG. 38A is a cross-sectional TEM image showing the surfaces of the particles and the crack portions. FIG. 38B1 and FIG. 38B2 show a HAADF-STEM image and a Ti mapping image of a region including the surface of the particle that is denoted by a circle marked with "1" in FIG. 38A, respectively. Similarly, FIG. 38C1 and FIG. 38C2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 20 nm from the surface in the crack portion that is denoted by a circle marked with "2" in FIG. 38A, respectively. FIG. 38D1 and FIG. 38D2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 500 nm from the surface in the crack portion that is denoted by a circle marked with "3" in FIG. 38A, respectively. FIG. 38E1 and FIG. 38E2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 1000 nm from the surface in the crack portion that is denoted by a circle marked with "4" in FIG. 38A, respectively.

[0504]

As shown in FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2 and FIGS. 38A, 38B1, 38B2, 38C1, 38C2, 38D1, 38D2, 38E1, and 38E2, in Sample 06 before heating, segregation of titanium on the surfaces of the particles is observed; however, no segregation is observed in the crack portion. In contrast, in Sample 07 after heating, segregation of titanium is observed both on the surfaces of the particles and in the crack portion. That is, it is found that titanium is segregated at the surface of the crack portion by heating.

[0505]

Next, results of TEM-EDX plane analysis of magnesium are shown in FIGS. 39A, 39B1, 39B2, 39C1, 39C2, 39D1, 39D2, 39E1, and 39E2, and FIGS. 40A, 40B1, 40B2, 40C1, 40C2, 40D1, 40D2, 40E1, and 40E2.

[0506]

FIG. 39A is a cross-sectional TEM image of Sample 06, which is the same as FIG. 37A.

FIGS. 39B1, 39C1, 39D1, and 39E1 are HAADF-STEM images, which are the same as FIGS. 37B1, 37C1, 37D1, and 37E1. FIG. 39B2 shows a Mg mapping image of a region which is the same as FIG. 39B1. FIG. 39C2 shows a Mg mapping image of a region which is the same as FIG. 39C1. FIG. 39D2 shows a Mg mapping image of a region which is the same as FIG. 39D1. FIG. 39E2 shows a Mg mapping image of a region which is the same as FIG. 39E1.

[0507]

FIG. 40A is a cross-sectional TEM image of Sample 07, which is the same as FIG. 38A. FIGS. 40B1, 40C1, 40D1, and 40E1 are HAADF-STEM images, which are the same as FIGS. 38B1, 38C1, 38D1, and 38E1. FIG. 40B2 shows a Mg mapping image of a region which is the same as FIG. 40B1. FIG. 40C2 shows a Mg mapping image of a region which is the same as FIG. 40C1. FIG. 40D2 shows a Mg mapping image of a region which is the same as FIG. 40D1. FIG. 40E2 shows a Mg mapping image of a region which is the same as FIG. 40E1.

[0508]

As shown in FIGS. 39A, 39B1, 39B2, 39C1, 39C2, 39D1, 39D2, 39E1, and 39E2, and FIGS. 40A, 40B1, 40B2, 40C1, 40C2, 40D1, 40D2, 40E1, and 40E2, in Sample 06 before heating, segregation of magnesium is not observed on the surfaces of the particles or in the crack portion. In contrast, in Sample 07 after heating, segregation of magnesium is observed both on the surfaces of the particles and in the crack portion.

[0509]

Next, to quantify titanium and magnesium, EDX point analysis was performed on the regions indicated by circles marked with 1 to 6 in FIG. 37A and the regions indicated by circles marked with 1 to 6 in FIG. 38A. In each region, two points were measured.

[0510]

FIGS. 41A and 41B show results of EDX point analysis in an atomic ratio of titanium to cobalt. FIG. 41A shows results of Sample 06 before heating. Detection points 1 to 6 in FIG. 41A correspond to the regions indicated by circles marked with 1 to 6 in FIG. 37A. FIG. 41B shows results of Sample 07 after heating. Detection points 1 to 6 in FIG. 41B correspond to the regions indicated by circles marked with 1 to 6 in FIG. 38A.

[0511]

As shown in FIGS. 41A and 41B, in the crack portion of Sample 06, Ti/Co is less than or equal to 0.01 in each measurement point; in contrast, in the crack portion of Sample 07, the amount of titanium is increased in many points, and there are measurement points where Ti/Co is greater than or equal to 0.05. Furthermore, Ti/Co on the surfaces of the particles of Sample 07 is between 0.10 and 0.18.

[0512]

Next, FIGS. 42A and 42B show results of EDX point analysis in an atomic ratio of magnesium to cobalt. The detection points are the same as those in FIGS. 41A and 41B.

[0513]

As shown in FIGS. 42A and 42B, in Sample 06, Mg/Co is less than or equal to 0.03 both on the surfaces of the particles and in the crack portion; in contrast, in Sample 07, there are many points where the amount of magnesium is increased both on the surfaces of the particles and in the crack portion. Furthermore, Mg/Co on the surfaces of the particles is between 0.15 and 0.50, and that in the crack portion is between 0 and 0.22.

[0514]

Next, CR2032 coin-type secondary batteries were fabricated using the positive electrode active material of Sample 07 after heating. A positive electrode formed by applying slurry in which a positive electrode active material (LCO) of Sample 02, AB, and polyvinylidene fluoride (PVDF) were mixed at a weight ratio of 95:3:2 to a positive electrode current collector was used. As the positive electrode current collector, 20-μm-thick aluminum foil was used. The amount of positive electrode active material layer containing the positive electrode active material, AB, and PVDF was 7.6 mg/cm^2.

[0515]

A lithium metal was used for the counter electrode.

[0516]

An electrolyte solution formed in such a manner that 1 mol/L $LiPF_6$ was dissolved in a solution in which ethylene carbonate (EC) and diethyl carbonate (DEC) were mixed at a volume ratio of 3:7, and vinylene carbonate (VC) was added to the solution at a 2 wt% was used.

[0517]

[Initial characteristics, rate characteristics]

Initial characteristics and rate characteristics of the secondary battery using the positive electrode active material of Sample 07 formed in the above manner were measured.

[0518]

In the measurement of the initial characteristics, charging was performed at CCCV, 0.2 C, 4.6 V, and a cutoff current of 0.05 C, and discharging was performed at CC, 0.2 C, and a cutoff voltage of 3.0 V. Here, 1C was set to 160 mA/g, which was a current value per weight of the positive electrode active material. The measurement temperature was 25 °C. Table 2 shows measurement results of initial characteristics.

[0519]

[Table 2]

Initial charge capacity [mAh/g]	Initial discharge capacity [mAh/g]	Initial charge and discharge efficiency [%]
221.2	217.5	98.3

[0520]

The rate characteristics were measured after the initial characteristics were measured. The measurement was performed by changing a discharge rate in the following order: 0.2 C charge/0.2 C discharge; 0.2 C charge/0.5 C discharge; 0.2 C charge/1.0 C discharge; 0.2 C charge/2.0 C discharge; 0.2 C charge/3.0 C discharge; 0.2 C charge/4.0 C discharge; and 0.2 C charge/5.0 C discharge. Note that the conditions other than the discharge rate are the same as those of the measurement of the initial characteristics. The measurement temperature was 25 °C.

[0521]

Table 3 shows the measurement results of the initial characteristics and the rate characteristics. In addition, FIG. 43 shows discharge curves of the rates.

[0522]

[Table 3]

Rate [C]	Discharge capacity [mAh/g]	Discharge capacity retention rate [%]
0.2	218.2	100.0
0.5	215.8	98.9
1.0	213.1	97.7
2.0	207.9	95.3
3.0	204.0	93.5
4.0	198.8	91.1
5.0	184.3	84.5

[0523]

[Temperature characteristics]

Next, a cell was fabricated under conditions similar to those of a cell for evaluating the rate characteristics except that the amount of positive electrode active material layer was 8.2 mg/cm^2, and the temperature characteristics were measured. Charging was performed at 25 °C, CCCV, 0.2 C, 4.6 V, and a cutoff current of 0.05 C. Discharging was performed at 25 °C, 0 °C, −10 °C, −20 °C, and 45 °C in this order, CC, 0.2 C, and a cutoff voltage of 3.0 V. FIG. 44 shows measurement results of temperature characteristics.

[0524]

[Cycle characteristics]

Next, a cell was fabricated under conditions similar to those of the cell for measuring the temperature characteristics, and the cycle characteristics were measured. In the measurement of the cycle characteristics, charging was performed at CCCV, 1.0 C, 4.55 V, and a cutoff current of 0.05 C, and discharging was performed at CC, 1.0 C, and a cutoff voltage of 3.0 V. The measurement temperature of the cycle characteristics was 45 °C and 100 cycles were measured. The discharge capacity retention rate after 100 cycles was 86 %. FIG. 45 is a graph showing the discharge capacity retention rate of the measured cycle characteristics.

[0525]

From the measurement results, the specific surface area of the positive electrode active material of Sample 07 was 0.13 m^2/g.

[0526]

Furthermore, from the measurement results of particle size distribution of the positive electrode active material of Sample 07, the average particle diameter was 21.5 μm, 10 %D was 13.1 μm, 50 %D was 22.0 μm, and 90 %D was 34.4 μm.

[0527]

The tap density of the positive electrode active material of Sample 07 is 2.21 g/cm^3. The tap density was measured with MULTI TESTER MT-1000 (manufactured by SEISHIN ENTERPRISE Co., Ltd.).

[0528]

As described above, it is found that the positive electrode active material of Sample 07 which is one embodiment of the present invention shows excellent initial charge and discharge capacity, rate characteristics, and cycle characteristics. In particular, the initial charge and discharge capacity is high, which is 98 % or higher; thus, it is probable that a side reaction is inhibited. In addition, even at a high discharge rate of 2 C, an excellent capacity of 96.1 % is shown using 0.2 C as a reference.

[Example 3]

[0529]

In this example, a positive electrode active material including a region containing titanium and magnesium in a superficial portion was formed by changing the ratio of Li to the first transition metal of starting materials, and evaluation results of characteristics are shown.

[0530]

[Formation of positive electrode active material]

In this example, positive electrode active materials of Samples 11 to 17, Samples 21 to

28, and Samples 31 to 40 in which cobalt was used as the first transition metal were prepared. Formation methods and conditions of these samples are as follows.

[0531]

<<Samples 11 to 17>>

First, a source of lithium, a source of cobalt, a source of magnesium, and a source of fluorine, which are to be starting materials, were individually weighed. In this example, lithium carbonate, cobalt oxide, magnesium oxide, and lithium fluoride were used as the source of lithium, the source of cobalt, the source of magnesium, and the source of fluorine, respectively.

[0532]

At that time, the starting materials of Sample 11 were weighed so that the ratio of Li to Co was 1.00. The starting materials of Sample 12 were weighed so that the ratio of Li to Co was 1.03. The starting materials of Sample 13 were weighed so that the ratio of Li to Co was 1.05. The starting materials of Sample 14 were weighed so that the ratio of Li to Co was 1.06. The starting materials of Sample 15 were weighed so that the ratio of Li to Co was 1.07. The starting materials of Sample 16 were weighed so that the ratio of Li to Co was 1.08. The starting materials of Sample 17 were weighed so that the ratio of Li to Co was 1.13.

[0533]

In addition, the starting materials of each of Samples 11 to 17 were weighed so that, when the number of atoms of cobalt contained in the starting materials was set to 1, the number of atoms of magnesium was 0.01 and the number of atoms of fluorine was 0.02.

[0534]

Next, the weighed starting materials were separately mixed with a ball mill for each sample.

[0535]

Then, the mixed starting materials were baked. The baking was performed at 1000 °C for 10 hours under the following conditions: the temperature rising rate was 200 °C/h; and the flow rate of dry air was 10 L/min.

[0536]

Through the above process, particles of a composite oxide containing lithium, cobalt, fluorine, and magnesium were synthesized.

[0537]

Next, TTIP was added to 2-propanol so that the amount of TTIP per weight of the positive electrode active material was 0.01 ml/g and then mixing was performed, so that a 2-propanol solution of tetra-i-propoxy titanium was formed.

[0538]

To the 2-propanol solution of TTIP, the particles of a composite oxide containing lithium, cobalt, fluorine, and magnesium were added and then mixing was performed.

[0539]

The above-described mixed solution was stirred with a magnetic stirrer for four hours, at 25 °C, and at a humidity of 90 %RH. Through the process, water in an atmosphere and TTIP caused hydrolysis and polycondensation reaction, and a layer containing titanium was formed on the surface of the lithium cobalt oxide particle containing magnesium and fluorine.

[0540]

The mixed solution which had been subjected to the above process was filtered to collect the residue. As a filter for filtration, Kiriyama filter paper (No. 4) was used.

[0541]

The collected residue was dried in a vacuum at 70 °C for one hour.

[0542]

The dried particles were heated. The heating was performed in an oxygen atmosphere under the following conditions: the temperature was 800 °C (the temperature rising rate was 200 °C/h); and the retention time was two hours.

[0543]

The heated particles were cooled and subjected to crushing treatment. In the crushing treatment, the particles were made to pass through a sieve. The sieve has an aperture width of 53 μm.

[0544]

The particles which were subjected to crushing treatment were used as the positive electrode active materials of Samples 11 to 17.

[0545]

<<Samples 21 to 27>>

The starting materials of Samples 21 to 27 were the same as those of Samples 11 to 16. At that time, the starting materials of Sample 21 were weighed so that the ratio of Li to Co was 1.00. The starting materials of Sample 22 were weighed so that the ratio of Li to Co was 1.03. The starting materials of Sample 23 were weighed so that the ratio of Li to Co was 1.05. The starting materials of Sample 24 were weighed so that the ratio of Li to Co was 1.06. The starting materials of Sample 25 were weighed so that the ratio of Li to Co was 1.07. The starting materials of Sample 26 were weighed so that the ratio of Li to Co was 1.08. The starting materials of Sample 27 were weighed so that the ratio of Li to Co was 1.13.

[0546]

Samples 21 to 27 were formed in manners similar to those of Samples 11 to 17 except

that the concentration of TTIP in the 2-propanol solution was adjusted so that the amount of TTIP per weight of the positive electrode active material was 0.02 ml/g.

[0547]

<<Sample 28>>

The ratio of Li to Co of the starting materials and the amount of TTIP of Sample 28 were the same as the ratio of Li to Co of the starting materials and the amount of TTIP of Sample 23. That is, in Sample 28, the starting materials were weighed so that the ratio of Li to Co was 1.05, and the amount of TTIP per weight of the positive electrode active material was 0.02 ml/g.

[0548]

Note that in Sample 28, after the starting materials were mixed, baking was performed at 950 °C.

[0549]

Sample 28 was formed in a manner similar to that of Sample 23 except for the baking temperature.

[0550]

It is probable that Samples 11 to 17 and Samples 21 to 28 are each a positive electrode active material which contains lithium cobaltate inside and includes a region containing titanium and magnesium in a superficial portion.

[0551]

<<Samples 31 to 40>>

Samples 31 to 40 were formed as comparative examples, each of which did not include a region containing titanium.

[0552]

The starting materials of Sample 31 were weighed so that the ratio of Li to Co was 1.00. The starting materials of Sample 32 were weighed so that the ratio of Li to Co was 1.01. The starting materials of Sample 33 were weighed so that the ratio of Li to Co was 1.02. The starting materials of Sample 34 were weighed so that the ratio of Li to Co was 1.03. The starting materials of Sample 35 were weighed so that the ratio of Li to Co was 1.035. The starting materials of Sample 36 were weighed so that the ratio of Li to Co was 1.04. The starting materials of Sample 37 were weighed so that the ratio of Li to Co was 1.051. The starting materials of Sample 38 were weighed so that the ratio of Li to Co was 1.061. The starting materials of Sample 39 were weighed so that the ratio of Li to Co was 1.081. The starting materials of Sample 40 were weighed so that the ratio of Li to Co was 1.130.

[0553]

In addition, the starting materials of each of Samples 31 to 40 were weighed so that, when

the number of atoms of cobalt contained in the starting materials was set to 1, the number of atoms of magnesium was 0.01 and the number of atoms of fluorine was 0.02.

[0554]

Next, the weighed starting materials were separately mixed with a ball mill for each sample.

[0555]

Then, the mixed starting materials were baked. The baking was performed at 1000 °C for 10 hours under the following conditions: the temperature rising rate was 200 °C/h; and the flow rate of dry air was 10 L/min.

[0556]

Through the above process, particles of a composite oxide containing lithium, cobalt, fluorine, and magnesium were synthesized.

[0557]

The synthesized particles were cooled and then heated. The heating was performed in an oxygen atmosphere under the following conditions: the temperature was 800 °C (the temperature rising rate was 200 °C/h); and the retention time was two hours.

[0558]

The heated particles were cooled and subjected to crushing treatment. In the crushing treatment, the particles were made to pass through a sieve. The sieve has an aperture width of 53 μm.

[0559]

The particles which were subjected to crushing treatment were used as the positive electrode active materials of Samples 31 to 40.

[0560]

Table 4 shows the formation conditions of Samples 11 to 17, Samples 21 to 28, and Samples 31 to 40.

[0561]

[Table 4]

	Li/Co	TTIP	Baking temperature
Sample 11	1.00	0.01 ml/g	1000 °C
Sample 12	1.03		
Sample 13	1.05		
Sample 14	1.06		
Sample 15	1.07		
Sample 16	1.08		
Sample 17	1.13		
Sample 21	1.00	0.02 ml/g	1000 °C
Sample 22	1.03		
Sample 23	1.05		
Sample 24	1.06		
Sample 25	1.07		
Sample 26	1.08		
Sample 27	1.13		
Sample 28	1.05	0.02 ml/g	950 °C
Sample 31	1.00	-	1000 °C
Sample 32	1.01		
Sample 33	1.02		
Sample 34	1.03		
Sample 35	1.035		
Sample 36	1.04		
Sample 37	1.051		
Sample 38	1.061		
Sample 39	1.081		
Sample 40	1.130		

[0562]

[XPS]

The positive electrode active materials of Samples 11 to 17, Samples 21 to 28, and Samples 31 to 40 were subjected to an XPS analysis. Table 5 shows results of the XPS analysis of Samples 11 to 17, Table 6 shows results of the XPS analysis of Samples 21 to 28, and Table 7 shows results of the XPS analysis of Samples 31 to 40. Tables 5 to 7 show a relative value of the concentration of each element under the condition where the concentration of cobalt is 1.

[0563]

[Table 5]

		Relative value under condition where concentration of Co is 1										
	Li/Co	Li	Co	O	C	F	Mg	Ti	Si	Ca	Na	S
Sample 11	1.00	0.82	1.00	2.87	0.25	0.37	0.57	0.15	0.00	0.04	0.27	0.01
Sample 12	1.03	0.69	1.00	2.84	0.93	0.63	0.63	0.14	0.00	0.05	0.04	0.00
Sample 13	1.05	0.96	1.00	3.22	0.66	0.47	0.61	0.14	0.00	0.03	0.09	0.03
Sample 14	1.06	1.04	1.00	3.13	0.34	0.77	0.77	0.15	0.00	0.05	0.34	0.04
Sample 15	1.07	0.84	1.00	3.42	0.58	0.60	0.23	0.18	0.00	0.08	0.27	0.00
Sample 16	1.08	1.08	1.00	3.10	0.66	0.59	0.04	0.16	0.00	0.03	0.16	0.02
Sample 17	1.13	1.29	1.00	3.70	0.83	0.87	0.00	0.19	0.00	0.04	0.20	0.02

[0564]

[Table 6]

		Relative value under condition where concentration of Co is 1										
	Li/Co	Li	Co	O	C	F	Mg	Ti	Si	Ca	Na	S
Sample 21	1.00	1.08	1.00	3.23	0.47	0.45	0.49	0.20	0.00	0.06	0.35	0.01
Sample 22	1.03	0.73	1.00	3.05	1.06	0.70	0.59	0.17	0.00	0.03	0.09	0.00
Sample 23	1.05	1.19	1.00	3.40	0.73	0.48	0.58	0.23	0.00	0.05	0.10	0.04
Sample 24	1.06	1.10	1.00	3.44	0.33	0.92	0.91	0.25	0.00	0.05	0.30	0.04
Sample 25	1.07	1.05	1.00	4.26	0.74	0.20	0.25	0.29	0.00	0.07	0.30	0.02
Sample 26	1.08	1.12	1.00	3.32	0.74	0.52	0.14	0.21	0.00	0.05	0.15	0.00
Sample 27	1.13	1.49	1.00	3.80	0.60	0.41	0.00	0.12	0.00	0.00	0.16	0.02
Sample 28	1.05	0.99	1.00	3.11	0.72	0.86	0.56	0.18	0.00	0.05	0.12	0.02

[0565]

[Table 7]

		Relative value under condition where concentration of Co is 1							
	Li/Co	Li	Co	O	C	F	Mg	Ca	Na
Sample 31	1.00	0.51	1.00	2.45	0.69	0.08	0.27	0.03	0.08
Sample 32	1.01	0.67	1.00	2.65	0.77	0.08	0.28	0.03	0.08
Sample 33	1.02	0.53	1.00	2.51	0.66	0.09	0.27	0.02	0.06
Sample 34	1.03	0.79	1.00	2.93	0.92	0.09	0.35	0.04	0.14
Sample 35	1.04	0.65	1.00	2.33	0.48	0.11	0.32	0.03	0.11
Sample 36	1.04	0.69	1.00	2.73	0.56	0.11	0.38	0.05	0.16
Sample 37	1.05	0.67	1.00	3.04	0.64	0.09	0.35	0.04	0.21
Sample 38	1.06	0.83	1.00	2.65	1.03	0.29	0.10	0.05	0.11
Sample 39	1.08	0.80	1.00	2.79	1.04	0.26	0.03	0.08	0.10
Sample 40	1.13	0.77	1.00	2.72	0.22	0.99	0.00	0.01	0.26

[0566]

FIGS. 46A and 46B are graphs in which the relative valueof magnesium and the relative valueof titanium are extracted from the analysis results of Tables 5 to 7. FIG. 46A is a graph showing the ratio of Li to Co and the relative valueof magnesium. FIG. 46B is a graph showing

the ratio of Li to Co and the relative valueof titanium.

[0567]

From the analysis results of Samples 31 to 40 in FIG. 46A, it is found that, in the case where a coating layer containing titanium is not included, the concentration of magnesium is high in samples where the ratio of Li to Co is greater than or equal to 1.00 and less than or equal to 1.05. This is probably because magnesium contained in the starting materials is segregated in a range where the element concentration can be detected by XPS by heating. In contrast, in samples where the ratio of Li to Co is greater than or equal to 1.06, the concentration of magnesium is low; thus, it is probable that the segregation of magnesium does not easily occur when the amount of lithium is too large.

[0568]

From the analysis results of Samples 11 to 16 and Samples 21 to 26 in FIG. 46A, it is found that the concentration of magnesium in a range where the element concentration can be detected by XPS is higher in the case where a region containing titanium is included in a superficial portion than in the case where the region containing titanium is not included.

[0569]

Moreover, in the case where the ratio of Li to Co is 1.06, in samples where the region containing titanium is not included, the concentration of magnesium in a range where the element concentration can be detected by XPS is low; in contrast, in samples where the region containing titanium is included, the concentration of magnesium in a range where the element concentration can be detected by XPS is high. That is, when the region containing titanium is formed in the superficial portion, magnesium is sufficiently segregated even in the case where the ratio of Li to Co is high.

[0570]

Note that even if the region containing titanium is included, the concentration of magnesium is lower in the case where the ratio of Li to Co is 1.07 than in the case where the ratio of Li to Co is 1.06. Furthermore, it is probable that, in the case where the ratio of Li to Co is greater than or equal to 1.08, the segregation of magnesium does not easily occur even if the region containing titanium is included.

[0571]

[Evaluation of cycle characteristics]

<<Energy density retention rate>>

Next, cycle characteristics were evaluated in a manner similar to that of Example 1 using positive electrode active materials of Samples 11 to 14, Sample 16, Samples 21 to 24, and Sample 26.

[0572]

The shape of the secondary battery, the materials and the mixture ratios of the positive electrode active material, the conductive additive, and the binder in the positive electrode, the counter electrode, the electrolyte solution, the exterior body, the conditions of the cycle characteristics test, and the like are the same as those in Example 1.

[0573]

FIG. 47A is a graph showing the energy density retention rates and the number of charge and discharge cycles at the time of charging at 4.6 V of secondary batteries using the positive electrode active materials of Samples 11 to 14 and Sample 16 which were formed so that the amount of TTIP per weight of the positive electrode active material was 0.01 ml/g. FIG. 47B is a graph showing the energy density retention rates and the number of charge and discharge cycles at the time of charging at 4.6 V of secondary batteries using the positive electrode active materials of Samples 21 to 24 and Sample 26 which were formed so that the amount of TTIP per weight of the positive electrode active material was 0.02 ml/g.

[0574]

As shown in FIG. 47A, in the case where TTIP is 0.01 ml/g, Samples 11 to 14, that is, the positive electrode active materials in which the ratio of Li to Co is greater than or equal to 1.00 and less than or equal to 1.06 have excellent cycle characteristics. In particular, Samples 11 and 12, that is, the positive electrode active materials in which the ratio of Li to Co is greater than or equal to 1.00 and less than or equal to 1.03 have extremely excellent cycle characteristics. In contrast, in Sample 16 in which the ratio of Li to Co is 1.08, the energy density retention rate decreases at a relatively early stage.

[0575]

As shown in FIG. 47B, in the case where TTIP is 0.02 ml/g, Samples 21 to 24, that is, the positive electrode active materials in which the ratio of Li to Co is greater than or equal to 1.00 and less than or equal to 1.06 have excellent cycle characteristics. In particular, Samples 23 and 24, that is, the positive electrode active materials in which the ratio of Li to Co is greater than or equal to 1.05 and less than or equal to 1.06 have extremely excellent cycle characteristics.

[0576]

FIG. 48 is a graph showing comparison between Sample 11 having the most excellent cycle characteristics in Samples 11 to 15 and Sample 23 having the most excellent cycle characteristics in Samples 21 to 25.

[0577]

As shown in FIG. 48, both Sample 11 and Sample 23 have excellent cycle characteristics; however, Sample 23 in which TTIP is 0.02 ml/g has more excellent cycle characteristics.

[0578]

<<Discharge capacity retention rate>>

Next, FIG. 49 shows evaluation results of a discharge capacity retention rate, which is one of the cycle characteristics of each of Samples 21 to 26 and Sample 28.

[0579]

The shape of the secondary battery, the material and the mixture ratio of the positive electrode active material, the conductive additive, and the binder in the positive electrode, the counter electrode, the electrolyte solution, the exterior body, the conditions of the cycle characteristics test, and the like of Samples 21 to 26 are the same as those in Example 1.

[0580]

The secondary battery using Sample 28 was formed in a manner similar to those of the secondary batteries using Samples 21 to 26 except that PVDF was used as a binder and the positive electrode active material (LCO), AB, and PVDF were mixed such that the weight ratio of LCO to AB and PVDF was 95:3:2, and evaluated.

[0581]

As shown in FIG. 49, Samples 21 to 24 and Sample 28 have excellent cycle characteristics. In particular, Sample 28 has extremely excellent cycle characteristics. In Sample 28, the discharge capacity retention rate after 50 cycles was higher than or equal to 85 %.

[0582]

In contrast, in Sample 25 and Sample 26 in which the ratios of Li to Co are 1.07 and 1.08, respectively, the discharge capacity retention rates decrease at a relatively early stage.

[0583]

From the above results, it is found that in the case where TTIP per weight of the positive electrode active material is 0.02 ml/g, the ratio of Li to Co preferably has a range of greater than or equal to 1.00 and less than 1.07. Moreover, it is found that a sample in which the ratio of Li to Co has a range of greater than or equal to 1.05 and less than or equal to 1.06 has extremely excellent cycle characteristics.

[0584]

FIGS. 50A to 50C show charge and discharge curves of the secondary batteries using Sample 28, which has extremely excellent cycle characteristics in FIG. 49, Sample 24, and Sample 25, in which the degradation occurs at a relatively early stage.

[0585]

FIG. 50A, FIG. 50B, and FIG. 50C show charge and discharge curves of the secondary batteries using Sample 28, Sample 24, and Sample 25, respectively. Each of the figures shows overlap of results of 50 cycles of charge and discharge. As indicated by an arrow in each figure,

the charge and discharge capacity decreases from the first cycle to the fiftieth cycle.

[0586]

As shown in FIGS. 50A and 50B, Sample 28 and Sample 24, which are positive electrode active materials of one embodiment of the present invention, have high charge and discharge capacity and excellent charge and discharge characteristics. In addition, it is found that a decrease in charge and discharge capacity of each of Sample 28 and Sample 24 in FIGS. 50A and 50B is significantly suppressed as compared with Sample 25 in FIG. 50C.

[Example 4]

[0587]

In this example, SEM observation results and SEM-EDX analysis results of the positive electrode active material of Sample 24 formed in Example 2 are described.

[0588]

Sample 24 was formed so that Li/Co was 1.06 and TTIP per weight of the positive electrode active material was 0.02 ml/g. FIG. 51A shows a SEM image of Sample 24. FIGS. 51B and 51C each show an enlarged image of a part in FIG. 51A.

[0589]

As shown in FIGS. 51A to 51C, there are a large number of projected regions in a superficial portion of the positive electrode active material.

[0590]

Next, FIGS. 52A-1, 52A-2, 52B-1, 52B-2, 52C-1, and 52C-2 show analysis results of the positive electrode active material of Sample 24 using SEM-EDX. FIG. 52A-1 shows a SEM image of a superficial portion of the positive electrode active material, FIG. 52A-2 shows a mapping image of titanium, FIG. 52B-1 shows a mapping image of magnesium, FIG. 52B-2 shows a mapping image of oxygen, FIG. 52C-1 shows a mapping image of aluminum, and FIG. 52C-2 shows a mapping image of cobalt. Note that in EDX element mapping images in FIGS. 52A-2, 52B-1, 52B-2, 52C-1, and 52C-2, a region where the number of elements is less than or equal to a lower limit of the detection is indicated in black, and as the number of elements is increased, the black region becomes white.

[0591]

The same regions in FIGS. 52A-1, 52A-2, and 52B-1 are surrounded by dotted lines. When the regions surrounded by dotted lines are compared with each other, it is found that in the projected regions in the superficial portion of the positive electrode active material, titanium and magnesium are distributed.

[0592]

Thus, it is found that Sample 24 is a positive electrode active material including the

projected fourth region 104 containing titanium and magnesium over the third region 103.

[0593]

As shown in Example 2, Sample 24 is one of the samples which show extremely excellent cycle characteristics. Thus, it is found that, even if the fourth region is provided in the superficial portion or when the fourth region is provided, the positive electrode active material with excellent cycle characteristics can be obtained.

[0594]

From the above results of Examples 1 to 3, it is found that when the region containing titanium is formed in the superficial portion, the positive electrode active material with excellent cycle characteristics can be obtained. In addition, it is found that, when the ratio of Li to Co is increased to increase the particle diameter of the positive electrode active material, the cycle characteristics might be degraded; however, the region containing titanium is formed in the superficial portion, whereby the range of the ratio of Li to Co in which excellent cycle characteristics is obtained can be widened. Furthermore, it is found that even when the fourth region containing titanium and magnesium is provided in the superficial portion of the positive electrode active material, excellent cycle characteristics is obtained.

[Example 5]

[0595]

In this example, an example of a method for producing a positive electrode active material coated with graphene oxide is shown, and observation results of the positive electrode active material produced by the method with an electron microscope are described.

[0596]

As shown in a process flow chart in FIG. 53, a process for forming a coating film on a positive electrode active material includes the steps of: weighing graphene oxide (S11); mixing and stirring the graphene oxide and pure water (S12); controlling pH (S13); adding an active material (S14); completing a suspension (S15); spraying the suspension using a spray dry apparatus (S16); and collecting particles in a container (S17).

[0597]

Note that in (S12), pure water is used as a dispersion medium; however, a dispersion medium is not particularly limited and ethanol or the like may be used. In addition, in (S14), the active material is a positive electrode active material.

[0598]

FIG. 54 is a schematic view of a spray dry apparatus 280. The spray dry apparatus 280 includes a chamber 281 and a nozzle 282. A suspension 284 is supplied to the nozzle 282 through a tube 283. The suspension 284 is supplied from the nozzle 282 to the chamber 281 in the form

of mist and dried in the chamber 281. The nozzle 282 may be heated with a heater 285. Here, a region of the chamber 281 which is close to the nozzle 282, for example, a region surrounded by dashed-two dotted line in FIG. 54, is also heated with the heater 285.

[0599]

In the case of using a suspension containing a positive electrode active material and graphene oxide as the suspension 284, powder of the positive electrode active material coated with the graphene oxide is collected in a collection container 286 through the chamber 281.

[0600]

The air in the chamber 281 may be suctioned by an aspirator or the like through a path indicated by an arrow 288.

[0601]

An example of conditions for forming the coating film is shown below.

[0602]

First, a suspension was formed by dispersing graphene oxide into a solvent.

[0603]

Although in pure water, graphene oxide is highly dispersible, pure water might react with an active material added later, so that Li might be dissolved or an active material might be damaged to change the surface structure. Thus, the graphene oxide was dispersed into a solution such that the ratio between ethanol and pure water was 4:6.

[0604]

Stirring to disperse the graphene oxide into a solution was performed under the following conditions: a stirrer and an ultrasonic wave generator were used; a rotation rate was 750 rpm; and irradiation time with ultrasonic waves was 2 minutes.

[0605]

Then, a LiOH aqueous solution was dropped to adjust pH to be pH7 (25 °C).

[0606]

The positive electrode active materials (in this example, lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-20F)) were added, and stirring was performed using a stirrer and an ultrasonic wave generator under the following conditions: a rotation rate was 750 rpm; and irradiation time with ultrasonic waves was 1 minute. Through the above process, the suspension was prepared. The above lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-20F) contain at least fluorine, magnesium, calcium, sodium, silicon, sulfur, and phosphorus, and each have a diameter of approximately 20 μm.

[0607]

Next, the suspension was sprayed uniformly with a spray nozzle (having a nozzle diameter of 20 μm) of the spray dry apparatus to obtain powder. The inlet temperature was 160 °C and the outlet temperature was 40 °C as the hot-air temperature of the spray dry apparatus, and the N_2 gas flow rate was 10 L/min.

[0608]

FIG. 55 shows a cross-sectional TEM image of the obtained powder. In addition, FIG. 56 shows a SEM image of the obtained powder. When a positive electrode active material which was the same as the sprayed positive electrode active material (C-20F, produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD.) was mixed with the graphene oxide with a planetary centrifugal mixer as a comparative example, coating was insufficient. FIG. 57 shows a SEM image of the comparative example.

[0609]

It is found that, as compared with FIG. 57, the coating film is uniformly formed on the surface of the powder in FIG. 56.

[0610]

FIGS. 58A and 58B illustrate a cross-sectional structure example of an active material layer 200 which is coated with graphene oxide with a spray dry apparatus and includes a graphene compound as a conductive additive.

[0611]

FIG. 58A is a longitudinal sectional view of the active material layer 200. The active material layer 200 includes positive electrode active material particles 100 coated with graphene oxide, a graphene compound 201 as a conductive additive, and a binder (not illustrated). Here, graphene or multilayer graphene may be used as the graphene compound 201, for example. The graphene compound 201 preferably has a sheet-like shape. The graphene compound 201 may have a sheet-like shape formed of a plurality of sheets of multilayer graphene and/or a plurality of sheets of graphene that partly overlap with each other.

[0612]

In the longitudinal section of the active material layer 200, as illustrated in FIG. 58B, the positive electrode active material 100 coated with a coating film 105 formed of graphene oxide is in contact with the graphene compound 201. A plurality of graphene compounds 201 are formed in such a way as to be partly in contact with the positive electrode active material 100 coated with the coating film 105 and adhere to the coating film 105 of the adjacent positive electrode active material 100, so that the graphene compounds 201 are in contact with each of the positive electrode active material 100.

[0613]

The graphene compound 201 and the coating film 105 are formed using carbon-based materials; thus, an excellent conductive path can be formed.

[0614]

The coating film 105 is effective in protecting the crystal structure of the positive electrode active material 100 so as not to be in contact with the electrolyte solution and in forming the excellent conductive path.

REFERENCE NUMERALS

[0615]

11a: positive electrode, 11b: negative electrode, 12a: lead, 12b: lead, 14: separator, 15a: bonding portion, 15b: bonding portion, 17: fixing member, 50: secondary battery, 51: exterior body, 61: folded portion, 62: seal portion, 63: seal portion, 71: crest line, 72: trough line, 73: space, 100: positive electrode active material, 101: first region, 101p: crystal plane, 102: second region, 102p: crystal plane, 103: third region, 103p: crystal plane, 104: fourth region, 105: coating film, 106: crack portion, 110: particle, 111: region, 112: layer containing titanium, 114: cobalt oxide layer, 120: particle, 121: region, 122: layer containing titanium, 124: cobalt oxide layer, 125: layer containing lithium titanate, 200: active material layer, 201: graphene compound, 214: separator, 280: spray dry apparatus, 281: chamber, 282: nozzle, 283: tube, 284: suspension, 285: heater, 286: collection container, 288: arrow, 300: coin-type secondary battery, 301: positive electrode can, 302: negative electrode can, 303: gasket, 304: positive electrode, 305: positive electrode current collector, 306: positive electrode active material layer, 307: negative electrode, 308: negative electrode current collector, 309: negative electrode active material layer, 310: separator, 500: laminated secondary battery, 501: positive electrode current collector, 502: positive electrode active material layer, 503: positive electrode, 504: negative electrode current collector, 505: negative electrode active material layer, 506: negative electrode, 507: separator, 508: electrolyte solution, 509: exterior body, 510: positive electrode lead electrode, 511: negative electrode lead electrode, 600: cylindrical secondary battery, 601: positive electrode cap, 602: battery can, 603: positive electrode terminal, 604: positive electrode, 605: separator, 606: negative electrode, 607: negative electrode terminal, 608: insulating plate, 609: insulating plate, 611: PTC element, 612: safety valve mechanism, 613: conductive plate, 614: conductive plate, 615: module, 616: wiring, 617: temperature control device, 900: circuit board, 910: label, 911: terminal, 912: circuit, 913: secondary battery, 914: antenna, 915: antenna, 916: layer, 917: layer, 918: antenna, 920: display device, 921: sensor, 922: terminal, 930: housing, 930a: housing, 930b: housing, 931: negative electrode, 932: positive electrode, 933: separator, 950: wound body, 951: terminal, 952: terminal, 980: laminated secondary battery, 981: film, 982: film, 993: wound body, 994: negative electrode,

995: positive electrode, 996: separator, 997: lead electrode, 998: lead electrode, 7100: portable display device, 7101: housing, 7102: display portion, 7103: operation button, 7104: secondary battery, 7200: portable information terminal, 7201: housing, 7202: display portion, 7203: band, 7204: buckle, 7205: operation button, 7206: input output terminal, 7207: icon, 7300: display device, 7304: display portion, 7400: mobile phone, 7401: housing, 7402: display portion, 7403: operation button, 7404: external connection port, 7405: speaker, 7406: microphone, 7407: secondary battery, 7408: lead electrode, 7409: current collector, 7500: vaporizer, 7501: atomizer, 7502: cartridge, 7504: secondary battery, 8000: display device, 8001: housing, 8002: display portion, 8003: speaker portion, 8004: secondary battery, 8021: ground-based charging apparatus, 8022: cable, 8024: secondary battery, 8100: lighting device, 8101: housing, 8102: light source, 8103: secondary battery, 8104: ceiling, 8105: wall, 8106: floor, 8107: window, 8200: indoor unit, 8201: housing, 8202: air outlet, 8203: secondary battery, 8204: outdoor unit, 8300: electric refrigerator-freezer, 8301: housing, 8302: refrigerator door, 8303: freezer door, 8304: secondary battery, 8400: automobile, 8401: headlight, 8406: electric motor, 8500: automobile, 8600: motor scooter, 8601: side mirror, 8602: secondary battery, 8603: indicator, 8604: storage unit under seat, 9600: tablet terminal, 9625: power saving mode changing switch, 9626: display mode changing switch, 9627: power switch, 9628: operation switch, 9629: fastener, 9630: housing, 9630a: housing, 9630b: housing, 9631: display portion, 9631a: display portion, 9631b: display portion, ~~9632a: region, 9632b: region,~~ 9633: solar cell, 9634: charge and discharge control circuit, 9635: power storage unit, 9636: DCDC converter, 9637: converter, ~~9638: operation key, 9639: button,~~ 9640: movable portion.

This application is based on Japanese Patent Application Serial No. 2016-133997 filed with Japan Patent Office on July 6, 2016, Japanese Patent Application Serial No. 2016-133143 filed with Japan Patent Office on July 5, 2016, Japanese Patent Application Serial No. 2017-002831 filed with Japan Patent Office on January 11, 2017, Japanese Patent Application Serial No. 2017-030693 filed with Japan Patent Office on February 22, 2017, Japanese Patent Application Serial No. 2017-084321 filed with Japan Patent Office on April 21, 2017,and Japanese Patent Application Serial No. 2017-119272 filed with Japan Patent Office on June 19, 2017 the entire contents of which are hereby incorporated by reference.

ABSTRACT

A positive electrode active material which can improve cycle characteristics of a secondary battery is provided. Two kinds of regions are provided in a superficial portion of a positive electrode active material such as lithium cobaltate which has a layered rock-salt crystal structure. The inner region is a non-stoichiometric compound containing a transition metal such as titanium, and the outer region is a compound of representative elements such as magnesium oxide. The two kinds of regions each have a rock-salt crystal structure. The inner layered rock-salt crystal structure and the two kinds of regions in the superficial portion are topotaxy; thus, a change of the crystal structure of the positive electrode active material generated by charging and discharging can be effectively suppressed. In addition, since the outer coating layer in contact with an electrolyte solution is the compound of representative elements which is chemically stable, the secondary battery having excellent cycle characteristics can be obtained.

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DESCRIPTION

POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE

TECHNICAL FIELD

[0001]

One embodiment of the present invention relates to an object, a method, or a manufacturing method. The present invention relates to a process, a machine, manufacture, or a composition of matter. One embodiment of the present invention relates to a semiconductor device, a display device, a light-emitting device, a power storage device, a lighting device, an electronic device, or a manufacturing method thereof. In particular, one embodiment of the present invention relates to a positive electrode active material that can be used in a secondary battery, a secondary battery, and an electronic device including a secondary battery.

[0002]

In this specification, the power storage device is a collective term describing elements and devices having a power storage function. For example, a storage battery such as a lithium-ion secondary battery (also referred to as secondary battery), a lithium-ion capacitor, and an electric double layer capacitor are included in the category of the power storage device.

[0003]

Note that electronic devices in this specification mean all devices including power storage devices, and electro-optical devices including power storage devices, information terminal devices including power storage devices, and the like are all electronic devices.

BACKGROUND ART

[0004]

In recent years, a variety of power storage devices such as lithium-ion secondary batteries, lithium-ion capacitors, and air batteries have been actively developed. In particular, demand for lithium-ion secondary batteries with high output and high energy density has rapidly grown with the development of the semiconductor industry, for portable information terminals such as mobile phones, smartphones, tablets, and laptop computers; portable music players; digital cameras; medical equipment; next-generation clean energy vehicles such as hybrid electric vehicles (HEV), electric vehicles (EV), and plug-in hybrid electric vehicles (PHEV); and the like. The lithium-ion secondary batteries are essential as rechargeable energy supply sources for today's information society.

[0005]

The performance required for lithium-ion secondary batteries includes increased energy density, improved cycle performance, safe operation under a variety of environments, and longer-term reliability.

[0006]

Thus, improvement of a positive electrode active material has been studied to increase the cycle performance and the capacity of the lithium ion secondary battery (Patent Document 1 and Patent Document 2).

[Reference]

[Patent Documents]

[0007]

[Patent Document 1] Japanese Published Patent Application No. 2012-018914

[Patent Document 2] Japanese Published Patent Application No. 2015-201432

DISCLOSURE OF INVENTION

[0008]

Development of lithium ion secondary batteries and positive electrode active materials used therein is susceptible to improvement in terms of charge and discharge characteristics, cycle characteristics, reliability, safety, cost, and the like.

[0009]

An object of one embodiment of the present invention is to provide a positive electrode active material which suppresses a reduction in capacity due to charge and discharge cycles when used in a lithium ion secondary battery. Another object of one embodiment of the present invention is to provide a high-capacity secondary battery. Another object of one embodiment of the present invention is to provide a secondary battery with excellent charge and discharge characteristics. Another object of one embodiment of the present invention is to provide a highly safe or highly reliable secondary battery.

[0010]

Another object of one embodiment of the present invention is to provide a novel material, a novel active material particle, a novel secondary battery, or a formation method thereof.

[0011]

Note that the descriptions of these objects do not disturb the existence of other objects. In one embodiment of the present invention, there is no need to achieve all the objects. Other objects can be derived from the description of the specification, the drawings, and the claims.

[0012]

To achieve the above objects, in one embodiment of the present invention, two kinds of regions which are different from a region inside the positive electrode active material are provided in a superficial portion of the positive electrode active material. It is preferable that the inner region contain a non-stoichiometric compound and the outer region contain a stoichiometric compound.

[0013]

In addition, it is preferable that the inner region contain titanium and the outer region contain magnesium. Furthermore, these two kinds of regions may overlap.

[0014]

In addition, it is preferable that the inner region be formed through a coating process such as a sol-gel method and the outer region be formed by segregation due to heating.

[0015]

One embodiment of the present invention is a positive electrode active material including a first region, a second region, and a third region. The first region is present inside the positive electrode active material. The second region and the third region are present in a superficial portion of the positive electrode active material. The third region is present in a region closer to a surface of the positive electrode active material than the second region is. The first region contains an oxide of lithium and a first transition metal and has a layered rock-salt crystal structure. The second region contains a non-stoichiometric compound containing an oxide of a second transition metal and the non-stoichiometric compound has a rock-salt crystal structure. The third region contains a compound of representative elements and the compound of representative elements has a rock-salt crystal structure.

[0016]

In the above structure, it is preferable that the first transition metal be cobalt, the second transition metal be titanium, and the compound of representative elements be magnesium oxide.

[0017]

In the above structure, the third region may contain fluorine. Furthermore, the second region and the third region may each contain cobalt.

[0018]

In the above structure, it is preferable that crystal orientations of the first region and the second region be partly aligned with each other and crystal orientations of the second region and the third region be partly aligned with each other.

[0019]

In the above structure, a degree of a mismatch between a (1−1−4) plane of the layered rock-salt crystal structure in the first region or a plane orthogonal to the (1−1−4) plane and a {100}

plane of the rock-salt crystal structure in the second region is preferably less than or equal to 0.12, and a degree of a mismatch between the {100} plane of the rock-salt crystal structure in the second region and a {100} plane of the rock-salt crystal structure in the third region is preferably less than or equal to 0.12.

[0020]

Another embodiment of the present invention is a positive electrode active material containing lithium, titanium, cobalt, magnesium, oxygen, and fluorine. When the concentration of cobalt which is present in a superficial portion of the positive electrode active material and is measured by X-ray photoelectron spectroscopy is 1, the concentration of titanium is greater than or equal to 0.05 and less than or equal to 0.4, the concentration of magnesium is greater than or equal to 0.4 and less than or equal to 1.5, and the concentration of fluorine is greater than or equal to 0.05 and less than or equal to 1.5.

[0021]

Another embodiment of the present invention is a method for forming a positive electrode active material including: a step of mixing a source of lithium, a source of cobalt, a source of magnesium, and a source of fluorine; a step of heating the mixture of the source of lithium, the source of cobalt, the source of magnesium, and the source of fluorine at 800 °C or higher and 1100 °C or lower for 2 hours or longer and 20 hours or shorter to obtain particles containing lithium, cobalt, magnesium, oxygen, and fluorine; a step of dissolving titanium alkoxide into alcohol; a step of mixing the particles containing lithium, cobalt, magnesium, oxygen, and fluorine into the alcohol solution of the titanium alkoxide and stirring the mixed solution in an atmosphere containing water vapor; a step of collecting precipitate from the mixed solution; and a step of heating the collected precipitate at 500°C or higher and 1200 °C or lower in an atmosphere containing oxygen under a condition where a retention time is 50 hours or shorter.

[0022]

In the above formation method, a ratio of the number of atoms of lithium in the source of lithium to the number of atoms of cobalt in the source of cobalt is preferably greater than or equal to 1.00 and less than 1.07.

[0023]

In the above formation method, a ratio between the number of atoms of magnesium in the source of magnesium and the number of atoms of fluorine in the source of fluorine is preferably Mg:F = 1:x ($1.5 \leq x \leq 4$).

[0024]

In the above formation method, the number of atoms of magnesium in the source of magnesium is preferably greater than or equal to 0.5 atomic% and less than or equal to 1.5 atomic%

of the number of atoms of cobalt in the source of cobalt.

[0025]

In the above formation method, lithium carbonate, cobalt oxide, magnesium oxide, and lithium fluoride can be used as a source of lithium, a source of cobalt, a source of magnesium, and a source of fluorine, respectively.

[0026]

When the surface of the positive electrode active material is covered with a coating film to protect the above crystal structure, a decrease in capacity due to charge and discharge cycles can be suppressed. As the coating film covering the surface of the positive electrode active material, a coating film containing carbon (a film containing a graphene compound) or a coating film containing lithium or a decomposition product of an electrolyte solution is used.

[0027]

In particular, powder in which the surface of the positive electrode active material is coated with graphene oxide using a spray dry apparatus is preferably obtained. The spray dry apparatus is a manufacturing apparatus using a spray dry method by which a dispersion medium is removed by supplying a hot wind to a suspension.

[0028]

When charge and discharge cycles are repeated, deformation of the particles of the positive electrode active materials, such as cracking or breaking, might occur. It is said that such deformation makes a new surface of the positive electrode active material exposed, and the surface is in contact with an electrolyte solution to cause a decomposition reaction or the like, so that the cycle characteristics and the charge and discharge characteristics of the secondary battery are degraded.

[0029]

Thus, a coating film is preferably provided to prevent the deformation of the particles of the positive electrode active materials, such as cracking or breaking.

[0030]

However, when suspension is formed and stirred by a rotary and revolutionary mixer to coat the surface of the positive electrode active material whose weight per unit volume is large with graphene oxide whose weight is relatively small, coating is insufficient.

[0031]

Thus, to coat the surfaces of the particles of the positive electrode active materials with the graphene oxide, a method in which the graphene oxide and a polar solvent (such as water) are mixed and ultrasonic treatment is performed, the particles of the positive electrode active materials are mixed therein to prepare the suspension, and dried powder is produced with a spray dry

apparatus is preferably used. The dried powder produced in this manner is referred to as a composite in some cases.

[0032]

The size of one drop of spray liquid sprayed from a nozzle of the spray dry apparatus depends on a nozzle diameter.

[0033]

When the particle diameter is smaller than the nozzle diameter, there are a plurality of particles in one drop of the spray liquid sprayed from the nozzle. When the surface of the particle after drying under the condition where the largest particle size is smaller than the nozzle diameter is observed, there are some portions where the surface is coated with the graphene oxide; however, the coating is insufficient.

[0034]

The nozzle diameter of the spray dry apparatus is preferably substantially equal to the largest particle size of the active material because the coverage of the active material can be improved. Moreover, the largest particle size of the positive electrode active material is preferably adjusted to be substantially equal to the nozzle diameter in forming the positive electrode active material.

[0035]

Since the graphene oxide is well dispersed into water, the suspension of water and the graphene oxide can be formed by stirring using ultrasonic waves. The positive electrode active material is added to the suspension, and the suspension is sprayed with the spray dry apparatus, whereby powder in which the surface of the positive electrode active material is coated with the graphene oxide can be obtained.

[0036]

Note that the suspension becomes more acidic as the amount of graphene oxide is increased. Thus, part of the surface of the positive electrode active material (e.g., $LiCoO_2$ containing Mg and F) might be etched. Then, a hydrogen ion exponent (pH) of the suspension before being sprayed is preferably adjusted to be close to approximately pH7, that is, close to neutral, or higher than or equal to pH8, that is, alkaline. For the pH adjustment, a LiOH aqueous solution is preferably used. For example, in the case where $LiCoO_2$ is used for the positive electrode active material and only pure water is used as the dispersion medium of the suspension, the surface of the positive electrode active material may be damaged. Thus, a mixed solution of ethanol and water is used as the dispersion medium of the suspension, whereby damage to the surface of the active material may be reduced.

[0037]

The suspension is formed in the above manner, whereby the positive electrode active material whose surface is coated with the graphene oxide can be prepared efficiently. When the surface is coated with the graphene oxide, the deformation of the particles of the positive electrode active materials, such as cracking or breaking can be prevented. Moreover, even if the positive electrode active material whose surface is coated with the graphene oxide is exposed to the air after the formation, the change of properties or degradation can be suppressed. Here, "after the formation" refers to a period from the termination of the formation of the positive electrode active material to the start of the fabrication of the secondary battery containing the positive electrode active material and includes the storing, the transporting, and the like of the positive electrode active material. In addition, when the coating film is formed, the positive electrode active material and the electrolyte solution can be prevented from being in direct contact with each other to react; thus, the secondary battery using the coating film has high reliability.

[0038]

For the spray dry method, a known apparatus can be utilized, for example, a countercurrent pressure-nozzle-type spray dry apparatus and a counter-cocurrent pressure-nozzle-type spray dry apparatus can be utilized.

[0039]

Note that the graphene oxide coating the surface of the active material may be reduced when used in the secondary battery. The reduced graphene oxide is referred to as "RGO" in some cases. In RGO, in some cases, part of oxygen atoms remains in a state of oxygen or atomic group containing oxygen that is bonded to carbon. For example, RGO includes a functional group, e.g., an epoxy group, a carbonyl group such as a carboxyl group, or a hydroxyl group.

[0040]

Another embodiment of the present invention is a secondary battery which includes a positive electrode containing the above-described positive electrode active material or the above-described positive electrode active material coated with a coating film and a negative electrode.

[0041]

The secondary battery can have any of a variety of shapes to fit the form of the device to be used, for example, a cylindrical shape, a rectangular shape, a coin-type shape, and a laminated (flat plate) shape.

[0042]

According to one embodiment of the present invention, a positive electrode active material which suppresses a reduction in capacity due to charge and discharge cycles when used in a lithium ion secondary battery is provided. In addition, a secondary battery with excellent charge and discharge characteristics is provided. In addition, a highly safe or highly reliable

secondary battery is provided. In addition, a novel material, a novel active material particle, a novel secondary battery, or a formation method thereof is provided.

BRIEF DESCRIPTION OF DRAWINGS

[0043]

In the accompanying drawings:

FIGS. 1A to 1C show examples of a positive electrode active material;

FIGS. 2A and 2B illustrate crystal structures of a positive electrode active material;

FIG. 3 illustrates crystal structures of a positive electrode active material;

FIGS. 4A-1, 4A-2, 4A-3, 4B, 4C, 4D-1, and 4D-2 show a sol-gel method;

FIGS. 5A to 5C illustrate a segregation model of elements contained in a positive electrode active material;

FIGS. 6A to 6D illustrate a segregation model of elements contained in a positive electrode active material;

FIGS. 7A and 7B are cross-sectional views of an active material layer containing a graphene compound as a conductive additive;

FIGS. 8A to 8C illustrate a method for charging a secondary battery;

FIGS. 9A to 9D illustrate a method for charging a secondary battery;

FIG. 10 illustrates a method for discharging a secondary battery;

FIGS. 11A to 11C illustrate a coin-type secondary battery;

FIGS. 12A to 12D illustrate a cylindrical secondary battery;

FIGS. 13A and 13B illustrate an example of a secondary battery;

FIGS. 14A-1, 14A-2, 14B-1, and 14B-2 illustrate examples of secondary batteries;

FIGS. 15A and 15B illustrate examples of secondary batteries;

FIG. 16 illustrates an example of a secondary battery;

FIGS. 17A to 17C illustrate a laminated secondary battery;

FIGS. 18A and 18B illustrate a laminated secondary battery;

FIG. 19 is an external view of a secondary battery;

FIG. 20 is an external view of a secondary battery;

FIGS. 21A to 21C illustrate a formation method of a secondary battery;

FIGS. 22A, 22B1, 22B2, 22C, and 22D illustrate a bendable secondary battery;

FIGS. 23A and 23B illustrate a bendable secondary battery;

FIGS. 24A to 24H illustrate examples of electronic devices;

FIGS. 25A to 25C illustrate an example of an electronic device;

FIG. 26 illustrates examples of electronic devices;

FIGS. 27A to 27C illustrate examples of electronic devices;

FIG. 28 is a transmission electron microscope image of a positive electrode active material in Example 1;

FIGS. 29A1, 29A2, 29B1, 29B2, 29C1, and 29C2 are FFT images of transmission electron microscope images of a positive electrode active material in Example 1;

FIGS. 30A1, 30A2, 30B1, 30B2, 30C1, and 30C2 are element mapping images of a positive electrode active material in Example 1;

FIGS. 31A1, 31A2, 31B1, 31B2, 31C1, and 31C2 are element mapping images of a positive electrode active material of a comparative example in Example 1;

FIG. 32 is a graph showing TEM-EDX line analysis results of a positive electrode active material in Example 1;

FIG. 33 is a graph showing charge and discharge characteristics of a secondary battery in Example 1;

FIG. 34 is a graph showing charge and discharge characteristics of a secondary battery of a comparative example in Example 1;

FIG. 35 is a graph showing cycle characteristics of a secondary battery in Example 1;

FIG. 36 is a graph showing cycle characteristics of a secondary battery in Example 1;

FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2 are TEM-EDX plane analysis images of a comparative example in Example 2;

FIGS. 38A, 38B1, 38B2, 38C1, 38C2, 38D1, 38D2, 38E1, and 38E2 are TEM-EDX plane analysis images of a positive electrode active material in Example 2;

FIGS. 39A, 39B1, 39B2, 39C1, 39C2, 39D1, 39D2, 39E1, and 39E2 are TEM-EDX plane analysis images of a comparative example in Example 2;

FIGS. 40A, 40B1, 40B2, 40C1, 40C2, 40D1, 40D2, 40E1, and 40E2 are TEM-EDX plane analysis images of a positive electrode active material in Example 2;

FIGS. 41A and 41B are each a graph showing EDX point analysis results in Example 2;

FIGS. 42A and 42B are each a graph showing EDX point analysis results in Example 2;

FIG. 43 is a graph showing rate characteristics of a secondary battery in Example 2;

FIG. 44 is a graph showing temperature characteristics of a secondary battery in Example 2;

FIG. 45 is a graph showing cycle characteristics of a secondary battery in Example 2;

FIGS. 46A and 46B are each a graph showing XPS analysis results of a positive electrode active material in Example 3;

FIGS. 47A and 47B are each a graph showing cycle characteristics of a secondary battery containing a positive electrode active material in Example 3;

FIG. 48 is a graph showing cycle characteristics of a secondary battery containing a positive electrode active material in Example 3;

FIG. 49 is a graph showing cycle characteristics of a secondary battery containing a positive electrode active material in Example 3;

FIGS. 50A to 50C are each a graph showing charge and discharge characteristics of a secondary battery containing a positive electrode active material in Example 3;

FIGS. 51A to 51C are SEM images of a positive electrode active material in Example 4;

FIGS. 52A-1, 52A-2, 52B-1, 52B-2, 52C-1, and 52C-2 are SEM-EDX images of a positive electrode active material in Example 4;

FIG. 53 is a process flow chart of Example 5;

FIG. 54 illustrates a spray dry apparatus in Example 5;

FIG. 55 is a TEM image showing one embodiment of the present invention in Example 5;

FIG. 56 is a SEM image showing one embodiment of the present invention in Example 5;

FIG. 57 is a SEM image showing a comparative example in Example 5; and

FIGS. 58A and 58B are cross-sectional views of an active material layer containing a graphene compound as a conductive additive in Example 5.

BEST MODE FOR CARRYING OUT THE INVENTION

[0044]

Embodiments of the present invention will be described below in detail with reference to the drawings. Note that the present invention is not limited to the description below, and it is easily understood by those skilled in the art that modes and details of the present invention can be modified in various ways. In addition, the present invention should not be construed as being limited to the description in the embodiments given below.

[0045]

Note that in drawings used in this specification, the sizes, thicknesses, and the like of components such as a positive electrode, a negative electrode, an active material layer, a separator, and an exterior body are exaggerated for simplicity in some cases. Therefore, the sizes of the components are not limited to the sizes in the drawings and relative sizes between the components.

[0046]

Note that in structures of the present invention described in this specification and the like, the same portions or portions having similar functions are denoted by common reference numerals in different drawings, and descriptions thereof are not repeated. Further, the same hatching

pattern is applied to portions having similar functions, and the portions are not especially denoted by reference numerals in some cases.

[0047]

In this specification and the like, the Miller index is used for the expression of crystal planes and orientations. In the crystallography, a superscript bar is placed over a number in the expression using the Miller index; however, in this specification and the like, crystal planes and orientations are expressed by placing a minus sign (–) at the front of a number instead of placing the bar over a number because of expression limitations. Furthermore, an individual direction which shows an orientation in crystal is denoted by "[ ]", a set direction which shows all of the equivalent orientations is denoted by "< >", an individual direction which shows a crystal plane is denoted by "( )", and a set plane having equivalent symmetry is denoted by "{ }". In the drawings, the crystal planes and orientations are expressed by a number with a bar placed thereover, which is an original crystallographic expression. Note that 1 Å is 10^{-10} m.

[0048]

In this specification and the like, segregation refers to a phenomenon in which, in a solid made of a plurality of elements (e.g., A, B, and C), a certain element (for example, B) is non-uniformly distributed.

[0049]

In this specification and the like, a layered rock-salt crystal structure included in a composite oxide containing lithium and a transition metal refers to a crystal structure in which a rock-salt ion arrangement where cations and anions are alternately arranged is included and the lithium and the transition metal are regularly arranged to form a two-dimensional plane, so that lithium can be two-dimensionally diffused. Note that a defect such as a cation or anion vacancy can exist. In the layered rock-salt crystal structure, strictly, a lattice of a rock-salt crystal is distorted in some cases.

[0050]

In this specification and the like, a rock-salt crystal structure refers to a structure in which cations and anions are alternately arranged. Note that a cation or anion vacancy may exist.

[0051]

Anions of a layered rock-salt crystal and anions of a rock-salt crystal each form a cubic closest packed structure (face-centered cubic lattice structure). When a layered rock-salt crystal and a rock-salt crystal are in contact with each other, there is a crystal plane at which directions of cubic closest packed structures formed of anions are aligned with each other. A space group of the layered rock-salt crystal is R–3m, which is different from a space group Fm–3m of a general rock-salt crystal and a space group Fd–3m of a rock-salt crystal having the simplest symmetry;

thus, the Miller index of the crystal plane satisfying the above conditions in the layered rock-salt crystal is different from that in the rock-salt crystal. In this specification, in the layered rock-salt crystal and the rock-salt crystal, a state where the directions of the cubic closest packed structures formed of anions are aligned with each other is referred to as a state where crystal orientations are substantially aligned with each other.

[0052]

Whether the crystal orientations in two regions are aligned with each other or not can be judged by a transmission electron microscope (TEM) image, a scanning transmission electron microscope (STEM) image, a high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) image, an annular bright-field scan transmission electron microscopy (ABF-STEM) image, and the like. X-ray diffraction, electron diffraction, neutron diffraction, and the like can be used for judging. In the TEM image and the like, alignment of cations and anions can be observed as repetition of bright lines and dark lines. When the orientations of cubic closest packed structures of the layered rock-salt crystal and the rock-salt crystal are aligned with each other, a state where an angle between the repetition of bright lines and dark lines in the layered rock-salt crystal and the repetition of bright lines and dark lines in the rock-salt crystal is less than or equal to 5°, preferably less than or equal to 2.5° is observed. Note that, in the TEM image and the like, a light element such as oxygen or fluorine is not clearly observed in some cases; however, in such a case, alignment of orientations can be judged by arrangement of metal elements.

[0053]

Furthermore, in this specification and the like, a state where structures of two-dimensional interfaces have similarity is referred to as "epitaxy". Crystal growth in which structures of two-dimensional interfaces have similarity is referred to as "epitaxial growth". In addition, a state where three-dimensional structures have similarity or orientations are crystallographically the same is referred to as "topotaxy". Thus, in the case of topotaxy, when part of a cross section is observed, orientations of crystals in two regions (e.g., a region serving as a base and a region formed through growth) are substantially aligned with each other.

[0054]

(Embodiment 1)

[Structure of positive electrode active material]

First, a positive electrode active material 100, which is one embodiment of the present invention, is described with reference to FIGS. 1A to 1C. The positive electrode active material 100 refers to a substance containing a transition metal which can receive and release lithium ion electrochemically. As illustrated in FIG. 1A, the positive electrode active material 100 includes

a first region 101 inside and a second region 102 and a third region 103 in a superficial portion.

[0055]

As illustrated in FIG. 1B, the second region 102 does not necessarily cover the entire first region 101. Similarly, the third region 103 does not necessarily cover the entire second region 102. In addition, the third region 103 may be present in contact with the first region 101.

[0056]

The thicknesses of the second region 102 and the third region 103 may each differ depending on the positions.

[0057]

Furthermore, the third region 103 may be present inside the positive electrode active material 100. For example, in the case where the first region 101 is a polycrystal, the third region 103 may be present in the vicinity of a grain boundary. Furthermore, the third region 103 may be present in a portion which includes crystal defects or a crack portion in the positive electrode active material 100 or in the vicinity thereof. In FIG. 1B, parts of grain boundaries are shown by dotted lines. In this specification and the like, crystal defects refer to defects which can be observed from a TEM image and the like, that is, a structure in which another element enters crystal, a cavity, and the like. In addition, a crack portion refers to a crack or a break formed in a particle like a crack portion 106 illustrated in FIG. 1C, for example.

[0058]

Similarly, as illustrated in FIG. 1B, the second region 102 may be present inside the positive electrode active material 100. For example, in the case where the first region 101 is a polycrystal, the second region 102 may be present in the vicinity of a grain boundary. Furthermore, the second region 102 may be present in a portion which includes crystal defects or a crack portion in the positive electrode active material 100 or in the vicinity thereof. Moreover, the third region 103 and the second region 102 inside the positive electrode active material 100 may overlap.

[0059]

<First region 101>

The first region 101 contains a composite oxide of lithium and a first transition metal. In other words, the first region 101 contains lithium, a first transition metal, and oxygen.

[0060]

The composite oxide of lithium and a first transition metal preferably has a layered rock-salt crystal structure.

[0061]

As the first transition metal, only cobalt may be used, cobalt and manganese may be used,

or cobalt, manganese, and nickel may be used.

[0062]

That is, the first region can include lithium cobalt oxide, lithium manganese oxide, lithium nickel oxide, lithium cobalt oxide in which manganese is substituted for part of cobalt, lithium nickel-manganese-cobalt oxide, or the like. In addition to the transition metal, the first region 101 may include a metal other than the transition metal, such as aluminum.

[0063]

The first region 101 serves as a region which particularly contributes to a charge and discharge reaction in the positive electrode active material 100. To increase capacity of a secondary battery containing the positive electrode active material 100, the volume of the first region 101 is preferably larger than those of the second region and the third region.

[0064]

A material having a layered rock-salt crystal structure has features such as high discharge capacity and low resistance due to lithium that can be diffused two-dimensionally, which is preferably used for the first region 101. In addition, in the case where the first region 101 has a layered rock-salt crystal structure, a segregation of a representative element such as magnesium, which is described later, tends to occur unexpectedly.

[0065]

Note that the first region 101 may be a single crystal or a polycrystal. For example, the first region 101 may be a polycrystal in which an average crystallite size is greater than or equal to 280 nm and less than or equal to 630 nm. In the case of a polycrystal, a grain boundary can be observed from the TEM or the like in some cases. In addition, the average of crystal grain sizes can be calculated from the half width of XRD.

[0066]

The polycrystal has a clear crystal structure; thus, a two-dimensional diffusion path of lithium ions can be sufficiently ensured. In addition, a polycrystal is easily produced as compared with a single crystal; thus, a polycrystal is preferably used for the first region 101.

[0067]

Furthermore, the entire first region 101 does not necessarily have a layered rock-salt crystal structure. For example, part of the first region 101 may be amorphous or has another crystal structure.

[0068]

<Second region 102>

The second region 102 contains an oxide of a second transition metal. In other words, the second region 102 contains a second transition metal and oxygen.

[0069]

As the second transition metal, a non-stoichiometric metal is preferably used. In other words, the second region 102 preferably includes a non-stoichiometric compound. For example, as the second transition metal, at least one of titanium, vanadium, manganese, iron, chromium, niobium, cobalt, zinc, zirconium, nickel, and the like can be used. Note that the second transition metal is preferably an element different from that of the first transition metal.

[0070]

In this specification and the like, a non-stoichiometric metal refers to a metal that can have a plurality of valences. In addition, a non-stoichiometric compound refers to a compound of a metal that can have a plurality of valences and another element.

[0071]

The second region 102 preferably has a rock-salt crystal structure.

[0072]

The second region 102 serves as a buffer region which connects the first region 101 to the third region 103 which is described later. In the non-stoichiometric compound, an interatomic distance can be changed in accordance with a change in valence of a metal contained in the non-stoichiometric compound. In addition, in the non-stoichiometric compound, a cation or anion vacancy and dislocation (what is called Magneli phase) are often formed. Thus, the second region 102, which serves as a buffer region, can absorb a strain generated between the first region 101 and the third region 103.

[0073]

Furthermore, the second region 102 may contain lithium in addition to the second transition metal and oxygen. For example, lithium titanate or lithium manganite may be contained. Moreover, the second region 102 may contain a representative element contained in the third region 103 which is described later. The second region 102 that contains an element contained in the first region 101, such as lithium, and an element contained in the third region 103 is preferable because the second region 102 serves as a buffer region.

[0074]

That is, the second region 102 can contain lithium titanate, titanium oxide, vanadium oxide, manganese oxide, iron oxide, copper oxide, chromium oxide, niobium oxide, cobalt oxide, zinc oxide, or the like.

[0075]

In addition, the second region 102 may contain the first transition metal. For example, the second transition metal may be present in part of a first transition metal site of the composite oxide including the first transition metal.

[0076]

For example, in the case where the second transition metal is titanium, titanium may be present as titanium oxide (TiO_2) or lithium titanate ($LiTiO_2$) in the second region 102. Alternatively, in the second region 102, titanium may be substituted for part of the first transition metal site of the composite oxide of lithium and the first transition metal.

[0077]

Moreover, the second region 102 may contain fluorine.

[0078]

The second region 102 preferably has a crystal structure which is the same as that of the third region 103 which is described later. In this case, orientations of crystals of the second region 102 and the third region 103 are likely to be aligned with each other.

[0079]

The second region 102 preferably has a rock-salt crystal structure; however, the entire second region 102 does not necessarily have a rock-salt crystal structure. For example, the second region 102 may have another crystal structure such as a spinel crystal structure, an olivine crystal structure, a corundum crystal structure, or a rutile crystal structure.

[0080]

Furthermore, a crystal structure may have a strain as long as a structure where six oxygen atoms are adjacent to cations is kept. In addition, a cation vacancy may be present in part of the second region 102.

[0081]

Moreover, part of the second region 102 may be amorphous.

[0082]

When the thickness of the second region 102 is too small, the function as the buffer region is degraded; however, when the thickness of the second region 102 is too large, the capacity might be decreased. Thus, the second region 102 is preferably provided in a range from the surface of the positive electrode active material 100 to a depth of 20 nm, preferably a depth of 10 nm, in a depth direction. The second transition metal may have a concentration gradient.

[0083]

<Third region 103>

The third region 103 contains a compound of representative elements. A compound of representative elements is a stoichiometric compound. As the compound of representative elements, a compound made of representative elements which are electrochemically stable is preferable, and at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride can be used, for example.

[0084]

The third region 103 is in contact with an electrolyte solution when the positive electrode active material 100 is used in a secondary battery. Thus, for the third region 103, a material which is hardly changed electrochemically in the process of charging and discharging and is not easily transformed by contact with the electrolyte solution is preferably used. The compound of representative elements which is a stoichiometric compound and electrochemically stable is preferably used for the third region 103. The positive electrode active material 100 includes the third region 103 in a superficial portion to improve stability in charging and discharging of the secondary battery. Here, a state where stability of a secondary battery is high refers to a state where the crystal structure of the composite oxide of lithium and the first transition metal contained in the first region 101 is more stable. Alternatively, it refers to a state where a change in capacity of the secondary battery is small even if charging and discharging are repeated or a state where a change in valence of a metal contained in the positive electrode active material 100 is suppressed even after charging and discharging are repeated.

[0085]

The third region 103 may contain fluorine. In the case where the third region 103 contains fluorine, fluorine may be substituted for some anions in the compound of the representative elements.

[0086]

Fluorine is substituted for some anions in the compound of the representative elements, whereby diffusion properties of lithium can be improved. Thus, the third region 103 is less likely to prevent charging and discharging. In addition, when fluorine is present in a superficial portion of a positive electrode active material particle, corrosion resistance against a hydrofluoric acid generated by decomposition of an electrolyte solution is increased in some cases.

[0087]

Moreover, the third region 103 may include lithium, the first transition metal, and the second transition metal.

[0088]

The compound of the representative elements contained in the third region 103 preferably has a rock-salt crystal structure. When the third region 103 has a rock-salt crystal structure, orientations of crystals are likely to be aligned with those of the second region 102. The orientations of crystals of the first region 101, the second region 102, and the third region 103 are substantially aligned with each other, whereby the second region 102 and the third region 103 can serve as a more stable coating layer.

[0089]

However, the entire third region 103 does not necessarily have a rock-salt crystal structure. For example, the third region 103 may have another crystal structure such as a spinel crystal structure, an olivine crystal structure, a corundum crystal structure, or a rutile crystal structure.

[0090]

Furthermore, a crystal structure may have a strain as long as a structure where six oxygen atoms are adjacent to cations is kept. In addition, a cation vacancy may be present in part of the third region 103.

[0091]

Moreover, part of the third region 103 may be amorphous.

[0092]

When the thickness of the third region 103 is too small, the function of increasing stability in charging and discharging is degraded; however, when the thickness of the third region 103 is too large, the capacity might be decreased. Thus, the thickness of the third region 103 is preferably greater than or equal to 0.5 nm and less than or equal to 50 nm, further preferably greater than or equal to 0.5 nm and less than or equal to 2 nm.

[0093]

In the case where the third region 103 contains fluorine, fluorine is preferably present in a bonding state other than magnesium fluoride (MgF_2), lithium fluoride (LiF), and cobalt fluoride (CoF_2). Specifically, when an XPS analysis is performed on the vicinity of the surface of the positive electrode active material 100, a peak position of bonding energy with fluorine is preferably higher than or equal to 682 eV and lower than or equal to 685 eV, further preferably approximately 684.3 eV. The bonding energy does not correspond to those of MgF_2, LiF, and CoF_2.

[0094]

In this specification and the like, a peak position of bonding energy with an element in an XPS analysis refers to a value of bonding energy at which the maximum intensity of an energy spectrum is obtained in a range corresponding to bonding energy of the element.

[0095]

In general, when charging and discharging are repeated, a side reaction occurs in a positive electrode active material, for example, a first transition metal such as manganese, cobalt, or nickel is dissolved in an electrolyte solution, oxygen is released, and a crystal structure becomes unstable, so that the positive electrode active material deteriorates. However, the positive electrode active material 100 of one embodiment of the present invention includes the second region 102 serving as a buffer region and the third region 103 which is electrochemically stable. Thus, the dissolution of the first transition metal can be effectively suppressed, and the crystal structure of the composite oxide of lithium and the transition metal contained in the first region

101 can be more stable. As a result, the cycle characteristics of the secondary battery including the positive electrode active material 100 can be significantly improved. In addition, in the case where charging and discharging are performed at a voltage higher than 4.3 V (vs. Li/Li^+), in particular, a high voltage of 4.5 V (vs. Li/Li^+) or more, the structure of one embodiment of the present invention is significantly effective.

[0096]

<Heteroepitaxial growth and topotaxy>

The second region 102 is preferably formed by heteroepitaxial growth from the first region 101. Furthermore, the third region 103 is preferably formed by heteroepitaxial growth from the second region 102. A region formed by heteroepitaxial growth becomes topotaxy which has crystal orientations substantially three-dimensionally aligned with those of a region serving as a base. Thus, the first region 101, the second region 102, and the third region 103 can be topotaxy.

[0097]

When the crystal orientations of the first region 101, the second region 102, and the third region 103 are substantially aligned with each other, the second region 102 and the third region 103 serve as a coating layer which has a stable bond with the first region 101. As a result, the positive electrode active material 100 including a strong coating layer can be provided.

[0098]

Since the second region 102 and the third region 103 have a stable bond with the first region 101, when the positive electrode active material 100 is used for the secondary battery, a change of the crystal structure in the first region 101 which is caused by charging and discharging can be effectively suppressed. Even when lithium is released from the first region 101 due to charging, the coating layer having a stable bond can suppress release of cobalt and oxygen from the first region 101. Furthermore, a chemically stable material can be used for a region in contact with the electrolyte solution. Thus, a secondary battery having excellent cycle characteristics can be provided.

[0099]

<Degree of mismatch between regions>

To perform heteroepitaxial growth, the degree of a mismatch between crystals in a region serving as a base and crystals on which crystal growth is performed is important.

[0100]

In this specification and the like, the degree of a mismatch f is defined by the following Formula 1. The average of the nearest neighbor distances between oxygen and cations of the crystals in the region serving as a base is represented by a, and the average of the natural nearest neighbor distances between anions and cations of the crystals on which crystal growth is performed

is represented by b.

[0101]

[Formula 1]

$$f = \left|\frac{b-a}{a}\right| \quad \text{(Formula 1)}$$

[0102]

To perform heteroepitaxial growth, the degree of a mismatch f between crystals in a region serving as a base and crystals on which crystal growth is performed needs to be less than or equal to 0.12. To perform more stable heteroepitaxial growth to form a layered shape, the degree of a mismatch f is preferably less than or equal to 0.08, further preferably less than or equal to 0.04.

[0103]

Thus, materials of the first region 101 and the second region 102 are preferably selected so that the degree of a mismatch f between the layered rock-salt crystal structure in the first region 101 and the rock-salt crystal structure in the second region 102 is less than or equal to 0.12.

[0104]

Furthermore, materials of the second region 102 and the third region 103 are preferably selected so that the degree of a mismatch f between the rock-salt crystal structure in the second region 102 and the rock-salt crystal structure in the third region 103 is less than or equal to 0.12.

[0105]

Examples of materials and crystal planes of the first region 101, the second region 102, and the third region 103 are shown below, which satisfy the above-described conditions: the degree of a mismatch f between the layered rock-salt crystal structure in the first region 101 and the rock-salt crystal structure in the second region 102 is less than or equal to 0.12; and the degree of a mismatch f between the rock-salt crystal structure in the second region 102 and the rock-salt crystal structure in the third region 103 is less than or equal to 0.12.

[0106]

<<Example 1: Lithium cobaltate, lithium titanate, and magnesium oxide>>

First, FIGS. 2A and 2B and FIG. 3 show an example in which the first transition metal is cobalt, the first region 101 contains lithium cobaltate having a layered rock-salt crystal structure, the second transition metal is titanium, the second region 102 contains lithium titanate having a rock-salt crystal structure, and the compound of the representative elements in the third region 103 is magnesium oxide having a rock-salt crystal structure.

[0107]

FIG. 2A illustrates a model of a layered rock-salt crystal structure (a space group R–3mH) of lithium cobaltate ($LiCoO_2$), a model of a rock-salt crystal structure (a space group Fd–3mZ) of

lithium titanate ($LiTiO_2$), and a model of a rock-salt crystal structure (a space group Fd–3mZ) of magnesium oxide. FIG. 2A illustrates models each of which is seen from the *b*-axis direction.

[0108]

From FIG. 2A, it is not seen that the layered rock-salt crystals and the rock-salt crystals make topotaxy. Then, here, the layered rock-salt crystals are seen from a different direction (e.g., a direction indicated by an arrow in FIG. 2A). FIG. 2B illustrates a model of the layered rock-salt crystals seen from the <1–1–4> plane direction and models of the rock-salt crystals seen from the <100> plane direction.

[0109]

As illustrated in FIG. 2B, when the layered rock-salt crystals are seen from the <1–1–4> plane direction, atomic arrangement of the layered rock-salt crystals is highly similar to those of the rock-salt crystals seen from the <100> plane direction. In addition, the nearest neighbor distances between metal and oxygen have similar values. For example, in the layered rock-salt lithium cobaltate, a distance between Li and O is 2.089 Å and a distance between Co and O is 1.925 Å. In the rock-salt lithium titanate, a distance between Li and O is 2.138 Å and a distance between Ti and O is 2.051 Å. In the rock-salt magnesium oxide, a distance between Mg and O is 2.106 Å.

[0110]

Then, with reference to FIG. 3, the degree of a mismatch between the regions when the (1–1–4) crystal plane of the layered rock-salt crystal and the {100} crystal plane of the rock-salt crystal are in contact with each other is described.

[0111]

As illustrated in FIG. 3, a distance between metals through oxygen in a (1–1–4) crystal plane 101p (1–1–4) of lithium cobaltate having the layered rock-salt crystal structure in the first region 101 is 4.01 Å. Furthermore, a distance between metals through oxygen in a {100} crystal plane 102p {100} of lithium titanate having the rock-salt crystal structure in the second region 102 is 4.19 Å. Thus, the degree of a mismatch *f* between the crystal plane 101p (1–1–4) and the crystal plane 102p {100} is 0.04.

[0112]

In addition, a distance between metals through oxygen in a {100} crystal plane 103p {100} of magnesium oxide having the rock-salt crystal structure in the third region 103 is 4.21 Å. Thus, the degree of a mismatch *f* between the crystal plane 102p {100} and the crystal plane 103p {100} is 0.02.

[0113]

In this manner, the degree of a mismatch between the first region 101 and the second region 102 and the degree of a mismatch between the second region 102 and the third region 103 are sufficiently small; thus, the first region 101, the second region 102, and the third region 103 can be topotaxy.

[0114]

Although not illustrated in FIG. 3, if the crystal plane 101p (1–1–4) in the first region 101 and the crystal plane 103p {100} in the third region 103 are in contact with each other, the degree of a mismatch f is 0.05. That is, owing to the second region 102, the degree of a mismatch can be small. Moreover, with the second region 102 that is a non-stoichiometric transition metal oxide, the first region 101, the second region 102, and the third region 103 can be more stable topotaxy. Thus, the second region 102 and the third region 103 can serve as a coating layer having a stable bond with the first region 101.

[0115]

In this embodiment, the (1–1–4) plane of the layered rock-salt crystal and the {100} plane of the rock-salt crystal are in contact with each other; however, one embodiment of the present invention is not limited thereto as long as crystal planes which can be topotaxy are in contact with each other.

[0116]

<<Example 2: Lithium cobaltate, manganese oxide, and calcium oxide>>

Next, an example in which the first transition metal is cobalt, the first region 101 contains lithium cobaltate having a layered rock-salt crystal structure, the second transition metal is manganese, the second region 102 contains manganese oxide having a rock-salt crystal structure, and the compound of the representative elements in the third region 103 is calcium oxide having a rock-salt crystal structure is shown.

[0117]

Also in this case, as in FIGS. 2A and 2B and FIG. 3, when the layered rock-salt crystals are seen from the <1–1–4> plane direction, atomic arrangement of the layered rock-salt crystals in the first region 101 is highly similar to those of the rock-salt crystals in the second region 102 and the third region 103 seen from the <100> plane direction.

[0118]

The degree of a mismatch between the regions when the (1–1–4) crystal plane of the layered rock-salt crystal and the {100} crystal plane of the rock-salt crystal are in contact with each other is described. A distance between metals through oxygen in a crystal plane (1–1–4) of lithium cobaltate having the layered rock-salt crystal structure in the first region 101 is 4.01 Å. Furthermore, a distance between metals through oxygen in a crystal plane {100} of manganese

oxide having the rock-salt crystal structure in the second region 102 is 4.45 Å. Thus, the degree of a mismatch *f* between the crystal plane (1−1−4) in the first region 101 and the crystal plane {100} in the second region 102 is 0.11.

[0119]

In addition, a distance between metals through oxygen in a crystal plane {100} of calcium oxide having the rock-salt crystal structure in the third region 103 is 4.82 Å. Thus, the degree of a mismatch *f* between the crystal plane {100} in the second region 102 and the crystal plane {100} in the third region 103 is 0.08.

[0120]

In this manner, the degree of a mismatch between the first region 101 and the second region 102 and the degree of a mismatch between the second region 102 and the third region 103 are sufficiently small; thus, the first region 101, the second region 102, and the third region 103 can be topotaxy.

[0121]

If the crystal plane (1−1−4) in the first region 101 and the crystal plane {100} in the third region 103 are in contact with each other, the degree of a mismatch *f* is 0.20; thus, it is difficult to perform the heteroepitaxial growth. That is, owing to the second region 102, the heteroepitaxial growth from the first region to the third region can be performed. Thus, the second region 102 and the third region 103 can serve as a coating layer having a stable bond with the first region 101.

[0122]

<<Example 3: Lithium nickel-manganese-cobalt oxide, manganese oxide, and calcium oxide>>

Next, an example in which the first transition metals are nickel, manganese, and cobalt, the first region 101 contains lithium nickel-manganese-cobalt oxide ($LiNi_{0.33}Co_{0.33}Mn_{0.33}O_2$) having a layered rock-salt crystal structure, the second transition metal is manganese, the second region 102 contains manganese oxide having a rock-salt crystal structure, and the compound of the representative elements in the third region 103 is calcium oxide having a rock-salt crystal structure is shown.

[0123]

Also in this case, as illustrated in FIGS. 2A and 2B and FIG. 3, when the layered rock-salt crystals are seen from the <1−1−4> plane direction, atomic arrangement of the layered rock-salt crystals is highly similar to those of the rock-salt crystals seen from the <100> plane direction. The degree of a mismatch between the regions when the (1−1−4) crystal plane of the layered rock-salt crystal and the {100} crystal plane of the rock-salt crystal are in contact with each other is described.

[0124]

A distance between metals through oxygen in a crystal plane (1−1−4) of lithium nickel-manganese-cobalt oxide having the layered rock-salt crystal structure in the first region 101 is 4.07 Å. Furthermore, a distance between metals through oxygen in a crystal plane {100} of manganese oxide having the rock-salt crystal structure in the second region 102 is 4.45 Å. Thus, the degree of a mismatch *f* between the crystal plane (1−1−4) in the first region 101 and the crystal plane {100} in the second region 102 is 0.09.

[0125]

In addition, a distance between metals through oxygen in a crystal plane {100} of calcium oxide having the rock-salt crystal structure in the third region 103 is 4.82 Å. Thus, the degree of a mismatch *f* between the crystal plane {100} in the second region 102 and the crystal plane {100} in the third region 103 is 0.08.

[0126]

In this manner, the degree of a mismatch between the first region 101 and the second region 102 and the degree of a mismatch between the second region 102 and the third region 103 are sufficiently small; thus, the first region 101, the second region 102, and the third region 103 can be topotaxy.

[0127]

If the crystal plane (1−1−4) in the first region 101 and the crystal plane {100} in the third region 103 are in contact with each other, the degree of a mismatch *f* is 0.18; thus, it is difficult to perform the heteroepitaxial growth. That is, the second region 102 is provided, whereby the heteroepitaxial growth from the first region to the third region can be performed. Thus, the second region 102 and the third region 103 can serve as a coating layer having a stable bond with the first region 101.

[0128]

<Boundaries between regions>

As described above, the first region 101, the second region 102, and the third region 103 have different compositions. The element contained in each region has a concentration gradient in some cases. For example, the second transition metal in the second region 102 may have a concentration gradient. In addition, the third region 103 may have a concentration gradient of a representative element because a representative element preferably segregates in the third region 103 as described later. Thus, the boundaries between the regions are not clear in some cases.

[0129]

The difference of compositions of the first region 101, the second region 102, and the third region 103 can be observed using a TEM image, a STEM image, fast Fourier transform (FFT) analysis, energy dispersive X-ray spectrometry (EDX), an analysis in the depth direction by time-

of-flight secondary ion mass spectrometry (ToF-SIMS), X-ray photoelectron spectroscopy (XPS), Auger electron spectroscopy, thermal desorption spectroscopy (TDS), or the like.

[0130]

For example, in the TEM image and the STEM image, difference of constituent elements is observed as difference of brightness; thus, difference of constituent elements of the first region 101, the second region 102, and the third region 103 can be observed. Furthermore, also in plane analysis of EDX (e.g., element mapping), it can be observed that the first region 101, the second region 102, and the third region 103 contain different elements.

[0131]

By line analysis of EDX and analysis in the depth direction using ToF-SIMS, a peak of concentration of each element contained in the first region 101, the second region 102, and the third region 103 can be detected.

[0132]

However, clear boundaries between the first region 101, the second region 102, and the third region 103 are not necessarily observed by the analyses.

[0133]

In this specification and the like, the third region 103 that is present in a superficial portion of the positive electrode active material 100 refers to a region from the surface of the positive electrode active material 100 to a region where a concentration of a representative element such as magnesium which is detected by analysis in the depth direction is 1/5 of a peak. As the analysis in the depth direction, the line analysis of EDX, analysis in the depth direction using ToF-SIMS, or the like, which is described above, can be used.

[0134]

Furthermore, a peak of a concentration of a representative element is preferably present in a region from the surface of the positive electrode active material 100 to a depth of 3 nm toward the center, further preferably to a depth of 1 nm, and still further preferably to a depth of 0.5 nm.

[0135]

Although the depth at which the concentration of the representative element becomes 1/5 of the peak is different depending on the manufacturing method, in the case of a manufacturing method described later, the depth is approximately 2 nm to 5 nm from the surface of the positive electrode active material.

[0136]

The third region 103 that is present inside the first region 101 in the vicinity of a grain boundary, a crystal defect, or the like also refers to a region where a concentration of a representative element which is detected by analysis in the depth direction is higher than or equal

to 1/5 of a peak.

[0137]

A distribution of fluorine in the positive electrode active material 100 preferably overlaps with a distribution of the representative element. Thus, fluorine also has a concentration gradient, and a peak of a concentration of fluorine is preferably present in a region from the surface of the positive electrode active material 100 to a depth of 3 nm toward the center, further preferably to a depth of 1 nm, and still further preferably to a depth of 0.5 nm.

[0138]

In this specification and the like, the second region 102 that is present in a superficial portion of the positive electrode active material 100 refers to a region where a concentration of the second transition metal which is detected by analysis in the depth direction is higher than or equal to 1/2 of a peak. The second region 102 that is present inside the first region 101 in the vicinity of a grain boundary, a crystal defect, or the like also refers to a region where a concentration of the second transition metal which is detected by analysis in the depth direction is higher than or equal to 1/2 of a peak. As the analysis method, the line analysis of EDX, analysis in the depth direction using ToF-SIMS, or the like, which is described above, can be used.

[0139]

Thus, the third region 103 and the second region 102 overlap in some cases. Note that the third region 103 is preferably present in a region closer to the surface of the positive electrode active material particle than the second region 102 is. In addition, the peak of the concentration of the representative element is preferably present in a region closer to the surface of the positive electrode active material particle than the peak of the concentration of the second transition metal is.

[0140]

The peak of the second transition metal is preferably present in a region from a depth of 0.2 nm or more to a depth of 10 nm or less from the surface of the positive electrode active material 100 toward the center, further preferably in a region from a depth of 0.5 nm or more to a depth of 3 nm or less.

[0141]

The measurement range of the XPS is from the surface of the particle of the positive electrode active material 100 to a region at a depth of approximately 5 nm. Thus, a concentration of an element present at a depth of approximately 5 nm from the surface can be analyzed quantitatively. Thus, the concentration of elements in the third region 103 and the second region 102 present at a depth of approximately 5 nm from the surface can be analyzed quantitatively.

[0142]

When the surface of the positive electrode active material 100 is subjected to the XPS analysis and the concentration of the first transition metal is defined as 1, a relative value of the concentration of the second transition metal is preferably greater than or equal to 0.05 and less than or equal to 0.4, further preferably greater than or equal to 0.1 and less than or equal to 0.3. In addition, a relative value of the concentration of the representative element is preferably greater than or equal to 0.4 and less than or equal to 1.5, further preferably greater than or equal to 0.45 and less than or equal to 1.00. Furthermore, a relative value of the concentration of fluorine is preferably greater than or equal to 0.05 and less than or equal to 1.5, further preferably greater than or equal to 0.3 and less than or equal to 1.00.

[0143]

Note that, as described above, elements contained in the first region 101, the second region 102, and the third region 103 may each have a concentration gradient; thus, the first region 101 may contain the element in the second region 102 or the third region 103, such as fluorine. Similarly, the third region 103 may contain the element in the first region 101 or the second region 102. In addition, the first region 101, the second region 102, and the third region 103 may each contain another element such as carbon, sulfur, silicon, sodium, calcium, chlorine, or zirconium.

[0144]

[Particle diameter]

If the particle diameter of the positive electrode active material 100 is too large, diffusion of lithium is difficult, whereas if the particle diameter is too small, it is difficult to maintain a crystal structure described later. Thus, D50 (also referred to as a median diameter) is preferably 5 μm or more and 100 μm or less, and further preferably 10 μm or more and 70 μm or less. In the case where the coating film is formed on the surface of the positive electrode active material 100 by a spray dry apparatus in a later step, it is preferable that the nozzle diameter and the maximum particle diameter of the positive electrode active material 100 be substantially the same. When the particle diameter is less than 5 μm and a spray dry apparatus having a nozzle diameter of 20 μm is used, secondary particles are covered collectively, which leads to a decrease in coverage.

[0145]

To increase the density of the positive electrode active material layer, it is effective to mix large particles (the longest portion is approximately 20 μm or more and 40 μm or less) and small particles (the longest portion is approximately 1 μm) and embed spaces between the large particles with the small particles. Thus, there may be two peaks of particle size distribution.

[0146]

The particle size of the positive electrode active material is influenced not only by the

particle sizes of starting materials but also by a ratio between lithium and the first transition metal (hereinafter expressed as a ratio of Li to the first transition metal) which are contained in the starting material.

[0147]

In the case where the particle size of the starting material is small, the grain growth needs to be performed at the time of baking so that the grain size of the positive electrode active material is in the above-described preferred range.

[0148]

To promote the grain growth at the time of baking, it is effective to make the ratio of Li to the first transition metal of the starting material larger than 1, that is, to make the amount of lithium a little larger. For example, when the ratio of Li to the first transition metal is approximately 1.06, a positive electrode active material in which D50 is larger than or equal to 15 μm is easily obtained. Note that, as described later, lithium may be lost to the outside of a system in the formation process of the positive electrode active material; thus, the ratio between lithium and the first transition metal of the obtained positive electrode active material does not agree with the ratio between lithium and the first transition metal of the starting material in some cases.

[0149]

However, if the amount of lithium is too large to make the particle size be in the preferred range, the capacity retention rate of a secondary battery containing the positive electrode active material might be decreased.

[0150]

Then, the present inventors found that with the second region 102 containing the second transition metal in the superficial portion, the particle size can be in the preferred range by control of the ratio of Li to the first transition metal and a positive electrode active material having high capacity retention rate can be formed.

[0151]

In the positive electrode active material of one embodiment of the present invention including a region containing the second transition metal in the superficial portion, the ratio of Li to the first transition metal in the starting material is preferably greater than or equal to 1.00 and less than or equal to 1.07, further preferably greater than or equal to 1.03 and less than or equal to 1.06.

[0152]

[Formation of second region]

The second region 102 can be formed by coating particles of the composite oxide of lithium and the first transition metal with a material containing the second transition metal.

[0153]

As the coating method of the material containing the second transition metal, a liquid phase method such as a sol-gel method, a solid phase method, a sputtering method, an evaporation method, a chemical vapor deposition (CVD) method, a pulsed laser deposition (PLD) method, or the like can be used. In this embodiment, the case where the sol-gel method which can be performed with a uniform coverage under an atmospheric pressure is used is described.

[0154]

<Sol-gel method>

A method for forming a material containing the second transition metal using a sol-gel method is described with reference to FIGS. 4A-1, 4A-2, 4A-3, 4B, 4C, 4D-1, and 4D-2.

[0155]

First, an alkoxide of the second transition metal is dissolved in alcohol.

[0156]

FIG. 4A-1 shows a general formula of the alkoxide of the second transition metal. In the formula of FIG. 4A-1, M2 indicates the alkoxide of the second transition metal. R represents an alkyl group having 1 to 18 carbon atoms or a substituted or unsubstituted aryl group having 6 to 13 carbon atoms. Although FIG. 4A-1 shows the general formula in which the second transition metal has a valence of 4, one embodiment of the present invention is not limited thereto. The second transition metal may have a valence of 2, a valence of 3, a valence of 5, a valence of 6, or a valence of 7. In this case, the alkoxide of the second transition metal includes an alkoxy group corresponding to the valence of the second transition metal.

[0157]

FIG. 4A-2 shows a general formula of the titanium alkoxide used when titanium is used as the second transition metal. R in FIG. 4A-2 represents an alkyl group having 1 to 18 carbon atoms or a substituted or unsubstituted aryl group having 6 to 13 carbon atoms.

[0158]

As the titanium alkoxide, tetramethoxytitanium, tetraethoxytitanium, tetra-*n*-propoxytitanium, tetra-*i*-propoxytitanium (also referred to as tetraisopropyl orthotitanate, titanium (IV) isopropoxide, titanium tetraisopropoxide (IV), TTIP, and the like), tetra-*n*-butoxytitanium, tetra-*i*-butoxytitanium, tetra-*sec*-butoxytitanium, tetra-*t*-butoxytitanium, or the like can be used.

[0159]

FIG. 4A-3 shows a chemical formula of titanium (IV) isopropoxide (TTIP) described in a formation method below, which is a kind of titanium alkoxide.

[0160]

As a solvent in which the alkoxide of the second transition metal is dissolved, an alcohol

such as methanol, ethanol, propanol, 2-propanol, butanol, or 2-butanol is preferably used.

[0161]

Next, particles of composite oxide of lithium, a transition metal, magnesium, and fluorine are mixed into the alcohol solution of the alkoxide of the second transition metal and stirred in an atmosphere containing water vapor.

[0162]

When the solution is put in an atmosphere containing H_2O, hydrolysis of water and an alkoxide of the second transition metal occurs as in FIG. 4B. Then, as in FIG. 4C, dehydration condensation occurs between the products of FIG. 4B. When the hydrolysis of FIG. 4B and the condensation reaction of FIG. 4C occur repeatedly, a sol of an oxide of the second transition metal is generated. This reaction also occurs on a particle 110 of the composite oxide as in FIGS. 4D-1 and 4D-2, and a layer containing the second transition metal is formed on the surface of the particle 110.

[0163]

After that, the particle 110 is collected, and the alcohol is vaporized. The details of the formation method are described later.

[0164]

Note that in this embodiment, an example in which the particles of the composite oxide of lithium, the first transition metal, the representative element, and fluorine are coated with the material containing the second transition metal before the particles are applied to a positive electrode current collector is described; however, one embodiment of the present invention is not limited thereto. After the positive electrode active material layer including the particles of the composite oxide of lithium, the first transition metal, the representative element, and fluorine is formed on the positive electrode current collector, the positive electrode current collector and the positive electrode active material layer may be both soaked into an alkoxide solution of the second transition metal to be coated with the material containing the second transition metal.

[0165]

[Segregation of third region]

The third region 103 can be formed by a sputtering method, a solid phase method, a liquid phase method such as a sol-gel method, or the like. However, the present inventors found that when a source of a representative element such as magnesium and a source of fluorine are mixed with a material of the first region 101 and then the mixture is heated, the representative element is segregated on a superficial portion of the positive electrode active material particle to form the third region 103. In addition, they found that with the third region 103 formed in this manner, the positive electrode active material 100 has excellent cycle characteristics.

[0166]

In the case where the third region 103 is formed through the heating as described above, the heating is preferably performed after the particle of the composite oxide is coated with the material containing the second transition metal. This is because even after the particle is coated with the material containing the second transition metal, the representative element such as magnesium is unexpectedly segregated on the surface of the particle when the heating is performed.

[0167]

Segregation models of the representative element are described with reference to FIGS. 5A to 5C and FIGS. 6A to 6D. It is probable that the segregation model of the representative element such as magnesium is slightly different in accordance with the ratio between lithium and the first transition metal contained in a starting material. Then, a segregation model in which the ratio of Li to the first transition metal in the starting material is less than 1.03, that is, the amount of lithium is small, is described with reference to FIGS. 5A to 5C. In addition, a segregation model in which the ratio of Li to the first transition metal in the starting material is greater than or equal to 1.03, that is, the amount of lithium is large, is described with reference to FIGS. 6A to 6D. In these segregation models in FIGS. 5A to 5C and FIGS. 6A to 6D, the first transition metal is cobalt, the second transition metal is titanium, and the representative element is magnesium.

[0168]

FIG. 5A illustrates a model diagram of the vicinity of the surface of the particle 110 of the composite oxide containing lithium, cobalt, magnesium, and fluorine, which is formed at a ratio of Li to Co in the starting material of less than 1.03. A region 111 in the drawings contains lithium, cobalt, magnesium, and fluorine, and lithium cobaltate ($LiCoO_2$) is a main component of the region 111. Lithium cobaltate has a layered rock-salt structure.

[0169]

It is generally known that, at the time of synthesizing particles of the composite oxide containing lithium, cobalt, magnesium, and fluorine, lithium partly moves outside a system (a particle on which lithium is formed). This is because lithium is volatilized at the time of baking, lithium is eluted to a solvent at the time of mixing the starting material, and the like. Thus, the ratio of Li to Co in the particle 110 of the composite oxide containing lithium, cobalt, magnesium, and fluorine becomes smaller than the ratio of Li to Co in the starting material in some cases.

[0170]

When the ratio of Li to Co in the starting material is less than 1.03, on the surface of the particle 110, lithium is released from the lithium cobaltate and cobalt oxide is easily generated. Thus, as illustrated in FIG. 5A, the surface of the particle 110 of the composite oxide is covered with a cobalt oxide (CoO) layer 114 in some cases.

[0171]

The cobalt oxide has a rock-salt crystal structure. Thus, in the particle 110 in FIG. 5A, the cobalt oxide layer 114 having a rock-salt crystal structure is provided over and in contact with the region 111 containing lithium cobaltate having a layered rock-salt crystal structure in some cases.

[0172]

The particle 110 is coated with a material containing titanium by a sol-gel method or the like. FIG. 5B illustrates a state where the particle 110 is coated with a layer 112 containing titanium by a sol-gel method. At the stage of FIG. 5B, the layer 112 containing titanium is a gel of titanium oxide; thus, the crystallinity is low.

[0173]

Next, the particle 110 coated with the layer 112 containing titanium is heated. Although the details of the heating conditions are described later, for example, FIG. 5C illustrates a state where the particle 110 is heated in an oxygen atmosphere at 800 °C for two hours to form the positive electrode active material 100, which is one embodiment of the present invention. By heating, titanium in the layer 112 containing titanium is diffused into the inside of the particle 110. At the same time, magnesium and fluorine contained in the region 111 are segregated on the surface of the particle 110.

[0174]

As described above, on the surface of the particle 110, cobalt oxide having a rock-salt structure is present. In addition, magnesium oxide also has a rock-salt crystal structure. Thus, it is probable that magnesium is more stable in the state of being present as magnesium oxide on the surface of the particle 110 as compared with the state of being present inside the particle 110. That could be why magnesium is segregated on the surface of the particle 110 when the particle 110 is heated.

[0175]

Moreover, it is considered that fluorine contained in the starting material promotes the segregation of magnesium.

[0176]

Fluorine has higher electronegativity than oxygen. Thus, it is probable that even in a stable compound such as magnesium oxide, when fluorine is added, uneven charge distribution occurs and a bond between magnesium and oxygen is weakened. Furthermore, it is probable that fluorine is substituted for oxygen in the magnesium oxide, whereby magnesium easily moves around the substituted fluorine.

[0177]

Moreover, this can also be described from a phenomenon in which a melting point of a mixture decreases. When magnesium oxide (melting point: 2852 °C) and lithium fluoride (melting point: 848 °C) are added at the same time, the melting point of the magnesium oxide is lowered. It is considered that the melting point is lowered, whereby magnesium easily moves at the time of heating, and the segregation of magnesium easily occurs.

[0178]

Lastly, the third region 103 becomes a solid solution of cobalt oxide and magnesium oxide which has a rock-salt crystal structure. Furthermore, fluorine is probably substituted for part of oxygen contained in the cobalt oxide and the magnesium oxide.

[0179]

Cobalt sites of lithium cobaltate are substituted for part of the diffused titanium and lithium titanate is substituted for another part of the diffused titanium. The second region 102 after the heating contains lithium titanate having a rock-salt crystal structure.

[0180]

The first region 101 after the heating contains lithium cobaltate having a layered rock-salt crystal structure.

[0181]

Next, the case where the ratio of Li to Co in the starting material is larger than or equal to 1.03 is described with reference to FIGS. 6A to 6D. FIG. 6A illustrates a model diagram of the vicinity of the surface of the particle 120 of the composite oxide containing lithium, cobalt, magnesium, and fluorine, which is formed at a ratio of Li to Co in the starting material of greater than or equal to 1.03. A region 121 in the drawings contains lithium, cobalt, magnesium, and fluorine.

[0182]

Since the particle 120 in FIG. 6A contains a sufficient amount of lithium, even when lithium is released from the particle 120 at the time of baking the particle 120 of the composite oxide of lithium, cobalt, magnesium, and fluorine or the like, lithium is diffused from the inside of the particle 120 to the surface thereof to compensate; as a result, a cobalt oxide layer is not easily formed on the surface.

[0183]

FIG. 6B illustrates a state where the particle 120 in FIG. 6A is coated with a layer 122 containing titanium by a sol-gel method. At the stage of FIG. 6B, the layer 122 containing titanium is a gel of titanium oxide; thus, the crystallinity is low.

[0184]

FIG. 6C illustrates the state where the particle 120 coated with the layer 122 containing

titanium in FIG. 6B starts to be heated. By heating, titanium in the layer 122 containing titanium is diffused into the inside of the particle 110. The diffused titanium is bonded with lithium contained in the region 121 to become lithium titanate, and a layer 125 containing the lithium titanate is formed.

[0185]

Since lithium is bonded with titanium to form lithium titanate, the amount of lithium is relatively insufficient at the surface of the particle 120. Thus, it is probable that, as illustrated in FIG. 6C, a cobalt oxide layer 124 is temporarily formed on the surface of the particle 120.

[0186]

FIG. 6D illustrates the state where the state of FIG. 6C is sufficiently heated to form the positive electrode active material 100, which is one embodiment of the present invention. It is considered that, since the cobalt oxide layer 124 having a rock-salt crystal structure is present on the surface, magnesium is more stable in the state of being present as magnesium oxide on the surface of the particle 120 as compared with the state of being present inside the particle 120. As in the case of FIGS. 5A to 5C, fluorine promotes the segregation of magnesium.

[0187]

Thus, as illustrated in FIG. 6D, magnesium and fluorine contained in the region 121 are segregated on the surface to be the third region 103 with the cobalt oxide.

[0188]

In this manner, the positive electrode active material 100, which includes the third region 103 containing magnesium oxide and cobalt oxide, the second region 102 containing lithium titanate, and the first region 101 containing lithium cobaltate, is formed.

[0189]

Note that in the case where the representative element is segregated by heating, when the composite oxide containing lithium and the first transition metal included in the first region 101 is a polycrystal or has crystal defects, the representative element can be segregated not only in the superficial portion but also in the vicinity of a grain boundary of the composite oxide containing lithium and the first transition metal or in the vicinity of crystal defects thereof. The representative element segregated in the vicinity of a grain boundary or in the vicinity of crystal defects can contribute to further improvement in stability of the crystal structure of the composite oxide containing lithium and the first transition metal included in the first region 101.

[0190]

When the composite oxide containing lithium and the first transition metal included in the first region 101 includes a crack portion, the representative element is also segregated in the crack portion by heating. In addition, not only the representative element but also the second

transition metal can be segregated. The crack portion is in contact with the electrolyte solution like the surface of the particle. Thus, the representative element and the second transition metal are segregated in the crack portion, and the third region 103 and the second region 102 are generated, whereby a chemically stable material can be used for the region in contact with the electrolyte solution. As a result, a secondary battery having excellent cycle characteristics can be provided.

[0191]

The ratio between a representative element (T) and fluorine (F) in a starting material is preferably in a range of T:F = 1:x ($1.5 \leq x \leq 4$) (atomic ratio) because the segregation of the representative element effectively occurs. Further preferably, the ratio between T and F is approximately 1:2 (atomic ratio).

[0192]

Since the third region 103 formed by segregation is formed by epitaxial growth, orientations of crystals in the second region 102 and the third region 103 are partly and substantially aligned with each other in some cases. That is, the second region 102 and the third region 103 become topotaxy in some cases. When the orientations of crystals in the second region 102 and the third region 103 are substantially aligned with each other, these regions can serve as a more favorable coating layer.

[0193]

However, not all of the representative elements such as magnesium which is added as a starting material need not be segregated in the third region 103. For example, the first region 101 may contain a small amount of representative element such as magnesium.

[0194]

<Fourth region 104>

In addition, as illustrated in FIG. 1C, the positive electrode active material 100 may include a fourth region 104 on the third region 103. Furthermore, when the positive electrode active material 100 contains a defect such as a crack portion 106, the fourth region 104 may be present to embed the defect such as the crack portion 106.

[0195]

The fourth region 104 contains some elements contained in the second region 102 and the third region 103. For example, the fourth region 104 contains the second transition metal and the representative element.

[0196]

The fourth region 104 may have a projection, a stripe shape, or a layered shape. The fourth region 104 is formed using the second transition metal and the representative element not

contained in the second region 102 or the third region 103 of the second transition metals and the representative elements contained in the starting material and the like. That is, with the fourth region 104, the amount of the second transition metal and the representative element contained in the second region 102 and the third region 103 can be kept in an appropriate range and the crystal structures of the second region 102 and the third region 103 can be stabilized in some cases. Moreover, with the fourth region 104, the defect such as the crack portion 106 included in the positive electrode active material 100 can be repaired.

[0197]

The presence of the fourth region 104 and the shape of the fourth region 104 can be observed by a scanning electron microscope (SEM) or the like. Elements contained in the fourth region 104 can be analyzed by SEM-EDX or the like.

[0198]

[Method for forming positive electrode active material]

Next, an example of a method for forming the positive electrode active material 100, which is one embodiment of the present invention, is described.

[0199]

<Step 11: Preparation of starting materials>

First, starting materials are prepared. From the starting materials prepared in this process, the first region 101 and the third region 103 are formed finally.

[0200]

As materials of lithium and the first transition metal contained in the first region 101, a source of lithium and a source of the first transition metal are prepared. In addition, as materials of the compound of the representative elements contained in the third region 103, a source of the representative element is prepared.

[0201]

In addition to these sources, a source of fluorine is preferably prepared. Fluorine used for the materials has an effect of segregating the representative elements contained in the third region 103 on the surface of the positive electrode active material 100 in a later step.

[0202]

As the source of lithium, for example, lithium carbonate and lithium fluoride can be used. As the source of the first transition metal, for example, an oxide of the first transition metal can be used. As the source of the representative element, for example, an oxide of the representative element contained in the third region and fluoride of the representative element contained in the third region can be used.

[0203]

As the source of fluorine, for example, lithium fluoride and fluoride of the representative element contained in the third region can be used. That is, lithium fluoride can be used as either the source of lithium or the source of fluorine.

[0204]

The amount of fluorine contained in the source of fluorine is preferably 1.0 time to 4 times (atomic ratio), further preferably 1.5 times to 3 times (atomic ratio) the amount of representative element contained in the source of the representative element.

[0205]

<Step 12: Mixing of starting materials>

Next, the source of lithium, the source of the first transition metal, and the source of the representative element are mixed. In addition, the source of fluorine is preferably added. For example, a ball mill and a bead mill can be used for the mixing.

[0206]

<Step 13: First heating>

Next, the materials mixed in Step 12 are heated. In this step, the heating is referred to as baking or first heating in some cases. The heating is preferably performed at higher than or equal to 800 °C and lower than or equal to 1100 °C, further preferably at higher than or equal to 900 °C and lower than or equal to 1000 °C. The heating time is preferably greater than or equal to 2 hours and less than or equal to 20 hours. The baking is preferably performed in a dried atmosphere such as dry air. In the dried atmosphere, for example, the dew point is preferably lower than or equal to −50 °C, further preferably lower than or equal to −100 °C. In this embodiment, the heating is performed at 1000 °C for 10 hours, the temperature rising rate is 200 °C/h, and dry air whose dew point is −109 °C flows at 10 L/min. After that, the heated materials are cooled to room temperature.

[0207]

By the heating in Step 13, the composite oxide of lithium and the first transition metal having a layered rock-salt crystal structure can be synthesized. At this time, the representative element and fluorine contained in the starting materials form a solid solution in the composite oxide. However, some representative elements have been already segregated on the surface of the composite oxide in some cases.

[0208]

In addition, as the starting materials, particles of the composite oxide containing lithium, cobalt, fluorine, and magnesium which are synthesized in advance may be used. In this case, Step 12 and Step 13 can be omitted. For example, lithium cobalt oxide particles (C-20F, produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD.) can be used as one of the starting

materials. The lithium cobalt oxide particle has a diameter of approximately 20 μm and contains fluorine, magnesium, calcium, sodium, silicon, sulfur, and phosphorus in a region which can be analyzed by XPS from the surface.

[0209]

<Step 14: Coating with second transition metal>

Next, the composite oxide of lithium and the first transition metal is cooled to room temperature. Then, the surface of the composite oxide particle of lithium and the first transition metal is coated with a material containing the second transition metal. In the formation method example, a sol-gel method is used.

[0210]

First, the alkoxide of the second transition metal which is dissolved in alcohol and the composite oxide particles of lithium and the first transition metal are mixed.

[0211]

For example, in the case where titanium is used as the second transition metal, TTIP can be used as the alkoxide of the second transition metal. As alcohol, isopropanol can be used, for example.

[0212]

Next, the above mixed solution is stirred in an atmosphere containing water vapor. The stirring can be performed with a magnetic stirrer, for example. The stirring time is not limited as long as water and TTIP in an atmosphere cause hydrolysis and polycondensation reaction. For example, the stirring can be performed at 25 °C and a humidity of 90 %RH (Relative Humidity) for 4 hours.

[0213]

As described above, when water and TTIP in an atmosphere are reacted, a sol-gel reaction can proceed more slowly as compared with the case where liquid water is added. Alternatively, when titanium alkoxide and water are reacted at room temperature, a sol-gel reaction can proceed more slowly as compared with the case where heating is performed at a temperature higher than the boiling point of alcohol which is a solvent, for example. A sol-gel reaction proceeds slowly, whereby a high-quality coating layer containing titanium with a uniform thickness can be formed.

[0214]

After the above process, precipitate is collected from the mixed solution. As the collection method, filtration, centrifugation, evaporation and drying, or the like can be used. In this embodiment, filtration is used. For the filtration, a paper filter is used, and the residue is washed by alcohol which is the same as the solvent in which titanium alkoxide is dissolved.

[0215]

Then, the collected residue is dried. In this embodiment, vacuum drying is performed at 70 °C for one hour.

[0216]

<Step 15: Second heating>

Next, the composite oxide particle coated with the material containing the second transition metal, which is formed in Step 14, is heated. This step is referred to as second heating in some cases. In the heating, the retention time within a specified temperature range is preferably shorter than or equal to 50 hours, further preferably longer than or equal to 2 hours and shorter than or equal to 10 hours, still further preferably longer than or equal to 1 hour and shorter than or equal to 3 hours. If the heating time is too short, there is concern that the segregation of the representative elements does not occur; however, if the heating time is too long, there is concern that the favorable second region 102 is not formed because diffusion of the second transition metal proceeds too much.

[0217]

The specified temperature is preferably higher than or equal to 500 °C and lower than or equal to 1200 °C, further preferably higher than or equal to 800 °C and lower than or equal to 1000 °C. If the specified temperature is too low, there is concern that the segregation of the representative elements and the second transition metal does not occur. However, if the specified temperature is too high, there is concern that the first transition metal in the composite oxide particle is reduced to decompose the composite oxide particle, that a layered structure of lithium and the first transition metal in the composite oxide particle cannot be kept, and the like.

[0218]

In this embodiment, the specified temperature is 800 °C and kept for 2 hours, the temperature rising rate is 200 °C/h, and the flow rate of dry air is 10 L/min.

[0219]

By the heating in Step 15, the composite oxide of lithium and the first transition metal and the oxide of the second transition metal covering the composite oxide become topotaxy. In other words, the first region 101 and the second region 102 become topotaxy.

[0220]

By the heating in Step 15, the representative elements which form a solid solution inside the composite oxide particle of lithium and the first transition metal are unevenly distributed on the surface to form a solid solution, that is, the representative elements are segregated, the compound of the representative elements is formed, and the third region 103 is formed. At this time, the compound of the representative elements is formed by heteroepitaxial growth from the second region 102. That is, the second region 102 and the third region 103 become topotaxy.

[0221]

Since the second region 102 and the third region 103 contain crystals whose orientations are substantially aligned with each other and have a stable bond with the first region 101, when the positive electrode active material 100 is used for the secondary battery, a change of the crystal structure in the first region 101 which is caused by charging and discharging can be effectively suppressed. Even when lithium is released from the first region 101 due to charging, the superficial portion having a stable bond can suppress release of oxygen and the first transition metal such as cobalt from the first region 101. Furthermore, a chemically stable material can be used for a region in contact with the electrolyte solution. Thus, a secondary battery having excellent cycle characteristics can be provided.

[0222]

Note that the entire first region 101 and second region 102 does not need to become topotaxy as long as part of the first region 101 and second region 102 becomes topotaxy. Furthermore, the entire second region 102 and third region 103 does not need to become topotaxy as long as part of the second region 102 and third region 103 becomes topotaxy.

[0223]

In the case where the compound of the representative elements contained in the third region contains oxygen, the heating in Step 15 is preferably performed in an atmosphere containing oxygen. Heating in an atmosphere containing oxygen promotes the formation of the third region 103.

[0224]

Furthermore, fluorine contained in the starting materials promotes the segregation of the representative elements.

[0225]

In this manner, in the method for forming the positive electrode active material of one embodiment of the present invention, after the elements forming the second region 102 are coated, heating is performed to form the third region 103, and two kinds of regions can be formed on the surface of the positive electrode active material 100. That is, in general, two coating steps are necessary for providing two kinds of regions in a superficial portion; however, in the method for forming the positive electrode active material of one embodiment of the present invention, only one coating step (sol-gel process) is needed, which is a formation method with high productivity.

[0226]

<Step 16: Cooling>

Next, the particles heated in Step 15 are cooled to room temperature. The time of decreasing temperature is preferably long because topotaxy is easily generated. For example, the

time of decreasing temperature from retention temperature to room temperature is preferably the same as the time of increasing temperature or longer, specifically longer than or equal to 10 hours and shorter than or equal to 50 hours.

[0227]

<Step 17: Collecting>

Next, the cooled particles are collected. Moreover, the particles are preferably made to pass through a sieve. Through the above process, the positive electrode active material 100 including the first region 101, the second region 102, and the third region 103 can be formed.

[0228]

This embodiment can be implemented in appropriate combination with any of the other embodiments.

[0229]

(Embodiment 2)

In this embodiment, examples of materials which can be used for a secondary battery containing the positive electrode active material 100 described in the above embodiment are described. In this embodiment, a secondary battery in which a positive electrode, a negative electrode, and an electrolyte solution are wrapped in an exterior body is described as an example.

[0230]

[Positive electrode]

The positive electrode includes a positive electrode active material layer and a positive electrode current collector.

[0231]

<Positive electrode active material layer>

The positive electrode active material layer contains at least a positive electrode active material. The positive electrode active material layer may contain, in addition to the positive electrode active material, other materials such as a coating film of the active material surface, a conductive additive, and a binder.

[0232]

As the positive electrode active material, the positive electrode active material 100 described in the above embodiment can be used. When the above-described positive electrode active material 100 is used, a secondary battery with high capacity and excellent cycle characteristics can be obtained.

[0233]

Examples of the conductive additive include a carbon material, a metal material, and a conductive ceramic material. Alternatively, a fiber material may be used as the conductive

additive. The content of the conductive additive with respect to the total amount of the active material layer is preferably greater than or equal to 1 wt% and less than or equal to 10 wt%, more preferably greater than or equal to 1 wt% and less than or equal to 5 wt%.

[0234]

A network for electric conduction can be formed in the active material layer by the conductive additive. The conductive additive also allows maintaining of a path for electric conduction between the positive electrode active material particles. The addition of the conductive additive to the active material layer increases the electric conductivity of the active material layer.

[0235]

Examples of the conductive additive include natural graphite, artificial graphite such as mesocarbon microbeads, and carbon fiber. Examples of carbon fiber include mesophase pitch-based carbon fiber, isotropic pitch-based carbon fiber, carbon nanofiber, and carbon nanotube. Carbon nanotube can be formed by, for example, a vapor deposition method. Other examples of the conductive additive include carbon materials such as carbon black (e.g., acetylene black (AB)), graphite (black lead) particles, graphene, and fullerene. Alternatively, metal powder or metal fibers of copper, nickel, aluminum, silver, gold, or the like, a conductive ceramic material, or the like can be used.

[0236]

Alternatively, a graphene compound may be used as the conductive additive.

[0237]

A graphene compound has excellent electrical characteristics of high conductivity and excellent physical properties of high flexibility and high mechanical strength. Furthermore, a graphene compound has a planar shape. A graphene compound enables low-resistance surface contact. Furthermore, a graphene compound has extremely high conductivity even with a small thickness in some cases and thus allows a conductive path to be formed in an active material layer efficiently even with a small amount. For this reason, it is preferable to use a graphene compound as the conductive additive because the area where the active material and the conductive additive are in contact with each other can be increased. It is preferable to form the graphene compound serving as a conductive additive as a coating film to cover the entire surface of the active material with a spray dry apparatus, in which case the electrical resistance may be reduced. Here, it is particularly preferable to use, for example, graphene, multilayer graphene, or RGO as a graphene compound. Note that RGO refers to a compound obtained by reducing graphene oxide (GO), for example.

[0238]

In the case where an active material with a small particle diameter (e.g., 1 μm or less) is used, the specific surface area of the active material is large and thus more conductive paths for the active material particles are needed. Thus, the amount of conductive additive tends to increase and the supported amount of active material tends to decrease relatively. When the supported amount of active material decreases, the capacity of the secondary battery also decreases. In such a case, a graphene compound that can efficiently form a conductive path even in a small amount is particularly preferably used as the conductive additive because the supported amount of active material does not decrease.

[0239]

A cross-sectional structure example of an active material layer 200 containing a graphene compound as a conductive additive is described below.

[0240]

FIG. 7A shows a longitudinal cross-sectional view of the active material layer 200. The active material layer 200 includes positive electrode active material particles 100, a graphene compound 201 serving as a conductive additive, and a binder (not illustrated). Here, graphene or multilayer graphene may be used as the graphene compound 201, for example. The graphene compound 201 preferably has a sheet-like shape. The graphene compound 201 may have a sheet-like shape formed of a plurality of sheets of multilayer graphene and/or a plurality of sheets of graphene that partly overlap with each other.

[0241]

The longitudinal cross section of the active material layer 200 in FIG. 7B shows substantially uniform dispersion of the sheet-like graphene compounds 201 in the active material layer 200. The graphene compounds 201 are schematically shown by thick lines in FIG. 7B but are actually thin films each having a thickness corresponding to the thickness of a single layer or a multi-layer of carbon molecules. The plurality of graphene compounds 201 are formed in such a way as to partly coat or adhere to the surfaces of the plurality of positive electrode active material particles 100, so that the graphene compounds 201 make surface contact with the positive electrode active material particles 100.

[0242]

Here, the plurality of graphene compounds are bonded to each other to form a net-like graphene compound sheet (hereinafter referred to as a graphene compound net or a graphene net). The graphene net covering the active material can function as a binder for bonding active materials. The amount of a binder can thus be reduced, or the binder does not have to be used. This can increase the proportion of the active material in the electrode volume or weight. That is to say, the capacity of the secondary battery can be increased.

[0243]

Here, it is preferable to perform reduction after a layer to be the active material layer 200 is formed in such a manner that graphene oxide is used as the graphene compound 201 and mixed with an active material. When graphene oxide with extremely high dispersibility in a polar solvent is used for the formation of the graphene compounds 201, the graphene compounds 201 can be substantially uniformly dispersed in the active material layer 200. The solvent is removed by volatilization from a dispersion medium in which graphene oxide is uniformly dispersed, and the graphene oxide is reduced; hence, the graphene compounds 201 remaining in the active material layer 200 partly overlap with each other and are dispersed such that surface contact is made, thereby forming a three-dimensional conduction path. Note that graphene oxide can be reduced either by heat treatment or with the use of a reducing agent, for example.

[0244]

Unlike a conductive additive in the form of particles, such as acetylene black, which makes point contact with an active material, the graphene compound 201 is capable of making low-resistance surface contact; accordingly, the electrical conduction between the positive electrode active material particles 100 and the graphene compounds 201 can be improved with a smaller amount of the graphene compound 201 than that of a normal conductive additive. This increases the proportion of the positive electrode active material particles 100 in the active material layer 200, resulting in increased discharge capacity of the secondary battery.

[0245]

Alternatively, the graphene compound may cover the entire surface of the active material in advance with a spray dry apparatus. After that, at the time of forming the positive electrode active material layer, a graphene compound can be further added to make the conductive path between the active materials more favorable.

[0246]

As the binder, a rubber material such as styrene-butadiene rubber (SBR), styrene-isoprene-styrene rubber, acrylonitrile-butadiene rubber, butadiene rubber, or ethylene-propylene-diene copolymer can be used, for example. Alternatively, fluororubber can be used as the binder.

[0247]

For the binder, for example, water-soluble polymers are preferably used. As the water-soluble polymers, a polysaccharide and the like can be used. As the polysaccharide, a cellulose derivative such as carboxymethyl cellulose (CMC), methyl cellulose, ethyl cellulose, hydroxypropyl cellulose, diacetyl cellulose, or regenerated cellulose, starch, or the like can be used. It is more preferred that such water-soluble polymers be used in combination with any of the above rubber materials.

[0248]

Alternatively, as the binder, a material such as polystyrene, poly(methyl acrylate), poly(methyl methacrylate) (PMMA), sodium polyacrylate, polyvinyl alcohol (PVA), polyethylene oxide (PEO), polypropylene oxide, polyimide, polyvinyl chloride, polytetrafluoroethylene, polyethylene, polypropylene, polyisobutylene, polyethylene terephthalate, nylon, polyvinylidene fluoride (PVdF), polyacrylonitrile (PAN), ethylene-propylene-diene polymer, polyvinyl acetate, or nitrocellulose is preferably used.

[0249]

A plurality of the above materials may be used in combination for the binder.

[0250]

For example, a material having a significant viscosity modifying effect and another material may be used in combination. For example, a rubber material or the like has high adhesion or high elasticity but may have difficulty in viscosity modification when mixed in a solvent. In such a case, a rubber material or the like is preferably mixed with a material having a significant viscosity modifying effect, for example. As a material having a significant viscosity modifying effect, for example, a water-soluble polymer is preferably used. An example of a water-soluble polymer having an especially significant viscosity modifying effect is the above-mentioned polysaccharide; for example, a cellulose derivative such as carboxymethyl cellulose (CMC), methyl cellulose, ethyl cellulose, hydroxypropyl cellulose, diacetyl cellulose, or regenerated cellulose, or starch can be used.

[0251]

Note that a cellulose derivative such as carboxymethyl cellulose obtains a higher solubility when converted into a salt such as a sodium salt or an ammonium salt of carboxymethyl cellulose, and accordingly, easily exerts an effect as a viscosity modifier. The high solubility can also increase the dispersibility of an active material and other components in the formation of slurry for an electrode. In this specification, cellulose and a cellulose derivative used as a binder of an electrode include salts thereof.

[0252]

The water-soluble polymers stabilize viscosity by being dissolved in water and allow stable dispersion of the active material and another material combined as a binder such as styrene-butadiene rubber in an aqueous solution. Furthermore, a water-soluble polymer is expected to be easily and stably adsorbed to an active material surface because it has a functional group. Many cellulose derivatives such as carboxymethyl cellulose have functional groups such as a hydroxyl group and a carboxyl group. Because of functional groups, polymers are expected to interact with each other and cover an active material surface in a large area.

[0253]

In the case where the binder covering or being in contact with the active material surface forms a film, the film is expected to serve as a passivation film to suppress the decomposition of the electrolyte solution. Here, the passivation film refers to a film without electric conductivity or a film with extremely low electric conductivity, and can inhibit the decomposition of an electrolyte solution at a potential at which a battery reaction occurs in the case where the passivation film is formed on the active material surface, for example. It is preferred that the passivation film can conduct lithium ions while suppressing electric conduction.

[0254]

<Positive electrode current collector>

The positive electrode current collector can be formed using a material that has high conductivity, such as a metal like stainless steel, gold, platinum, aluminum, or titanium, or an alloy thereof. It is preferred that a material used for the positive electrode current collector not dissolve at the potential of the positive electrode. Alternatively, the positive electrode current collector can be formed using an aluminum alloy to which an element that improves heat resistance, such as silicon, titanium, neodymium, scandium, or molybdenum, is added. Still alternatively, a metal element that forms silicide by reacting with silicon can be used. Examples of the metal element that forms silicide by reacting with silicon include zirconium, titanium, hafnium, vanadium, niobium, tantalum, chromium, molybdenum, tungsten, cobalt, and nickel. The current collector can have any of various shapes including a foil-like shape, a plate-like shape (sheet-like shape), a net-like shape, a punching-metal shape, and an expanded-metal shape. The current collector preferably has a thickness of 5 μm to 30 μm.

[0255]

[Negative electrode]

The negative electrode includes a negative electrode active material layer and a negative electrode current collector. The negative electrode active material layer may contain a conductive additive and a binder.

[0256]

<Negative electrode active material>

As a negative electrode active material, for example, an alloy-based material or a carbon-based material can be used.

[0257]

For the negative electrode active material, an element which enables charge-discharge reactions by an alloying reaction and a dealloying reaction with lithium can be used. For example, a material containing at least one of silicon, tin, gallium, aluminum, germanium, lead, antimony,

bismuth, silver, zinc, cadmium, indium, and the like can be used. Such elements have higher capacity than carbon. In particular, silicon has a significantly high theoretical capacity of 4200 mAh/g. For this reason, silicon is preferably used as the negative electrode active material. Alternatively, a compound containing any of the above elements may be used. Examples of the compound include SiO, Mg_2Si, Mg_2Ge, SnO, SnO_2, Mg_2Sn, SnS_2, V_2Sn_3, $FeSn_2$, $CoSn_2$, Ni_3Sn_2, Cu_6Sn_5, Ag_3Sn, Ag_3Sb, Ni_2MnSb, $CeSb_3$, $LaSn_3$, $La_3Co_2Sn_7$, $CoSb_3$, InSb, and SbSn. Here, an element that enables charge-discharge reactions by an alloying reaction and a dealloying reaction with lithium, a compound containing the element, and the like may be referred to as an alloy-based material.

[0258]

In this specification and the like, SiO refers, for example, to silicon monoxide. SiO can alternatively be expressed as SiO_x. Here, x preferably has an approximate value of 1. For example, x is preferably 0.2 or more and 1.5 or less, more preferably 0.3 or more and 1.2 or less.

[0259]

As the carbon-based material, graphite, graphitizing carbon (soft carbon), non-graphitizing carbon (hard carbon), a carbon nanotube, graphene, carbon black, and the like can be used.

[0260]

Examples of graphite include artificial graphite and natural graphite. Examples of artificial graphite include meso-carbon microbeads (MCMB), coke-based artificial graphite, and pitch-based artificial graphite. As artificial graphite, spherical graphite having a spherical shape can be used. For example, MCMB is preferably used because it may have a spherical shape. Moreover, MCMB may preferably be used because it can relatively easily have a small surface area. Examples of natural graphite include flake graphite and spherical natural graphite.

[0261]

Graphite has a low potential substantially equal to that of a lithium metal (higher than or equal to 0.05 V and lower than or equal to 0.3 V vs. Li/Li^+) when lithium ions are intercalated into the graphite (while a lithium-graphite intercalation compound is formed). For this reason, a lithium-ion secondary battery can have a high operating voltage. In addition, graphite is preferred because of its advantages such as a relatively high capacity per unit volume, relatively small volume expansion, low cost, and higher level of safety than that of a lithium metal.

[0262]

Alternatively, for the negative electrode active material, an oxide such as titanium dioxide (TiO_2), lithium titanium oxide ($Li_4Ti_5O_{12}$), lithium-graphite intercalation compound (Li_xC_6), niobium pentoxide (Nb_2O_5), tungsten oxide (WO_2), or molybdenum oxide (MoO_2) can be used.

[0263]

Still alternatively, for the negative electrode active material, $Li_{3-x}M_xN$ (M = Co, Ni, or Cu) with a Li_3N structure, which is a nitride containing lithium and a transition metal, can be used. For example, $Li_{2.6}Co_{0.4}N_3$ is preferable because of high charge and discharge capacity (900 mAh/g and 1890 mAh/cm^3).

[0264]

A nitride containing lithium and a transition metal is preferably used, in which case lithium ions are contained in the negative electrode active material and thus the negative electrode active material can be used in combination with a material for a positive electrode active material which does not contain lithium ions, such as V_2O_5 or Cr_3O_8. In the case of using a material containing lithium ions as a positive electrode active material, the nitride containing lithium and a transition metal can be used for the negative electrode active material by extracting the lithium ions contained in the positive electrode active material in advance.

[0265]

Alternatively, a material which causes a conversion reaction can be used for the negative electrode active material; for example, a transition metal oxide which does not form an alloy with lithium, such as cobalt oxide (CoO), nickel oxide (NiO), and iron oxide (FeO), may be used. Other examples of the material which causes a conversion reaction include oxides such as Fe_2O_3, CuO, Cu_2O, RuO_2, and Cr_2O_3, sulfides such as $CoS_{0.89}$, NiS, and CuS, nitrides such as Zn_3N_2, Cu_3N, and Ge_3N_4, phosphides such as NiP_2, FeP_2, and CoP_3, and fluorides such as FeF_3 and BiF_3.

[0266]

For the conductive additive and the binder that can be included in the negative electrode active material layer, materials similar to those of the conductive additive and the binder that can be included in the positive electrode active material layer can be used.

[0267]

<Negative electrode current collector>

For the negative electrode current collector, a material similar to that of the positive electrode current collector can be used. Note that a material which is not alloyed with a carrier ion such as lithium is preferably used for the negative electrode current collector.

[0268]

[Electrolyte solution]

The electrolyte solution contains a solvent and an electrolyte. As a solvent of the electrolyte solution, an aprotic organic solvent is preferably used. For example, one of ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate, chloroethylene carbonate, vinylene carbonate, γ-butyrolactone, γ-valerolactone, dimethyl carbonate (DMC), diethyl carbonate (DEC),

ethyl methyl carbonate (EMC), methyl formate, methyl acetate, ethyl acetate, methyl propionate, ethyl propionate, propyl propionate, methyl butyrate, 1,3-dioxane, 1,4-dioxane, dimethoxyethane (DME), dimethyl sulfoxide, diethyl ether, methyl diglyme, acetonitrile, benzonitrile, tetrahydrofuran, sulfolane, and sultone can be used, or two or more of these solvents can be used in an appropriate combination in an appropriate ratio.

[0269]

Alternatively, when one or more kinds of ionic liquids (room temperature molten salts) which have features of non-flammability and non-volatility is used as a solvent of the electrolyte solution, a secondary battery can be prevented from exploding or catching fire even when the secondary battery internally shorts out or the internal temperature increases owing to overcharging or the like. An ionic liquid contains a cation and an anion. The ionic liquid contains an organic cation and an anion. Examples of the organic cation used for the electrolyte solution include aliphatic onium cations such as a quaternary ammonium cation, a tertiary sulfonium cation, and a quaternary phosphonium cation, and aromatic cations such as an imidazolium cation and a pyridinium cation. Examples of the anion used for the electrolyte solution include a monovalent amide-based anion, a monovalent methide-based anion, a fluorosulfonate anion, a perfluoroalkylsulfonate anion, a tetrafluoroborate anion, a perfluoroalkylborate anion, a hexafluorophosphate anion, and a perfluoroalkylphosphate anion.

[0270]

As an electrolyte dissolved in the above-described solvent, one of lithium salts such as $LiPF_6$, $LiClO_4$, $LiAsF_6$, $LiBF_4$, $LiAlCl_4$, LiSCN, LiBr, LiI, Li_2SO_4, $Li_2B_{10}Cl_{10}$, $Li_2B_{12}Cl_{12}$, $LiCF_3SO_3$, $LiC_4F_9SO_3$, $LiC(CF_3SO_2)_3$, $LiC(C_2F_5SO_2)_3$, $LiN(CF_3SO_2)_2$, $LiN(C_4F_9SO_2)(CF_3SO_2)$, and $LiN(C_2F_5SO_2)_2$ can be used, or two or more of these lithium salts can be used in an appropriate combination in an appropriate ratio.

[0271]

The electrolyte solution used for a secondary battery is preferably highly purified and contains a small amount of dust particles and elements other than the constituent elements of the electrolyte solution (hereinafter also simply referred to as impurities). Specifically, the weight ratio of impurities to the electrolyte solution is less than or equal to 1 %, preferably less than or equal to 0.1 %, and further preferably less than or equal to 0.01 %.

[0272]

Furthermore, an additive agent such as vinylene carbonate, propane sultone (PS), *tert*-butylbenzene (TBB), fluoroethylene carbonate (FEC), LiBOB, or a dinitrile compound such as succinonitrile or adiponitrile may be added to the electrolyte solution. The concentration of a material to be added with respect to the whole solvent is, for example, higher than or equal to 0.1

wt% and lower than or equal to 5 wt%.

[0273]

Alternatively, a gelled electrolyte obtained in such a manner that a polymer is swelled with an electrolyte solution may be used.

[0274]

When a polymer gel electrolyte is used, safety against liquid leakage and the like is improved. Furthermore, a secondary battery can be thinner and more lightweight.

[0275]

As a polymer that undergoes gelation, a silicone gel, an acrylic gel, an acrylonitrile gel, a polyethylene oxide-based gel, a polypropylene oxide-based gel, a fluorine-based polymer gel, or the like can be used.

[0276]

Examples of the polymer include a polymer having a polyalkylene oxide structure, such as polyethylene oxide (PEO); PVDF; polyacrylonitrile; and a copolymer containing any of them. For example, PVDF-HFP, which is a copolymer of PVDF and hexafluoropropylene (HFP) can be used. The formed polymer may be porous.

[0277]

Instead of the electrolyte solution, a solid electrolyte including an inorganic material such as a sulfide-based inorganic material or an oxide-based inorganic material, or a solid electrolyte including a high-molecular material such as a polyethylene oxide (PEO)-based high-molecular material may alternatively be used. When the solid electrolyte is used, a separator and a spacer are not necessary. Furthermore, since the battery can be entirely solidified, there is no possibility of liquid leakage to increase the safety of the battery dramatically.

[0278]

[Separator]

The secondary battery preferably includes a separator. As the separator, for example, paper; nonwoven fabric; glass fiber; ceramics; or synthetic fiber using nylon (polyamide), vinylon (polyvinyl alcohol-based fiber), polyester, acrylic, polyolefin, or polyurethane can be used. The separator is preferably formed to have an envelope-like shape to wrap one of the positive electrode and the negative electrode.

[0279]

The separator may have a multilayer structure. For example, an organic material film such as polypropylene or polyethylene can be coated with a ceramic-based material, a fluorine-based material, a polyamide-based material, a mixture thereof, or the like. Examples of the ceramic-based material include aluminum oxide particles and silicon oxide particles. Examples

of the fluorine-based material include PVDF and a polytetrafluoroethylene. Examples of the polyamide-based material include nylon and aramid (meta-based aramid and para-based aramid).

[0280]

Deterioration of the separator in charging and discharging at high voltage can be suppressed and thus the reliability of the secondary battery can be improved because oxidation resistance is improved when the separator is coated with the ceramic-based material. In addition, when the separator is coated with the fluorine-based material, the separator is easily brought into close contact with an electrode, resulting in high output characteristics. When the separator is coated with the polyamide-based material, in particular, aramid, the safety of the secondary battery is improved because heat resistance is improved.

[0281]

For example, both surfaces of a polypropylene film may be coated with a mixed material of aluminum oxide and aramid. Alternatively, a surface of the polypropylene film in contact with the positive electrode may be coated with the mixed material of aluminum oxide and aramid, and a surface of the polypropylene film in contact with the negative electrode may be coated with the fluorine-based material.

[0282]

With the use of a separator having a multilayer structure, the capacity of the secondary battery per volume can be increased because the safety of the secondary battery can be maintained even when the total thickness of the separator is small.

[0283]

[Exterior body]

For an exterior body included in the secondary battery, a metal material such as aluminum and a resin material can be used, for example. An exterior body in the form of a film can also be used. As the film, for example, a film having a three-layer structure in which a highly flexible metal thin film of aluminum, stainless steel, copper, nickel, or the like is provided over a film formed of a material such as polyethylene, polypropylene, polycarbonate, ionomer, or polyamide, and an insulating synthetic resin film of a polyamide-based resin, a polyester-based resin, or the like is provided as the outer surface of the exterior body over the metal thin film can be used.

[0284]

[Charging and discharging methods]

The secondary battery can be charged and discharged in the following manner, for example.

[0285]

<<CC charge>>

First, CC charge, which is one of charging methods, is described. CC charge is a charging method in which a constant current is made to flow to a secondary battery in the whole charging period and charge is terminated when the voltage reaches a predetermined voltage. The secondary battery is assumed to be an equivalent circuit with internal resistance R and secondary battery capacitance C as illustrated in FIG. 8A. In that case, a secondary battery voltage V_B is the sum of a voltage V_R applied to the internal resistance R and a voltage V_C applied to the secondary battery capacitance C.

[0286]

While the CC charge is performed, a switch is on as illustrated in FIG. 8A, so that a constant current I flows to the secondary battery. During the period, the current I is constant; thus, according to the Ohm's law ($V_R = R \times I$), the voltage V_R applied to the internal resistance R is also constant. In contrast, the voltage V_C applied to the secondary battery capacitance C increases over time. Accordingly, the secondary battery voltage V_B increases over time.

[0287]

When the secondary battery voltage V_B reaches a predetermined voltage, e.g., 4.3 V, the charge is terminated. On termination of the CC charge, the switch is turned off as illustrated in FIG. 8B, and the current I becomes 0. Thus, the voltage V_R applied to the internal resistance R becomes 0 V. Consequently, the secondary battery voltage V_B is decreased by the lost voltage drop in the internal resistance R.

[0288]

FIG. 8C shows an example of the secondary battery voltage V_B and charging current during a period in which the CC charge is performed and after the CC charge is terminated. The secondary battery voltage V_B increases while the CC charge is performed, and slightly decreases after the CC charge is terminated.

[0289]

<<CCCV charge>>

Next, CCCV charge, which is a charging method different from the above-described method, is described. CCCV charge is a charging method in which CC charge is performed until the voltage reaches a predetermined voltage and then constant voltage (CV) charge is performed until the amount of current flow becomes small, specifically, a termination current value.

[0290]

While the CC charge is performed, a switch of a constant current power source is on and a switch of a constant voltage power source is off as illustrated in FIG. 9A, so that the constant current I flows to the secondary battery. During the period, the current I is constant; thus, according to the Ohm's law ($V_R = R \times I$), the voltage V_R applied to the internal resistance R is also

constant. In contrast, the voltage V_C applied to the secondary battery capacitance C increases over time. Accordingly, the secondary battery voltage V_B increases over time.

[0291]

When the secondary battery voltage V_B reaches a predetermined voltage, e.g., 4.3 V, switching is performed from the CC charge to the CV charge. While the CV charge is performed, the switch of the constant voltage power source is on and the switch of the constant current power source is off as illustrated in FIG. 9B; thus, the secondary battery voltage V_B is constant. In contrast, the voltage V_C applied to the secondary battery capacitance C increases over time. Since $V_B = V_R + V_C$ is satisfied, the voltage V_R applied to the internal resistance R decreases over time. As the voltage V_R applied to the internal resistance R decreases, the current I flowing to the secondary battery also decreases according to the Ohm's law ($V_R = R \times I$).

[0292]

When the current I flowing to the secondary battery becomes a predetermined current, e.g., approximately 0.01 C, charge is terminated. On termination of the CCCV charge, all the switches are turned off as illustrated in FIG. 9C, so that the current I becomes 0. Thus, the voltage V_R applied to the internal resistance R becomes 0 V. However, the voltage V_R applied to the internal resistance R becomes sufficiently small by the CV charge; thus, even when a voltage drop no longer occurs in the internal resistance R, the secondary battery voltage V_B hardly decreases.

[0293]

FIG. 9D shows an example of the secondary battery voltage V_B and charging current while the CCCV charge is performed and after the CCCV charge is terminated. Even after the CCCV charge is terminated, the secondary battery voltage V_B hardly decreases.

[0294]

<<CC discharge>>

Next, CC discharge, which is one of discharging methods, is described. CC discharge is a discharging method in which a constant current is made to flow from the secondary battery in the whole discharging period, and discharge is terminated when the secondary battery voltage V_B reaches a predetermined voltage, e.g., 2.5 V.

[0295]

FIG. 10 shows an example of the secondary battery voltage V_B and discharging current while the CC discharge is performed. As discharge proceeds, the secondary battery voltage V_B decreases.

[0296]

Next, a discharge rate and a charge rate will be described. The discharge rate refers to the relative ratio of discharging current to battery capacity and is expressed in a unit C. A current

of approximately 1 C in a battery with a rated capacity X (Ah) is X A. The case where discharge is performed at a current of $2X$ A is rephrased as follows: discharge is performed at 2 C. The case where discharge is performed at a current of $X/5$ A is rephrased as follows: discharge is performed at 0.2 C. Similarly, the case where charging is performed at a current of $2X$ A is rephrased as follows: charging is performed at 2 C, and the case where charging is performed at a current of $X/5$ A is rephrased as follows: charging is performed at 0.2 C.

[0297]

(Embodiment 3)

In this embodiment, examples of a shape of a secondary battery containing the positive electrode active material 100 described in the above embodiment are described. For the materials used for the secondary battery described in this embodiment, the description of the above embodiment can be referred to.

[0298]

[Coin-type secondary battery]

First, an example of a coin-type secondary battery is described. FIG. 11A is an external view of a coin-type (single-layer flat type) secondary battery, and FIG. 11B is a cross-sectional view thereof.

[0299]

In a coin-type secondary battery 300, a positive electrode can 301 doubling as a positive electrode terminal and a negative electrode can 302 doubling as a negative electrode terminal are insulated from each other and sealed by a gasket 303 made of polypropylene or the like. A positive electrode 304 includes a positive electrode current collector 305 and a positive electrode active material layer 306 provided in contact with the positive electrode current collector 305. A negative electrode 307 includes a negative electrode current collector 308 and a negative electrode active material layer 309 provided in contact with the negative electrode current collector 308.

[0300]

Note that only one surface of each of the positive electrode 304 and the negative electrode 307 used for the coin-type secondary battery 300 is provided with an active material layer.

[0301]

For the positive electrode can 301 and the negative electrode can 302, a metal having a corrosion-resistant property to an electrolyte solution, such as nickel, aluminum, or titanium, an alloy of such a metal, or an alloy of such a metal and another metal (e.g., stainless steel) can be used. Alternatively, the positive electrode can 301 and the negative electrode can 302 are preferably covered with nickel, aluminum, or the like in order to prevent corrosion due to the electrolyte solution. The positive electrode can 301 and the negative electrode can 302 are

electrically connected to the positive electrode 304 and the negative electrode 307, respectively.

[0302]

The negative electrode 307, the positive electrode 304, and the separator 310 are immersed in the electrolyte solution. Then, as illustrated in FIG. 11B, the positive electrode 304, the separator 310, the negative electrode 307, and the negative electrode can 302 are stacked in this order with the positive electrode can 301 positioned at the bottom, and the positive electrode can 301 and the negative electrode can 302 are subjected to pressure bonding with the gasket 303 located therebetween. In such a manner, the coin-type secondary battery 300 can be manufactured.

[0303]

When the positive electrode active material described in the above embodiment is used in the positive electrode 304, the coin-type secondary battery 300 with high capacity and excellent cycle characteristics can be obtained.

[0304]

Here, a current flow in charging a secondary battery is described with reference to FIG. 11C. When a secondary battery using lithium is regarded as a closed circuit, lithium ions transfer and a current flows in the same direction. Note that in the secondary battery using lithium, an anode and a cathode change places in charge and discharge, and an oxidation reaction and a reduction reaction occur on the corresponding sides; hence, an electrode with a high reaction potential is called a positive electrode and an electrode with a low reaction potential is called a negative electrode. For this reason, in this specification, the positive electrode is referred to as a "positive electrode" or a "plus electrode" and the negative electrode is referred to as a "negative electrode" or a "minus electrode" in all the cases where charge is performed, discharge is performed, a reverse pulse current is supplied, and a charging current is supplied. The use of the terms "anode" and "cathode" related to an oxidation reaction and a reduction reaction might cause confusion because the anode and the cathode change places at the time of charging and discharging. Thus, the terms "anode" and "cathode" are not used in this specification. If the term "anode" or "cathode" is used, it should be mentioned that the anode or the cathode is which of the one at the time of charging or the one at the time of discharging and corresponds to which of a positive (plus) electrode or a negative (minus) electrode.

[0305]

Two terminals in FIG. 11C are connected to a charger, and the coin-type secondary battery 300 is charged. As the charge of the coin-type secondary battery 300 proceeds, a potential difference between electrodes increases.

[0306]

[Cylindrical secondary battery]

Next, an example of a cylindrical secondary battery will be described with reference to FIGS. 12A to 12D. A cylindrical secondary battery 600 includes, as illustrated in FIG. 12A, a positive electrode cap (battery lid) 601 on the top surface and a battery can (outer can) 602 on the side and bottom surfaces. The positive electrode cap and the battery can (outer can) 602 are insulated from each other by a gasket (insulating packing) 610.

[0307]

FIG. 12B is a schematic cross-sectional view of the cylindrical secondary battery. Inside the battery can 602 having a hollow cylindrical shape, a battery element in which a strip-like positive electrode 604 and a strip-like negative electrode 606 are wound with a strip-like separator 605 located therebetween is provided. Although not illustrated, the battery element is wound around a center pin. One end of the battery can 602 is close and the other end thereof is open. For the battery can 602, a metal having a corrosion-resistant property to an electrolyte solution, such as nickel, aluminum, or titanium, an alloy of such a metal, or an alloy of such a metal and another metal (e.g., stainless steel) can be used. Alternatively, the battery can 602 is preferably covered with nickel, aluminum, or the like in order to prevent corrosion due to the electrolyte solution. Inside the battery can 602, the battery element in which the positive electrode, the negative electrode, and the separator are wound is provided between a pair of insulating plates 608 and 609 that face each other. Furthermore, a nonaqueous electrolyte solution (not illustrated) is injected inside the battery can 602 provided with the battery element. As the nonaqueous electrolyte solution, a nonaqueous electrolyte solution that is similar to that of the coin-type secondary battery can be used.

[0308]

Since the positive electrode and the negative electrode of the cylindrical secondary battery are wound, active materials are preferably formed on both sides of the current collectors. A positive electrode terminal (positive electrode current collecting lead) 603 is connected to the positive electrode 604, and a negative electrode terminal (negative electrode current collecting lead) 607 is connected to the negative electrode 606. Both the positive electrode terminal 603 and the negative electrode terminal 607 can be formed using a metal material such as aluminum. The positive electrode terminal 603 and the negative electrode terminal 607 are resistance-welded to a safety valve mechanism 612 and the bottom of the battery can 602, respectively. The safety valve mechanism 612 is electrically connected to the positive electrode cap 601 through a positive temperature coefficient (PTC) element 611. The safety valve mechanism 612 cuts off electrical connection between the positive electrode cap 601 and the positive electrode 604 when the internal pressure of the battery exceeds a predetermined threshold value. The PTC element 611, which

serves as a thermally sensitive resistor whose resistance increases as temperature rises, limits the amount of current by increasing the resistance, in order to prevent abnormal heat generation. Barium titanate ($BaTiO_3$)-based semiconductor ceramic can be used for the PTC element.

[0309]

Alternatively, as illustrated in FIG. 12C, a plurality of cylindrical secondary batteries 600 may be sandwiched between a conductive plate 613 and a conductive plate 614 to form a module 615. The plurality of cylindrical secondary batteries 600 may be connected in parallel, connected in series, or connected in series after being connected in parallel. With the module 615 including the plurality of cylindrical secondary batteries 600, large electric power can be extracted.

[0310]

FIG. 12D is a top view of the module 615. The conductive plate 613 is shown by a dotted line for clarity of the drawing. As illustrated in FIG. 12D, the module 615 may include a wiring 616 which electrically connects the plurality of cylindrical secondary batteries 600 to each other. It is possible to provide the conductive plate 613 over the wiring 616 to overlap with each other. In addition, a temperature control device 617 may be provided between the plurality of cylindrical secondary batteries 600. When the cylindrical secondary batteries 600 are overheated, the temperature control device 617 can cool them, and when the cylindrical secondary batteries 600 are cooled too much, the temperature control device 617 can heat them. Thus, the performance of the module 615 is not easily influenced by the outside air temperature.

[0311]

When the positive electrode active material described in the above embodiment is used in the positive electrode 604, the cylindrical secondary battery 600 with high capacity and excellent cycle characteristics can be obtained.

[0312]

[Structural examples of secondary battery]

Other structural examples of secondary batteries will be described with reference to FIGS. 13A and 13B, FIGS. 14A-1, 14A-2, 14B-1, and 14B-2, FIGS. 15A and 15B, and FIG. 16.

[0313]

FIGS. 13A and 13B are external views of a secondary battery. The secondary battery includes a circuit board 900 and a secondary battery 913. A label 910 is attached to the secondary battery 913. As shown in FIG. 13B, the secondary battery further includes a terminal 951, a terminal 952, an antenna 914, and an antenna 915.

[0314]

The circuit board 900 includes terminals 911 and a circuit 912. The terminals 911 are connected to the terminals 951 and 952, the antennas 914 and 915, and the circuit 912. Note that

a plurality of terminals 911 serving as a control signal input terminal, a power supply terminal, and the like may be provided.

[0315]

The circuit 912 may be provided on the rear surface of the circuit board 900. The shape of each of the antennas 914 and 915 is not limited to a coil shape and may be a linear shape or a plate shape. Furthermore, a planar antenna, an aperture antenna, a traveling-wave antenna, an EH antenna, a magnetic-field antenna, a dielectric antenna, or the like may be used. Alternatively, the antenna 914 or the antenna 915 may be a flat-plate conductor. The flat-plate conductor can serve as one of conductors for electric field coupling. That is, the antenna 914 or the antenna 915 can serve as one of two conductors of a capacitor. Thus, electric power can be transmitted and received not only by an electromagnetic field or a magnetic field but also by an electric field.

[0316]

The line width of the antenna 914 is preferably larger than that of the antenna 915. This makes it possible to increase the amount of electric power received by the antenna 914.

[0317]

The secondary battery includes a layer 916 between the secondary battery 913 and the antennas 914 and 915. The layer 916 has a function of blocking an electromagnetic field from the secondary battery 913, for example. As the layer 916, for example, a magnetic body can be used.

[0318]

Note that the structure of the secondary battery is not limited to that shown in FIGS. 13A and 13B.

[0319]

For example, as shown in FIGS. 14A-1 and 14A-2, two opposite surfaces of the secondary battery 913 in FIGS. 13A and 13B may be provided with respective antennas. FIG. 14A-1 is an external view showing one side of the opposite surfaces, and FIG. 14A-2 is an external view showing the other side of the opposite surfaces. For portions similar to those in FIGS. 13A and 13B, a description of the secondary battery illustrated in FIGS. 13A and 13B can be referred to as appropriate.

[0320]

As illustrated in FIG. 14A-1, the antenna 914 is provided on one of the opposite surfaces of the secondary battery 913 with the layer 916 located therebetween, and as illustrated in FIG. 14A-2, an antenna 918 is provided on the other of the opposite surfaces of the secondary battery 913 with a layer 917 located therebetween. The layer 917 has a function of blocking an electromagnetic field from the secondary battery 913, for example. As the layer 917, for example,

a magnetic body can be used.

[0321]

With the above structure, both of the antennas 914 and 918 can be increased in size. The antenna 918 has a function of communicating data with an external device, for example. An antenna with a shape that can be applied to the antenna 914, for example, can be used as the antenna 918. As a system for communication using the antenna 918 between the secondary battery and another device, a response method that can be used between the secondary battery and another device, such as NFC, can be employed.

[0322]

Alternatively, as illustrated in FIG. 14B-1, the secondary battery 913 in FIGS. 13A and 13B may be provided with a display device 920. The display device 920 is electrically connected to the terminal 911. It is possible that the label 910 is not provided in a portion where the display device 920 is provided. For portions similar to those in FIGS. 13A and 13B, a description of the secondary battery illustrated in FIGS. 13A and 13B can be referred to as appropriate.

[0323]

The display device 920 can display, for example, an image showing whether charge is being carried out, an image showing the amount of stored power, or the like. As the display device 920, electronic paper, a liquid crystal display device, an electroluminescent (EL) display device, or the like can be used. For example, the use of electronic paper can reduce power consumption of the display device 920.

[0324]

Alternatively, as illustrated in FIG. 14B-2, the secondary battery 913 illustrated in FIGS. 13A and 13B may be provided with a sensor 921. The sensor 921 is electrically connected to the terminal 911 via a terminal 922. For portions similar to those in FIGS. 13A and 13B, a description of the secondary battery illustrated in FIGS. 13A and 13B can be referred to as appropriate.

[0325]

The sensor 921 has a function of measuring, for example, displacement, position, speed, acceleration, angular velocity, rotational frequency, distance, light, liquid, magnetism, temperature, chemical substance, sound, time, hardness, electric field, electric current, voltage, electric power, radiation, flow rate, humidity, gradient, oscillation, odor, or infrared rays. With the sensor 921, for example, data on an environment (e.g., temperature) where the secondary battery is placed can be determined and stored in a memory inside the circuit 912.

[0326]

Furthermore, structural examples of the secondary battery 913 will be described with

reference to FIGS. 15A and 15B and FIG. 16.

[0327]

The secondary battery 913 illustrated in FIG. 15A includes a wound body 950 provided with the terminals 951 and 952 inside a housing 930. The wound body 950 is soaked in an electrolyte solution inside the housing 930. The terminal 952 is in contact with the housing 930. An insulator or the like inhibits contact between the terminal 951 and the housing 930. Note that in FIG. 15A, the housing 930 divided into two pieces is illustrated for convenience; however, in the actual structure, the wound body 950 is covered with the housing 930 and the terminals 951 and 952 extend to the outside of the housing 930. For the housing 930, a metal material (such as aluminum) or a resin material can be used.

[0328]

Note that as illustrated in FIG. 15B, the housing 930 in FIG. 15A may be formed using a plurality of materials. For example, in the secondary battery 913 in FIG. 15B, a housing 930a and a housing 930b are bonded to each other, and the wound body 950 is provided in a region surrounded by the housing 930a and the housing 930b.

[0329]

For the housing 930a, an insulating material such as an organic resin can be used. In particular, when a material such as an organic resin is used for the side on which an antenna is formed, blocking of an electric field from the secondary battery 913 can be inhibited. When an electric field is not significantly blocked by the housing 930a, an antenna such as the antennas 914 and 915 may be provided inside the housing 930a. For the housing 930b, a metal material can be used, for example.

[0330]

FIG. 16 illustrates the structure of the wound body 950. The wound body 950 includes a negative electrode 931, a positive electrode 932, and separators 933. The wound body 950 is obtained by winding a sheet of a stack in which the negative electrode 931 overlaps with the positive electrode 932 with the separator 933 provided therebetween. Note that a plurality of stacks each including the negative electrode 931, the positive electrode 932, and the separator 933 may be stacked.

[0331]

The negative electrode 931 is connected to the terminal 911 in FIGS. 13A and 13B via one of the terminals 951 and 952. The positive electrode 932 is connected to the terminal 911 in FIGS. 13A and 13B via the other of the terminals 951 and 952.

[0332]

When the positive electrode active material described in the above embodiment is used

in the positive electrode 932, the secondary battery 913 with high capacity and excellent cycle characteristics can be obtained.

[0333]

[Laminated secondary battery]

Next, an example of a laminated secondary battery will be described with reference to FIGS. 17A to 17C, FIG. 18A and 18B, FIG. 19, FIG. 20, FIGS. 21A to 21C, FIGS. 22A, 22B1, 22B2, 22C, and 22D, and FIGS. 23A and 23B. When the laminated secondary battery has flexibility and is used in an electronic device at least part of which is flexible, the secondary battery can be bent as the electronic device is bent.

[0334]

A laminated secondary battery 980 is described with reference to FIGS. 17A to 17C. The laminated secondary battery 980 includes a wound body 993 illustrated in FIG. 17A. The wound body 993 includes a negative electrode 994, a positive electrode 995, and a separator 996. The wound body 993 is, like the wound body 950 illustrated in FIG. 16, obtained by winding a sheet of a stack in which the negative electrode 994 overlaps with the positive electrode 995 with the separator 996 therebetween.

[0335]

Note that the number of stacks each including the negative electrode 994, the positive electrode 995, and the separator 996 may be determined as appropriate depending on capacity and an element volume which are required. The negative electrode 994 is connected to a negative electrode current collector (not illustrated) via one of a lead electrode 997 and a lead electrode 998. The positive electrode 995 is connected to a positive electrode current collector (not illustrated) via the other of the lead electrode 997 and the lead electrode 998.

[0336]

As illustrated in FIG. 17B, the wound body 993 is packed in a space formed by bonding a film 981 and a film 982 having a depressed portion that serve as exterior bodies by thermocompression bonding or the like, whereby the laminated secondary battery 980 can be formed as illustrated in FIG. 17C. The wound body 993 includes the lead electrode 997 and the lead electrode 998, and is soaked in an electrolyte solution inside a space surrounded by the film 981 and the film 982 having a depressed portion.

[0337]

For the film 981 and the film 982 having a depressed portion, a metal material such as aluminum or a resin material can be used, for example. With the use of a resin material for the film 981 and the film 982 having a depressed portion, the film 981 and the film 982 having a depressed portion can be changed in their forms when external force is applied; thus, a flexible

secondary battery can be fabricated.

[0338]

Although FIGS. 17B and 17C illustrate an example where a space is formed by two films, the wound body 993 may be placed in a space formed by bending one film.

[0339]

When the positive electrode active material described in the above embodiment is used in the positive electrode 995, the laminated secondary battery 980 with high capacity and excellent cycle characteristics can be obtained.

[0340]

In FIGS. 17A to 17C, an example in which the laminated secondary battery 980 includes a wound body in a space formed by films serving as exterior bodies is described; however, as illustrated in FIGS. 18A and 18B, a secondary battery may include a plurality of strip-shaped positive electrodes, a plurality of strip-shaped separators, and a plurality of strip-shaped negative electrodes in a space formed by films serving as exterior bodies, for example.

[0341]

A laminated secondary battery 500 illustrated in FIG. 18A includes a positive electrode 503 including a positive electrode current collector 501 and a positive electrode active material layer 502, a negative electrode 506 including a negative electrode current collector 504 and a negative electrode active material layer 505, a separator 507, an electrolyte solution 508, and an exterior body 509. The separator 507 is provided between the positive electrode 503 and the negative electrode 506 in the exterior body 509. The exterior body 509 is filled with the electrolyte solution 508. The electrolyte solution described in Embodiment 2 can be used for the electrolyte solution 508.

[0342]

In the laminated secondary battery 500 illustrated in FIG. 18A, the positive electrode current collector 501 and the negative electrode current collector 504 also serve as terminals for an electrical contact with an external portion. For this reason, the positive electrode current collector 501 and the negative electrode current collector 504 may be arranged so as to be partly exposed to the outside of the exterior body 509. Alternatively, a lead electrode and the positive electrode current collector 501 or the negative electrode current collector 504 may be bonded to each other by ultrasonic welding, and instead of the positive electrode current collector 501 and the negative electrode current collector 504, the lead electrode may be exposed to the outside of the exterior body 509.

[0343]

As the exterior body 509 of the laminated secondary battery 500, for example, a laminate

film having a three-layer structure can be employed in which a highly flexible metal thin film of aluminum, stainless steel, copper, nickel, or the like is provided over a film formed of a material such as polyethylene, polypropylene, polycarbonate, ionomer, or polyamide, and an insulating synthetic resin film of a polyamide-based resin, a polyester-based resin, or the like is provided over the metal thin film as the outer surface of the exterior body.

[0344]

FIG. 18B illustrates an example of a cross-sectional structure of the laminated secondary battery 500. Although FIG. 18A illustrates an example including only two current collectors for simplicity, an actual battery includes a plurality of electrode layers.

[0345]

The example in FIG. 18B includes 16 electrode layers. The laminated secondary battery 500 has flexibility even though including 16 electrode layers. FIG. 18B illustrates a structure including 8 layers of negative electrode current collectors 504 and 8 layers of positive electrode current collectors 501, i.e., 16 layers in total. Note that FIG. 18B illustrates a cross section of the lead portion of the negative electrode, and the 8 negative electrode current collectors 504 are bonded to each other by ultrasonic welding. It is needless to say that the number of electrode layers is not limited to 16, and may be more than 16 or less than 16. With a large number of electrode layers, the secondary battery can have high capacity. In contrast, with a small number of electrode layers, the secondary battery can have small thickness and high flexibility.

[0346]

FIGS. 19 and 20 each illustrate an example of the external view of the laminated secondary battery 500. In FIGS. 19 and 20, the positive electrode 503, the negative electrode 506, the separator 507, the exterior body 509, a positive electrode lead electrode 510, and a negative electrode lead electrode 511 are included.

[0347]

FIG. 21A illustrates external views of the positive electrode 503 and the negative electrode 506. The positive electrode 503 includes the positive electrode current collector 501, and the positive electrode active material layer 502 is formed on a surface of the positive electrode current collector 501. The positive electrode 503 also includes a region where the positive electrode current collector 501 is partly exposed (hereinafter referred to as a tab region). The negative electrode 506 includes the negative electrode current collector 504, and the negative electrode active material layer 505 is formed on a surface of the negative electrode current collector 504. The negative electrode 506 also includes a region where the negative electrode current collector 504 is partly exposed, that is, a tab region. The areas and the shapes of the tab regions included in the positive electrode and the negative electrode are not limited to those illustrated in

FIG. 21A.

[0348]

[Method for manufacturing laminated secondary battery]

Here, an example of a method for manufacturing the laminated secondary battery whose external view is illustrated in FIG. 19 will be described with reference to FIGS. 21B and 21C.

[0349]

First, the negative electrode 506, the separator 507, and the positive electrode 503 are stacked. FIG. 21B illustrates a stack including the negative electrode 506, the separator 507, and the positive electrode 503. The secondary battery described here as an example includes 5 negative electrodes and 4 positive electrodes. Next, the tab regions of the positive electrodes 503 are bonded to each other, and the tab region of the positive electrode on the outermost surface and the positive electrode lead electrode 510 are bonded to each other. The bonding can be performed by ultrasonic welding, for example. In a similar manner, the tab regions of the negative electrodes 506 are bonded to each other, and the negative electrode lead electrode 511 is bonded to the tab region of the negative electrode on the outermost surface.

[0350]

After that, the negative electrode 506, the separator 507, and the positive electrode 503 are placed over the exterior body 509.

[0351]

Subsequently, the exterior body 509 is folded along a dashed line as illustrated in FIG. 21C. Then, the outer edge of the exterior body 509 is bonded. The bonding can be performed by thermocompression bonding, for example. At this time, a part (or one side) of the exterior body 509 is left unbonded (to provide an inlet) so that the electrolyte solution 508 can be introduced later.

[0352]

Next, the electrolyte solution 508 is introduced into the exterior body 509 from the inlet of the exterior body 509. The electrolyte solution 508 is preferably introduced in a reduced pressure atmosphere or in an inert gas atmosphere. Lastly, the inlet is bonded. In the above manner, the laminated secondary battery 500 can be manufactured.

[0353]

When the positive electrode active material described in the above embodiment is used in the positive electrode 503, the laminated secondary battery 500 with high capacity and excellent cycle characteristics can be obtained.

[0354]

[Bendable secondary battery]

Next, an example of a bendable secondary battery is described with reference to FIGS. 22A, 22B1, 22B2, 22C and 22D and FIGS. 23A and 23B.

[0355]

FIG. 22A is a schematic top view of a bendable secondary battery 50. FIGS. 22B1, 22B2, and 22C are schematic cross-sectional views taken along cutting line C1-C2, cutting line C3-C4, and cutting line A1-A2, respectively, in FIG. 22A. The battery 50 includes an exterior body 51 and a positive electrode 11a, and a negative electrode 11b held in the exterior body 51. A lead 12a electrically connected to the positive electrode 11a and a lead 12b electrically connected to the negative electrode 11b are extended to the outside of the exterior body 51. In addition to the positive electrode 11a and the negative electrode 11b, an electrolyte solution (not illustrated) is enclosed in a region surrounded by the exterior body 51.

[0356]

FIGS. 23A and 23B illustrate the positive electrode 11a and the negative electrode 11b included in the battery 50. FIG. 23A is a perspective view illustrating the stacking order of the positive electrode 11a, the negative electrode 11b, and the separator 14. FIG. 23B is a perspective view illustrating the lead 12a and the lead 12b in addition to the positive electrode 11a and the negative electrode 11b.

[0357]

As illustrated in FIG. 23A, the battery 50 includes a plurality of strip-shaped positive electrodes 11a, a plurality of strip-shaped negative electrodes 11b, and a plurality of separators 14. The positive electrode 11a and the negative electrode 11b each include a projected tab portion and a portion other than the tab. A positive electrode active material layer is formed on one surface of the positive electrode 11a other than the tab portion, and a negative electrode active material layer is formed on one surface of the negative electrode 11b other than the tab portion.

[0358]

The positive electrodes 11a and the negative electrodes 11b are stacked so that surfaces of the positive electrodes 11a on each of which the positive electrode active material layer is not formed are in contact with each other and that surfaces of the negative electrodes 11b on each of which the negative electrode active material layer is not formed are in contact with each other.

[0359]

Furthermore, the separator 14 is provided between the surface of the positive electrode 11a on which the positive electrode active material is formed and the surface of the negative electrode 11b on which the negative electrode active material is formed. In FIG. 23A, the separator 14 is shown by a dotted line for easy viewing.

[0360]

In addition, as illustrated in FIG. 23B, the plurality of positive electrodes 11a are electrically connected to the lead 12a in a bonding portion 15a. The plurality of negative electrodes 11b are electrically connected to the lead 12b in a bonding portion 15b.

[0361]

Next, the exterior body 51 is described with reference to FIGS. 22B1, 22B2, 22C, and 22D.

[0362]

The exterior body 51 has a film-like shape and is folded in half with the positive electrodes 11a and the negative electrodes 11b between facing portions of the exterior body 51. The exterior body 51 includes a folded portion 61, a pair of seal portions 62, and a seal portion 63. The pair of seal portions 62 is provided with the positive electrodes 11a and the negative electrodes 11b positioned therebetween and thus can also be referred to as side seals. The seal portion 63 has portions overlapping with the lead 12a and the lead 12b and can also be referred to as a top seal.

[0363]

Part of the exterior body 51 that overlaps with the positive electrodes 11a and the negative electrodes 11b preferably has a wave shape in which crest lines 71 and trough lines 72 are alternately arranged. The seal portions 62 and the seal portion 63 of the exterior body 51 are preferably flat.

[0364]

FIG. 22B1 shows a cross section cut along the part overlapping with the crest line 71. FIG. 22B2 shows a cross section cut along the part overlapping with the trough line 72. FIGS. 22B1 and 22B2 correspond to cross sections of the battery 50, the positive electrodes 11a, and the negative electrodes 11b in the width direction.

[0365]

The distance between an end portion of the negative electrode 11b in the width direction and the seal portion 62 is referred to as a distance La. When the battery 50 changes in shape, for example, is bent, the positive electrode 11a and the negative electrode 11b change in shape such that the positions thereof are shifted from each other in the length direction as described later. At the time, if the distance La is too short, the exterior body 51 and the positive electrode 11a and the negative electrode 11b are rubbed hard against each other, so that the exterior body 51 is damaged in some cases. In particular, when a metal film of the exterior body 51 is exposed, there is concern that the metal film is corroded by the electrolyte solution. Thus, the distance La is preferably set as long as possible. However, if the distance La is too long, the volume of the battery 50 is increased.

[0366]

The distance La between the end portion of the negative electrode 11b and the seal portion 62 is preferably increased as the total thickness of the stacked positive electrodes 11a and negative electrodes 11b is increased.

[0367]

Specifically, when the total thickness of the stacked positive electrodes 11a and negative electrodes 11b and the separators 214 is referred to as a thickness t, the distance La is preferably 0.8 times or more and 3.0 times or less, further preferably 0.9 times or more and 2.5 times or less, still further preferably 1.0 times or more and 2.0 times or less as large as the thickness t. When the distance La is in the above-described range, a compact battery which is highly reliable for bending can be obtained.

[0368]

Furthermore, when a distance between the pair of seal portions 62 is referred to as a distance Lb, it is preferable that the distance Lb be sufficiently longer than a width Wb of the negative electrode 11b. In this case, even when the positive electrode 11a and the negative electrode 11b come into contact with the exterior body 51 by change in the shape of the battery 50 such as repeated bending, the position of part of the positive electrode 11a and the negative electrode 11b can be shifted in the width direction; thus, the positive and negative electrodes 11a and 11b and the exterior body 51 can be effectively prevented from being rubbed against each other.

[0369]

For example, the difference between the distance Lb (i.e., the distance between the pair of seal portions 62) and the width Wb of the negative electrode 11b is preferably 1.6 times or more and 6.0 times or less, further preferably 1.8 times or more and 5.0 times or less, still further preferably 2.0 times or more and 4.0 times or less as large as the total thickness t of the positive electrode 11a and the negative electrode 11b.

[0370]

In other words, the distance Lb, the width Wb, and the thickness t preferably satisfy the relation of the following Formula 2.

[0371]

[Formula 2]

$$\frac{Lb-Wb}{2t} \geq a \qquad \text{(Formula 2)}$$

[0372]

In the formula, a is 0.8 or more and 3.0 or less, preferably 0.9 or more and 2.5 or less, further preferably 1.0 or more and 2.0 or less.

[0373]

FIG. 22C illustrates a cross section including the lead 12a and corresponds to a cross section of the battery 50, the positive electrode 11a, and the negative electrode 11b in the length direction. As illustrated in FIG. 22C, a space 73 is preferably provided between end portions of the positive electrode 11a and the negative electrode 11b in the length direction and the exterior body 51 in the folded portion 61.

[0374]

FIG. 22D is a schematic cross-sectional view of the battery 50 in a state of being bent. FIG. 22D corresponds to a cross section along cutting line B1-B2 in FIG. 22A.

[0375]

When the battery 50 is bent, a part of the exterior body 51 positioned on the outer side in bending is unbent and the other part positioned on the inner side changes its shape as it shrinks. More specifically, the part of the exterior body 51 positioned on the outer side in bending changes its shape such that the wave amplitude becomes smaller and the length of the wave period becomes larger. In contrast, the part of the exterior body 51 positioned on the inner side in bending changes its shape such that the wave amplitude becomes larger and the length of the wave period becomes smaller. When the exterior body 51 changes its shape in this manner, stress applied to the exterior body 51 due to bending is relieved, so that a material itself that forms the exterior body 51 does not need to expand and contract. As a result, the battery 50 can be bent with weak force without damage to the exterior body 51.

[0376]

Furthermore, as illustrated in FIG. 22D, when the battery 50 is bent, the positions of the positive electrode 11a and the negative electrode 11b are shifted relatively. At this time, ends of the stacked positive electrodes 11a and negative electrodes 11b on the seal portion 63 side are fixed by the fixing member 17. Thus, the plurality of positive electrodes 11a and the plurality of negative electrodes 11b are more shifted at a position closer to the folded portion 61. Therefore, stress applied to the positive electrode 11a and the negative electrode 11b is relieved, and the positive electrode 11a and the negative electrode 11b themselves do not need to expand and contract. As a result, the battery 50 can be bent without damage to the positive electrode 11a and the negative electrode 11b.

[0377]

Furthermore, the space 73 is provided between the end portions of the positive and negative electrodes 11a and 11b and the exterior body 51, whereby the relative positions of the positive electrode 11a and the negative electrode 11b can be shifted while the end portions of the

positive electrode 11a and the negative electrode 11b located on an inner side when the battery 50 is bent do not contact the exterior body 51.

[0378]

In the battery 50 illustrated in FIGS. 22A, 22B1, 22B2, 22C and 22D and FIGS. 23A and 23B, the exterior body, the positive electrode 11a, and the negative electrode 11b are less likely to be damaged and the battery characteristics are less likely to deteriorate even when the battery 50 is repeatedly bent and unbent. When the positive electrode active material described in the above embodiment is used for the positive electrode 11a included in the battery 50, a battery with more excellent cycle characteristics can be obtained.

[0379]

(Embodiment 4)

In this embodiment, examples of electronic devices including the secondary battery of one embodiment of the present invention are described.

[0380]

First, FIGS. 24A to 24G show examples of electronic devices including the bendable secondary battery described in Embodiment 3. Examples of an electronic device including a flexible secondary battery include television sets (also referred to as televisions or television receivers), monitors of computers or the like, digital cameras or digital video cameras, digital photo frames, mobile phones (also referred to as cellular phones or mobile phone devices), portable game machines, portable information terminals, audio reproducing devices, and large game machines such as pachinko machines.

[0381]

In addition, a flexible secondary battery can be incorporated along a curved inside/outside wall surface of a house or a building or a curved interior/exterior surface of an automobile.

[0382]

FIG. 24A illustrates an example of a mobile phone. A mobile phone 7400 is provided with a display portion 7402 incorporated in a housing 7401, an operation button 7403, an external connection port 7404, a speaker 7405, a microphone 7406, and the like. Note that the mobile phone 7400 includes a secondary battery 7407. When the secondary battery of one embodiment of the present invention is used as the secondary battery 7407, a lightweight mobile phone with a long lifetime can be provided.

[0383]

FIG. 24B illustrates the mobile phone 7400 that is bent. When the whole mobile phone 7400 is curved by external force, the secondary battery 7407 included in the mobile phone 7400 is also curved. FIG. 24C illustrates the curved secondary battery 7407. The secondary battery

7407 is a thin storage battery. The secondary battery 7407 is curved and fixed. Note that the secondary battery 7407 includes a lead electrode 7408 electrically connected to a current collector 7409. The current collector 7409 is, for example, copper foil, and partly alloyed with gallium; thus, adhesion between the current collector 7409 and an active material layer in contact with the current collector 7409 is improved and the secondary battery 7407 can have high reliability even in a state of being bent.

[0384]

FIG. 24D illustrates an example of a bangle display device. A portable display device 7100 includes a housing 7101, a display portion 7102, an operation button 7103, and a secondary battery 7104. FIG. 24E illustrates the bent secondary battery 7104. When the curved secondary battery 7104 is on a user's arm, the housing changes its form and the curvature of a part or the whole of the secondary battery 7104 is changed. Note that the radius of curvature of a curve at a point refers to the radius of the circular arc that best approximates the curve at that point. The reciprocal of the radius of curvature is curvature. Specifically, part or the whole of the housing or the main surface of the secondary battery 7104 is changed in the range of radius of curvature from 40 mm to 150 mm. When the radius of curvature at the main surface of the secondary battery 7104 is greater than or equal to 40 mm and less than or equal to 150 mm, the reliability can be kept high. When the secondary battery of one embodiment of the present invention is used as the secondary battery 7104, a lightweight portable display device with a long lifetime can be provided.

[0385]

FIG. 24F illustrates an example of a watch-type portable information terminal. A portable information terminal 7200 includes a housing 7201, a display portion 7202, a band 7203, a buckle 7204, an operation button 7205, an input output terminal 7206, and the like.

[0386]

The portable information terminal 7200 is capable of executing a variety of applications such as mobile phone calls, e-mailing, viewing and editing texts, music reproduction, Internet communication, and a computer game.

[0387]

The display surface of the display portion 7202 is curved, and images can be displayed on the curved display surface. In addition, the display portion 7202 includes a touch sensor, and operation can be performed by touching the screen with a finger, a stylus, or the like. For example, by touching an icon 7207 displayed on the display portion 7202, application can be started.

[0388]

With the operation button 7205, a variety of functions such as time setting, power on/off,

on/off of wireless communication, setting and cancellation of a silent mode, and setting and cancellation of a power saving mode can be performed. For example, the functions of the operation button 7205 can be set freely by setting the operation system incorporated in the portable information terminal 7200.

[0389]

The portable information terminal 7200 can employ near field communication that is a communication method based on an existing communication standard. For example, mutual communication between the portable information terminal 7200 and a headset capable of wireless communication can be performed, and thus hands-free calling is possible.

[0390]

Moreover, the portable information terminal 7200 includes the input output terminal 7206, and data can be directly transmitted to and received from another information terminal via a connector. In addition, charging via the input output terminal 7206 is possible. Note that the charging operation may be performed by wireless power feeding without using the input output terminal 7206.

[0391]

The display portion 7202 of the portable information terminal 7200 includes the secondary battery of one embodiment of the present invention. When the secondary battery of one embodiment of the present invention is used, a lightweight portable information terminal with a long lifetime can be provided. For example, the secondary battery 7104 illustrated in FIG. 24E that is in the state of being curved can be provided in the housing 7201. Alternatively, the secondary battery 7104 illustrated in FIG. 24E can be provided in the band 7203 such that it can be curved.

[0392]

A portable information terminal 7200 preferably includes a sensor. As the sensor, for example a human body sensor such as a fingerprint sensor, a pulse sensor, or a temperature sensor, a touch sensor, a pressure sensitive sensor, an acceleration sensor, or the like is preferably mounted.

[0393]

FIG. 24G illustrates an example of an armband display device. A display device 7300 includes a display portion 7304 and the secondary battery of one embodiment of the present invention. The display device 7300 can include a touch sensor in the display portion 7304 and can serve as a portable information terminal.

[0394]

The display surface of the display portion 7304 is bent, and images can be displayed on the bent display surface. A display state of the display device 7300 can be changed by, for

example, near field communication, which is a communication method based on an existing communication standard.

[0395]

The display device 7300 includes an input output terminal, and data can be directly transmitted to and received from another information terminal via a connector. In addition, charging via the input output terminal is possible. Note that the charging operation may be performed by wireless power feeding without using the input output terminal.

[0396]

When the secondary battery of one embodiment of the present invention is used as the secondary battery included in the display device 7300, a lightweight display device with a long lifetime can be provided.

[0397]

In addition, FIG. 24H, FIGS. 25A to 25C, and FIG. 26 show examples of electronic devices including the secondary battery with excellent cycle characteristics described in the above embodiment.

[0398]

When the secondary battery of one embodiment of the present invention is used as a secondary battery of a daily electronic device, a lightweight product with a long lifetime can be provided. As the daily electronic devices, an electric toothbrush, an electric shaver, electric beauty equipment, and the like are given. As secondary batteries of these products, in consideration of handling ease for users, small and lightweight stick type secondary batteries with high capacity are desired.

[0399]

FIG. 24H is a perspective view of a device which is called a vaporizer. In FIG. 24H, a vaporizer 7500 includes an atomizer 7501 including a heating element, a secondary battery 7504 supplying power to the atomizer, and a cartridge 7502 including a liquid supply bottle, a sensor, and the like. To improve safety, a protection circuit which prevents overcharge and overdischarge of the secondary battery 7504 may be electrically connected to the secondary battery 7504. The secondary battery 7504 in FIG. 24H includes an output terminal for connecting to a charger. When the vaporizer 7500 is held by a user, the secondary battery 7504 becomes a tip portion; thus, it is preferable that the secondary battery 7504 have a short total length and be lightweight. With the secondary battery of one embodiment of the present invention which has high capacity and excellent cycle characteristics, the small and lightweight vaporizer 7500 which can be used for a long time for a long period can be provided.

[0400]

Next, FIGS. 25A and 25B illustrate an example of a foldable tablet terminal. A tablet terminal 9600 illustrated in FIGS. 25A and 25B includes a housing 9630a, a housing 9630b, a movable portion 9640 connecting the housings 9630a and 9630b, a display portion 9631 including a display portion 9631a and a display portion 9631b, a display mode changing switch 9626, a power switch 9627, a power saving mode changing switch 9625, a fastener 9629, and an operation switch 9628. A flexible panel is used for the display portion 9631, whereby a tablet terminal with a larger display portion can be provided. FIG. 25A illustrates the tablet terminal 9600 that is opened, and FIG. 25B illustrates the tablet terminal 9600 that is closed.

[0401]

The tablet terminal 9600 includes a power storage unit 9635 inside the housings 9630a and 9630b. The power storage unit 9635 is provided across the housings 9630a and 9630b, passing through the movable portion 9640.

[0402]

Part of the display portion 9631a can be a touch panel region and data can be input when a displayed operation key is touched. Although a structure in which a half region in the display portion 9631a has only a display function and the other half region has a touch panel function is shown as an example, the display portion 9631a is not limited to the structure. The whole region in the display portion 9631a may have a touch panel function. For example, the display portion 9631a can display keyboard buttons in the whole region to be a touch panel, and the display portion 9631b can be used as a display screen.

[0403]

Like the display portion 9631a, part of the display portion 9631b can be a touch panel region. A switching button for showing/hiding a keyboard of the touch panel is touched with a finger, a stylus, or the like, so that keyboard buttons can be displayed on the display portion 9631b.

[0404]

Touch input can be performed in the touch panel region and the touch panel region at the same time.

[0405]

The display mode switch 9626 can switch the display between a portrait mode and a landscape mode, and between monochrome display and color display, for example. The power saving mode changing switch 9625 can control display luminance in accordance with the amount of external light in use of the tablet terminal 9600, which is measured with an optical sensor incorporated in the tablet terminal 9600. Another detection device including a sensor for detecting inclination, such as a gyroscope sensor or an acceleration sensor, may be incorporated in the tablet terminal, in addition to the optical sensor.

[0406]

Although the display portion 9631a and the display portion 9631b have the same area in FIG. 25A, one embodiment of the present invention is not limited to this example. The display portion 9631a and the display portion 9631b may have different areas or different display quality. For example, one display panel may be capable of higher-definition display than the other display panel.

[0407]

The tablet terminal is closed in FIG. 25B. The tablet terminal includes the housing 9630, a solar cell 9633, and a charge and discharge control circuit 9634 including a DCDC converter 9636. The power storage unit of one embodiment of the present invention is used as the power storage unit 9635.

[0408]

The tablet terminal 9600 can be folded such that the housings 9630a and 9630b overlap with each other when not in use. Thus, the display portions 9631a and 9631b can be protected, which increases the durability of the tablet terminal 9600. With the power storage unit 9635 including the secondary battery of one embodiment of the present invention which has high capacity and excellent cycle characteristics, the tablet terminal 9600 which can be used for a long time for a long period can be provided.

[0409]

The tablet terminal illustrated in FIGS. 25A and 25B can also have a function of displaying various kinds of data (e.g., a still image, a moving image, and a text image), a function of displaying a calendar, a date, or the time on the display portion, a touch-input function of operating or editing data displayed on the display portion by touch input, a function of controlling processing by various kinds of software (programs), and the like.

[0410]

The solar cell 9633, which is attached on the surface of the tablet terminal, supplies electric power to a touch panel, a display portion, an image signal processor, and the like. Note that the solar cell 9633 can be provided on one or both surfaces of the housing 9630 and the power storage unit 9635 can be charged efficiently. The use of a lithium-ion battery as the power storage unit 9635 brings an advantage such as reduction in size.

[0411]

The structure and operation of the charge and discharge control circuit 9634 illustrated in FIG. 25B will be described with reference to a block diagram in FIG. 25C. The solar cell 9633, the power storage unit 9635, the DC-DC converter 9636, a converter 9637, switches SW1 to SW3, and the display portion 9631 are illustrated in FIG. 25C, and the power storage unit 9635, the DC-

DC converter 9636, the converter 9637, and the switches SW1 to SW3 correspond to the charge and discharge control circuit 9634 in FIG. 25B.

[0412]

First, an example of the operation in the case where power is generated by the solar cell 9633 using external light is described. The voltage of electric power generated by the solar cell is raised or lowered by the DCDC converter 9636 to a voltage for charging the power storage unit 9635. When the power from the solar cell 9633 is used for the operation of the display portion 9631, the switch SW1 is turned on and the voltage of the power is raised or lowered by the converter 9637 to a voltage needed for operating the display portion 9631. When display on the display portion 9631 is not performed, the switch SW1 is turned off and the switch SW2 is turned on, so that the power storage unit 9635 can be charged.

[0413]

Note that the solar cell 9633 is described as an example of a power generation means; however, one embodiment of the present invention is not limited to this example. The power storage unit 9635 may be charged using another power generation means such as a piezoelectric element or a thermoelectric conversion element (Peltier element). For example, the power storage unit 9635 may be charged with a non-contact power transmission module that transmits and receives power wirelessly (without contact) to charge the battery or with a combination of other charging means.

[0414]

FIG. 26 illustrates other examples of electronic devices. In FIG. 26, a display device 8000 is an example of an electronic device including a secondary battery 8004 of one embodiment of the present invention. Specifically, the display device 8000 corresponds to a display device for TV broadcast reception and includes a housing 8001, a display portion 8002, speaker portions 8003, the secondary battery 8004, and the like. The secondary battery 8004 of one embodiment of the present invention is provided in the housing 8001. The display device 8000 can receive electric power from a commercial power supply. Alternatively, the display device 8000 can use electric power stored in the secondary battery 8004. Thus, the display device 8000 can operate with the use of the secondary battery 8004 of one embodiment of the present invention as an uninterruptible power supply even when electric power cannot be supplied from a commercial power supply due to power failure or the like.

[0415]

A semiconductor display device such as a liquid crystal display device, a light-emitting device in which a light-emitting element such as an organic EL element is provided in each pixel, an electrophoretic display device, a digital micromirror device (DMD), a plasma display panel

(PDP), or a field emission display (FED) can be used for the display portion 8002.

[0416]

Note that the display device includes, in its category, all of information display devices for personal computers, advertisement displays, and the like other than TV broadcast reception.

[0417]

In FIG. 26, an installation lighting device 8100 is an example of an electronic device using a secondary battery 8103 of one embodiment of the present invention. Specifically, the lighting device 8100 includes a housing 8101, a light source 8102, the secondary battery 8103, and the like. Although FIG. 26 illustrates the case where the secondary battery 8103 is provided in a ceiling 8104 on which the housing 8101 and the light source 8102 are installed, the secondary battery 8103 may be provided in the housing 8101. The lighting device 8100 can receive electric power from a commercial power supply. Alternatively, the lighting device 8100 can use electric power stored in the secondary battery 8103. Thus, the lighting device 8100 can operate with the use of the secondary battery 8103 of one embodiment of the present invention as an uninterruptible power supply even when electric power cannot be supplied from a commercial power supply due to power failure or the like.

[0418]

Note that although the installation lighting device 8100 provided in the ceiling 8104 is illustrated in FIG. 26 as an example, the secondary battery of one embodiment of the present invention can be used as an installation lighting device provided in, for example, a wall 8105, a floor 8106, a window 8107, or the like other than the ceiling 8104. Alternatively, the secondary battery can be used in a tabletop lighting device or the like.

[0419]

As the light source 8102, an artificial light source which emits light artificially by using power can be used. Specifically, an incandescent lamp, a discharge lamp such as a fluorescent lamp, and a light-emitting element such as an LED or an organic EL element are given as examples of the artificial light source.

[0420]

In FIG. 26, an air conditioner including an indoor unit 8200 and an outdoor unit 8204 is an example of an electronic device including a secondary battery 8203 of one embodiment of the present invention. Specifically, the indoor unit 8200 includes a housing 8201, an air outlet 8202, the secondary battery 8203, and the like. Although FIG. 26 illustrates the case where the secondary battery 8203 is provided in the indoor unit 8200, the secondary battery 8203 may be provided in the outdoor unit 8204. Alternatively, the secondary batteries 8203 may be provided in both the indoor unit 8200 and the outdoor unit 8204. The air conditioner can receive electric

power from a commercial power supply. Alternatively, the air conditioner can use electric power stored in the secondary battery 8203. Particularly in the case where the secondary batteries 8203 are provided in both the indoor unit 8200 and the outdoor unit 8204, the air conditioner can operate with the use of the secondary battery 8203 of one embodiment of the present invention as an uninterruptible power supply even when electric power cannot be supplied from a commercial power supply due to power failure or the like.

[0421]

Note that although the split-type air conditioner including the indoor unit and the outdoor unit is illustrated in FIG. 26 as an example, the secondary battery of one embodiment of the present invention can be used in an air conditioner in which the functions of an indoor unit and an outdoor unit are integrated in one housing.

[0422]

In FIG. 26, an electric refrigerator-freezer 8300 is an example of an electronic device using a secondary battery 8304 of one embodiment of the present invention. Specifically, the electric refrigerator-freezer 8300 includes a housing 8301, a refrigerator door 8302, a freezer door 8303, the secondary battery 8304, and the like. The secondary battery 8304 is provided in the housing 8301 in FIG. 26. The electric refrigerator-freezer 8300 can receive electric power from a commercial power supply. Alternatively, the electric refrigerator-freezer 8300 can use electric power stored in the secondary battery 8304. Thus, the electric refrigerator-freezer 8300 can operate with the use of the secondary battery 8304 of one embodiment of the present invention as an uninterruptible power supply even when electric power cannot be supplied from a commercial power supply due to power failure or the like.

[0423]

Note that among the electronic devices described above, a high-frequency heating apparatus such as a microwave oven and an electronic device such as an electric rice cooker require high power in a short time. The tripping of a circuit breaker of a commercial power source in use of electronic devices can be prevented by using the secondary battery of one embodiment of the present invention as an auxiliary power source for supplying power which cannot be supplied enough by a commercial power source.

[0424]

In addition, in a time period when electronic devices are not used, particularly when the proportion of the amount of power which is actually used to the total amount of power which can be supplied from a commercial power source (such a proportion referred to as a usage rate of power) is low, power can be stored in the secondary battery, whereby the usage rate of power can be reduced in a time period when the electronic devices are used. For example, in the case of the

electric refrigerator-freezer 8300, power can be stored in the secondary battery 8304 in night time when the temperature is low and the refrigerator door 8302 and the freezer door 8303 are not often opened and closed. On the other hand, in daytime when the temperature is high and the refrigerator door 8302 and the freezer door 8303 are frequently opened and closed, the secondary battery 8304 is used as an auxiliary power source; thus, the usage rate of power in daytime can be reduced.

[0425]

According to one embodiment of the present invention, the secondary battery can have excellent cycle characteristics and improve reliability. Furthermore, in accordance with one embodiment of the present invention, a secondary battery with high capacity can be obtained; thus, the secondary battery itself can be made more compact and lightweight as a result of improved characteristics of the secondary battery. Thus, the secondary battery of one embodiment of the present invention is used in the electronic device described in this embodiment, whereby a more lightweight electronic device with a longer lifetime can be obtained. This embodiment can be implemented in appropriate combination with any of the other embodiments.

[0426]

(Embodiment 5)

In this embodiment, examples of vehicles including the secondary battery of one embodiment of the present invention are described.

[0427]

The use of secondary batteries in vehicles enables production of next-generation clean energy vehicles such as hybrid electric vehicles (HEVs), electric vehicles (EVs), and plug-in hybrid electric vehicles (PHEVs).

[0428]

FIGS. 27A to 27C each illustrate an example of a vehicle using the secondary battery of one embodiment of the present invention. An automobile 8400 illustrated in FIG. 27A is an electric vehicle that runs on the power of an electric motor. Alternatively, the automobile 8400 is a hybrid electric vehicle capable of driving appropriately using either an electric motor or an engine. One embodiment of the present invention can provide a high-mileage vehicle. The automobile 8400 includes the secondary battery. As the secondary battery, the modules of the secondary batteries illustrated in FIGS. 12C and 12D may be arranged to be used in a floor portion in the automobile. Alternatively, a battery pack in which a plurality of secondary batteries each of which is illustrated in FIGS. 17A to 17C are combined may be placed in a floor portion in the automobile. The secondary battery is used not only for driving an electric motor 8406, but also for supplying electric power to a light-emitting device such as a headlight 8401 or a room light

(not illustrated).

[0429]

The secondary battery can also supply electric power to a display device of a speedometer, a tachometer, or the like included in the automobile 8400. Furthermore, the secondary battery can supply electric power to a semiconductor device included in the automobile 8400, such as a navigation system.

[0430]

FIG. 27B illustrates an automobile 8500 including the secondary battery. The automobile 8500 can be charged when the secondary battery is supplied with electric power through external charging equipment by a plug-in system, a contactless power feeding system, or the like. In FIG. 27B, secondary batteries 8024 and 8025 included in the automobile 8500 is charged with the use of a ground-based charging apparatus 8021 through a cable 8022. In charging, a given method such as CHAdeMO (registered trademark) or Combined Charging System may be employed as a charging method, the standard of a connector, or the like as appropriate. The ground-based charging apparatus 8021 may be a charging station provided in a commerce facility or a power source in a house. With the use of a plug-in technique, the secondary battery 8024 included in the automobile 8500 can be charged by being supplied with electric power from the outside, for example. The charging can be performed by converting AC electric power into DC electric power through a converter such as an AC-DC converter.

[0431]

Furthermore, although not illustrated, the vehicle may include a power receiving device so that it can be charged by being supplied with electric power from an above-ground power transmitting device in a contactless manner. In the case of the contactless power feeding system, by fitting a power transmitting device in a road or an exterior wall, charging can be performed not only when the electric vehicle is stopped but also when driven. In addition, the contactless power feeding system may be utilized to perform transmission and reception of electric power between vehicles. A solar cell may be provided in the exterior of the automobile to charge the secondary battery when the automobile stops or moves. To supply electric power in such a contactless manner, an electromagnetic induction method or a magnetic resonance method can be used.

[0432]

FIG. 27C shows an example of a motorcycle using the secondary battery of one embodiment of the present invention. A motor scooter 8600 illustrated in FIG. 27C includes a secondary battery 8602, side mirrors 8601, and indicators 8603. The secondary battery 8602 can supply electric power to the indicators 8603.

[0433]

Furthermore, in the motor scooter 8600 illustrated in FIG. 27C, the secondary battery 8602 can be held in a storage unit under seat 8604. The secondary battery 8602 can be held in the storage unit under seat 8604 even with a small size. The secondary battery 8602 is detachable; thus, the secondary battery 8602 is carried indoors when it is charged, and is stored before the motorcycle is driven.

[0434]

In accordance with one embodiment of the present invention, the secondary battery can have improved cycle characteristics and the capacity of the secondary battery can be increased. Thus, the secondary battery itself can be made more compact and lightweight. The compact and lightweight secondary battery contributes to a reduction in the weight of a vehicle, and thus increases the driving radius. Furthermore, the secondary battery included in the vehicle can be used as a power source for supplying electric power to products other than the vehicle. In such a case, the use of a commercial power source can be avoided at peak time of electric power demand, for example. If the use of a commercial power source can be avoided at peak time of electric power demand, the avoidance can contribute to energy saving and a reduction in carbon dioxide emissions. Moreover, if the cycle characteristics are excellent, the secondary battery can be used for a long period; thus, the use amount of rare metals such as cobalt can be reduced.

[0435]

This embodiment can be implemented in appropriate combination with the other embodiments.

[Example 1]

[0436]

In this example, the positive electrode active materials which are embodiments of the present invention are formed, and the observation results of the positive electrode active materials by STEM, the results of TEM images subjected to fast Fourier transform, and the analysis results obtained by energy dispersive X-ray spectroscopy (EDX) are described. In addition, the evaluation results of characteristics of secondary batteries containing the positive electrode active materials are described.

[0437]

[Formation of positive electrode active material]

<<Sample 01>>

In this example, a positive electrode active material of Sample 01, which contains lithium cobaltate as a composite oxide of lithium and a first transition metal contained in a first region, lithium titanate as an oxide of a second transition metal contained in a second region, and magnesium oxide as an oxide of a representative element contained in a third region, was formed.

[0438]

In this example, lithium cobalt oxide particles (C-20F, produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD.) were used as a starting material. Thus, in this example, Step 12 and Step 13 described in Embodiment 1 were omitted. Note that the above-described lithium cobalt oxide particles each have a particle diameter of approximately 20 μm, and contain fluorine, magnesium, calcium, sodium, silicon, sulfur, and phosphorus in a region which can be analyzed by XPS.

[0439]

Next, as Step 14, the lithium cobalt oxide particles containing magnesium and fluorine were coated with a material containing titanium by a sol-gel method. Specifically, TTIP was dissolved in isopropanol, and an isopropanol solution of TTIP was formed. Then, the lithium cobalt oxide particles were mixed into the solution so that TTIP to lithium cobalt oxide containing magnesium and fluorine was 0.01 ml/g.

[0440]

The above mixed solution was stirred with a magnetic stirrer for four hours, at 25 °C, and at a humidity of 90 %RH. Through the process, water in an atmosphere and TTIP caused hydrolysis and polycondensation reaction, and a layer containing titanium was formed on the surface of the lithium cobalt oxide particle containing magnesium and fluorine.

[0441]

The mixed solution which had been subjected to the above process was filtered to collect the residue. As a filter for filtration, Kiriyama filter paper (No. 4) was used.

[0442]

The collected residue was dried in a vacuum at 70 °C for one hour.

[0443]

Next, the lithium cobalt oxide particles coated with the material containing titanium was heated. With a muffle furnace, the heating was performed under the following conditions: the flow rate of dry air was 10 L/min; the temperature was 800 °C (the temperature rising rate was 200 °C/h); and the retention time was two hours. The dew point of the dry air is preferably lower than or equal to −109 °C.

[0444]

Then, the heated particles were cooled to room temperature. The time of decreasing temperature from the retention temperature to room temperature was 10 hours to 15 hours. After that, crushing treatment was performed. In the crushing treatment, the particles were made to pass through a sieve. The sieve has an aperture width of 53 μm.

[0445]

Lastly, the cooled particles were collected, and the positive electrode active material of Sample 01 was obtained.

[0446]

<<Sample 02>>

Sample 02 was formed as a comparative example by heating lithium cobalt oxide particles containing magnesium and fluorine without being coated with a material containing titanium.

[0447]

Lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-20F) were used as the lithium cobalt oxide particles containing magnesium and fluorine.

[0448]

The lithium cobalt oxide particles containing magnesium and fluorine were heated. The heating was performed under the following conditions: the temperature was 800 °C (the temperature rising rate was 200 °C/h); the retention time was two hours; and the flow rate of oxygen was 10 L/min.

[0449]

The heated particles were cooled and made to pass through a sieve like Sample 01 to obtain a positive electrode active material of Sample 02.

[0450]

It is probable that Sample 02 is a positive electrode active material which contains lithium cobalt oxide inside and includes a region containing magnesium in a superficial portion.

[0451]

<<Sample 03>>

Sample 03 was formed as a comparative example in the following manner: a region containing titanium was formed in lithium cobalt oxide particles which do not contain magnesium by a sol-gel method and then the lithium cobalt oxide particles were heated.

[0452]

Lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-10N) were used. In the lithium cobalt oxide particles, magnesium is not detected and fluorine is detected at approximately 1 atomic% by XPS.

[0453]

A region containing titanium was formed by a sol-gel method in the lithium cobalt oxide particles, and the lithium cobalt oxide particles were dried, heated, cooled, and made to pass through a sieve, like Sample 01. The obtained lithium cobalt oxide particles were used as a positive electrode active material of Sample 03.

[0454]

It is probable that Sample 03 is a positive electrode active material which contains lithium cobalt oxide inside and includes a region containing titanium in a superficial portion.

[0455]

<<Sample 04>>

For Sample 04, as a comparative example, lithium cobalt oxide particles were used as it is without being heated.

[0456]

Lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-10N) were used.

[0457]

Sample 04 is a positive electrode active material which does not have a coating layer.

[0458]

<<Sample 05>>

For Sample 05, as a comparative example, lithium cobalt oxide particles containing magnesium and fluorine were used as it is without being heated.

[0459]

Lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-20F) were used as the lithium cobalt oxide particles containing magnesium and fluorine. That is, Sample 05 was used as the same as the starting material of Sample 01.

[0460]

Table 1 shows the conditions of Sample 01 to Sample 05.

[0461]

[Table 1]

	Conditions
Sample 01	$LiCoO_2$+Mg+F, coated with a material containing Ti, heated
Sample 02	$LiCoO_2$+Mg+F, heated
Sample 03	$LiCoO_2$, coated with a material containing Ti, heated
Sample 04	$LiCoO_2$, not heated
Sample 05	$LiCoO_2$+Mg+F, not heated

[0462]

[STEM]

The obtained positive electrode active material of Sample 01 was observed by an electron microscope (JEM-ARM200F, manufactured by JEOL Ltd.) under the condition where the

acceleration voltage was 200 kV. FIG. 28 shows the obtained electron microscope image. As shown in FIG. 28, the positive electrode active material probably includes three different regions: the first region 101; the second region 102; and the third region 103. The third region 103 is observed as a region brighter than the first region 101 and the second region 102. Furthermore, crystal orientations of the first region 101 and the second region 102 are partly aligned with each other, and crystal orientations of the second region 102 and the third region 103 are partly aligned with each other.

[0463]

[STEM-FFT]

FIG. 29A1 shows a fast Fourier transform (FFT) image of a region 103FFT in the STEM image of FIG. 28. In FIG. 29A2, a center point O of FIG. 29A1 is shown by a cross, and bright points A, B, and C are each surrounded by a circle. Similarly, FIG. 29B1 shows an FFT image of a region 102FFT. In FIG. 29B2, a center point O of FIG. 29B1 is shown by a cross, and bright points A, B, and C are each surrounded by a circle. In addition, FIG. 29C1 shows an FFT image of a region 101FFT. In FIG. 29C2, a center point O of FIG. 29C1 is shown by a cross, and bright points A, B, and C are each surrounded by a circle.

[0464]

In FIG. 29A2, a distance *d* between the bright point A and the center point O is 0.256 nm, a distance *d* between the bright point B and the center point O is 0.241 nm, and a distance *d* between the bright point C and the center point O is 0.209 nm. In addition, ∠COA is 121°, ∠COB is 52°, and ∠AOB is 69°. From these results, the region 103FFT probably contains magnesium oxide (MgO, cubic crystal).

[0465]

Similarly, in FIG. 29B2, the distance *d* between the bright point A and the center point O is 0.238 nm, the distance *d* between the bright point B and the center point O is 0.225 nm, and the distance *d* between the bright point C and the center point O is 0.198 nm. In addition, ∠COA is 123°, ∠COB is 52°, and ∠AOB is 71°. From these results, the region 102FFT probably contains lithium titanate ($LiTiO_2$, cubic crystal).

[0466]

In FIG. 29C2, the distance *d* between the bright point A and the center point O is 0.240 nm, the distance *d* between the bright point B and the center point O is 0.235 nm, and the distance *d* between the bright point C and the center point O is 0.196 nm. In addition, ∠COA is 126°, ∠COB is 52°, and ∠AOB is 74°. From these results, the region 101FFT probably contains lithium cobaltate ($LiCoO_2$, rhombohedral).

[0467]

[EDX]

FIGS. 30A1, 30A2, 30B1, 30B2, 30C1, and 30C2 show a high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) image and element mapping images with EDX of the positive electrode active material of Sample 01. FIG. 30A1 shows a HAADF-STEM image, FIG. 30A2 shows a mapping image of oxygen atoms, FIG. 30B1 shows a mapping image of cobalt atoms, FIG. 30B2 shows a mapping image of fluorine atoms, FIG. 30C1 shows a mapping image of titanium atoms, and FIG. 30C2 shows a mapping image of magnesium atoms. Note that in EDX element mapping images in FIGS. 30A2, 30B1, 30B2, 30C1, and 30C2 and FIGS. 31A2, 31B1, 31B2, 31C1, and 31C2, a region where the number of elements is less than or equal to a lower limit of the detection is indicated in white, and as the number of elements is increased, the white region becomes black.

[0468]

As shown in FIGS. 30A2 and 30B1, it is found that the oxygen atoms and the cobalt atoms are distributed in the whole of the positive electrode active material particle. In contrast, as shown in FIGS. 30B2, 30C1, and 30C2, it is found that the fluorine atoms, the titanium atoms, and the magnesium atoms are unevenly distributed in a region close to the surface of the positive electrode active material.

[0469]

Next, FIGS. 31A1, 31A2, 31B1, 31B2, 31C1, and 31C2 show a HAADF-STEM image and element mapping images with EDX of the positive electrode active material of Sample 05, which is a comparative example. FIG. 31A1 shows a HAADF-STEM image, FIG. 31A2 shows a mapping image of oxygen atoms, FIG. 31B1 shows a mapping image of cobalt atoms, FIG. 31B2 shows a mapping image of fluorine atoms, FIG. 31C1 shows a mapping image of titanium atoms, and FIG. 31C2 shows a mapping image of magnesium atoms.

[0470]

As shown in FIGS. 31B2 and 31C2, it is found that, even in Sample 05 which is not heated, a certain amount of magnesium and fluorine is unevenly distributed in the vicinity of the surface.

[0471]

[EDX line analysis]

FIG. 32 shows results of line analysis with TEM-EDX performed on a cross section of the vicinity of the surface of the positive electrode active material of Sample 01. FIG. 32 is a graph showing data detected on a line connecting the outside of the positive electrode active material of Sample 01 to the inside of the positive electrode active material, and a distance of 0 nm indicates the outside of the positive electrode active material and a distance of 14 nm indicates the inside of the particle. With EDX, the analysis region tends to be large, so that elements not

only at a center of an electron beam irradiation region but also in a region around the center may be detected.

[0472]

As shown in FIG. 32, it is found that there are peaks of magnesium and titanium in the vicinity of the surface of the positive electrode active material of Sample 01, the distribution of magnesium is closer to the surface than the distribution of titanium is. It is also found that the peak of magnesium is closer to the surface than the peak of titanium is. In addition, it is probable that cobalt and oxygen are present from the outermost surface of the positive electrode active material particle.

[0473]

As shown in FIG. 32, fluorine is hardly detected. This is probably because fluorine, which is a light element, is difficult to detect with EDX.

[0474]

From the above STEM images, FFT images, element mapping images with EDX, and EDX line analysis, it is found that Sample 01 is a positive electrode active material of one embodiment of the present invention, which includes the first region containing lithium cobaltate, the second region containing lithium, titanium, cobalt, and oxygen, and the third region containing magnesium and oxygen. It is found that, in Sample 01, part of the second region and part of the third region overlap.

[0475]

In the graph of FIG. 32, the amount of detected oxygen is stable at a distance of 4 nm or more. Thus, in this example, the average value O_{ave} of the amount of detected oxygen in the stable region is obtained, and a distance x of the measurement point at which the measurement value closest to 0.5 O_{ave}, the value of 50 % of the average value O_{ave}, is obtained is assumed to be the surface of the positive electrode active material particle.

[0476]

In this example, the average value O_{ave} of the amount of detected oxygen in a range from a distance of 4 nm to a distance of 14 nm is 674.2. The x axis of the measurement point at which the measurement value closest to 337.1, which is 50 % of 674.2, is obtained indicates a distance of 1.71 nm. Thus, in this example, a distance of 1.71 nm in the graph of FIG. 32 is assumed to be the surface of the positive electrode active material particle.

[0477]

When the surface of the positive electrode active material particle is set at a distance of 1.71 nm in FIG. 32, the peak of magnesium and the peak of titanium are present at 0.72 nm and 1.00 nm, respectively, from the surface of the positive electrode active material particle.

[0478]

The concentration of magnesium is higher than or equal to 1/5 of the peak from the surface of the positive electrode active material particle to a distance of 4.42 nm, that is, to a region at 2.71 nm from the surface. The measurement value of magnesium is less than 1/5 of the peak at a distance of 4.57 nm or more, that is, at a depth of 2.86 nm or more from the surface of the positive electrode active material particle. Thus, it is found that in Sample 01, a region from the surface to a depth of 2.71 nm is the third region.

[0479]

Furthermore, the concentration of titanium is higher than or equal to 1/2 of the peak from a distance of 2.14 nm to a distance of 3.42 nm. Thus, it is found that a region from a depth of 0.43 nm to a depth of 1.71 nm from the surface of the positive electrode active material particle is the second region.

[0480]

Next, evaluation results of charge and discharge characteristics of secondary batteries which are fabricated using the positive electrode active materials of Sample 01 to Sample 05 formed in the above manner are described.

[0481]

[Fabrication of secondary batteries]

CR2032 coin-type secondary batteries (with a diameter of 20 mm and a height of 3.2 mm) were fabricated using the positive electrode active materials of Sample 01 to Sample 05 formed in the above manner.

[0482]

A positive electrode formed by applying slurry in which a positive electrode active material (LCO), acetylene black (AB), and polyvinylidene fluoride (PVDF) were mixed at a weight ratio of 95:2.5:2.5 to a current collector was used.

[0483]

A lithium metal was used for the counter electrode.

[0484]

As an electrolyte contained in an electrolyte solution, 1 mol/L lithium hexafluorophosphate ($LiPF_6$) was used, and as the electrolyte solution, a solution in which ethylene carbonate (EC) and diethyl carbonate (DEC) at a volume ratio of 3:7 and vinylene carbonate (VC) at a 2 weight% were mixed was used.

[0485]

A positive electrode can and a negative electrode can were formed of stainless steel (SUS).

[0486]

[Evaluation of charge and discharge characteristics]

Next, charge and discharge characteristics of the secondary batteries of Sample 01 and Sample 05 formed in the above manner were evaluated. The measurement temperature was 25 °C. Twenty cycles of charging and discharging were performed at 4.6 V (CCCV, 0.5 C, a cutoff current of 0.01 C) and 2.5 V (CC, 0.5 C), respectively. Here, 1C was set to 137 mA/g, which was a current value per weight of the positive electrode active material.

[0487]

FIG. 33 is a graph showing charge and discharge characteristics of the secondary battery using the positive electrode active material of Sample 01. FIG. 33 shows excellent charge and discharge characteristics with a wide plateau. In addition, results of 20 cycles of charging and discharging almost overlap, which means that the cycle characteristics are excellent.

[0488]

FIG. 34 is a graph showing charge and discharge characteristics of the secondary battery of Sample 05, which is a comparative example. In the initial cycles, excellent charge and discharge characteristics are shown; however, as indicated by arrows in FIG. 34, charge capacity and discharge capacity decrease with an increase in cycles.

[0489]

[Evaluation of cycle characteristics]

<<Charging at 4.4 V>>

The cycle characteristics of the secondary batteries of Sample 01 and Sample 05 charged at 4.4 V were evaluated. The measurement temperature was 25 °C. The charging was performed at 4.4 V (CCCV, 0.5 C, a cutoff current of 0.01 C), and the discharging was performed at 2.5 V (CC, 0.5 C).

[0490]

FIG. 35 is a graph showing the cycle characteristics of the secondary batteries charged at 4.4 V. In FIG. 35, a solid line and a dotted line indicate secondary batteries containing the positive electrode active materials of Sample 01 and Sample 05, respectively. As shown in FIG. 35, in the secondary battery containing Sample 01, an energy density retention rate is 99.5 % even after 50 cycles of charging and discharging were performed, which shows extremely excellent cycle characteristics. In the secondary battery containing Sample 05, an energy density retention rate is 94.3 % after 50 cycles were performed.

[0491]

<<Charging at 4.6 V>>

The cycle characteristics of the secondary batteries of Sample 01 to Sample 04 charged at 4.6 V were evaluated. The measurement temperature was 25 °C. The charging was

performed to 4.6 V (CCCV, 0.5 C, a cutoff current of 0.01 C), and the discharging was performed to 2.5 V (CC, 0.5 C).

[0492]

FIG. 36 is a graph showing the cycle characteristics charged at 4.6 V. As shown in FIG. 36, in the secondary battery containing Sample 01, which is the positive electrode active material of one embodiment of the present invention, an energy density retention rate is 94.1 % even after 50 cycles of charging and discharging were performed at a high voltage of 4.6 V, which shows extremely excellent cycle characteristics. On the other hand, the secondary batteries containing the positive electrode active materials of Sample 02, Sample 03, and Sample 04, which are comparative examples, are inferior to that of Sample 01, and in Sample 04, for example, energy density retention rate is 33.2 % after 50 cycles were performed.

[0493]

As described above, it is found that the positive electrode active material with the structure of one embodiment of the present invention can achieve an advantageous effect when charging and discharging is performed at a voltage higher than 4.4 V.

[Example 2]

[0494]

In this example, the positive electrode active materials which are embodiments of the present invention are formed, and results of analysis which is different from that in Example 1 are described. In addition, evaluation results of characteristics of secondary batteries containing the positive electrode active materials under conditions different from those in Example 1 are described.

[0495]

In this example, a positive electrode active material which contains lithium cobaltate as a composite oxide of lithium and a first transition metal contained in a first region, lithium titanate as an oxide of a second transition metal contained in a second region, and magnesium oxide as an oxide of a representative element contained in a third region, was formed.

[0496]

[Formation of positive electrode active material and fabrication of secondary battery]

<<Sample 06 and Sample 07>>

In this example, lithium cobalt oxide particles (C-20F, produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD.) were used as a starting material.

[0497]

Next, as Step 14, the lithium cobalt oxide particles were coated with titanium oxide by a sol-gel method and dried. Step 14 was performed in a manner similar to that in Example 1 except

that mixture was performed so that TTIP to lithium cobalt oxide was 0.004 ml/g. The lithium cobalt oxide particles which are coated with the titanium oxide and are not heated yet are referred to as Sample 06.

[0498]

Next, Sample 06, which is the lithium cobalt oxide particles coated with the titanium oxide, was heated. With a muffle furnace, the heating was performed at 800 °C in an oxygen atmosphere under the following conditions: the retention time was two hours; and the flow rate of oxygen was 10 L/min.

[0499]

Then, as in Example 1, the particles were cooled and collected to obtain the positive electrode active material. The heated positive electrode active material is referred to as Sample 07.

[0500]

[TEM-EDX]

Sample 06 and Sample 07, in particular, cracks generated in the particles and the vicinity of the cracks were subjected to analysis with TEM-EDX.

[0501]

First, results of TEM-EDX plane analysis of titanium are shown in FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2 and FIGS. 38A, 38B1, 38B2, 38C1, 38C2, 38D1, 38D2, 38E1, and 38E2.

[0502]

FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2 show TEM-EDX analysis results of Sample 06 before heating. FIG. 37A is a cross-sectional TEM image showing the surfaces of the particles and the crack portions. FIG. 37B1 and FIG. 37B2 show a HAADF-STEM image and a Ti mapping image of a region including the surface of the particle that is denoted by a circle marked with "1" in FIG. 37A, respectively. Similarly, FIG. 37C1 and FIG. 37C2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 20 nm from the surface in the crack portion that is denoted by a circle marked with "2" in FIG. 37A, respectively. FIG. 37D1 and FIG. 37D2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 500 nm from the surface in the crack portion that is denoted by a circle marked with "3" in FIG. 37A, respectively. FIG. 37E1 and FIG. 37E2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 1000 nm from the surface in the crack portion that is denoted by a circle marked with "4" in FIG. 37A, respectively. Note that in EDX element mapping images in FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2, FIGS. 38A, 38B1, 38B2, 38C1, 38C2,

38D1, 38D2, 38E1, and 38E2, FIGS. 39A, 39B1, 39B2, 39C1, 39C2, 39D1, 39D2, 39E1, and 39E2, and FIGS. 40A, 40B1, 40B2, 40C1, 40C2, 40D1, 40D2, 40E1, and 40E2, a region where the number of elements is less than or equal to a lower limit of the detection is indicated in white, and as the number of elements is increased, the white region becomes black.

[0503]

FIGS. 38A, 38B1, 38B2, 38C1, 38C2, 38D1, 38D2, 38E1, and 38E2 show TEM-EDX analysis results of Sample 07 after heating. FIG. 38A is a cross-sectional TEM image showing the surfaces of the particles and the crack portions. FIG. 38B1 and FIG. 38B2 show a HAADF-STEM image and a Ti mapping image of a region including the surface of the particle that is denoted by a circle marked with "1" in FIG. 38A, respectively. Similarly, FIG. 38C1 and FIG. 38C2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 20 nm from the surface in the crack portion that is denoted by a circle marked with "2" in FIG. 38A, respectively. FIG. 38D1 and FIG. 38D2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 500 nm from the surface in the crack portion that is denoted by a circle marked with "3" in FIG. 38A, respectively. FIG. 38E1 and FIG. 38E2 show a HAADF-STEM image and a Ti mapping image of a region at a depth of approximately 1000 nm from the surface in the crack portion that is denoted by a circle marked with "4" in FIG. 38A, respectively.

[0504]

As shown in FIGS. 37A, 37B1, 37B2, 37C1, 37C2, 37D1, 37D2, 37E1, and 37E2 and FIGS. 38A, 38B1, 38B2, 38C1, 38C2, 38D1, 38D2, 38E1, and 38E2, in Sample 06 before heating, segregation of titanium on the surfaces of the particles is observed; however, no segregation is observed in the crack portion. In contrast, in Sample 07 after heating, segregation of titanium is observed both on the surfaces of the particles and in the crack portion. That is, it is found that titanium is segregated at the surface of the crack portion by heating.

[0505]

Next, results of TEM-EDX plane analysis of magnesium are shown in FIGS. 39A, 39B1, 39B2, 39C1, 39C2, 39D1, 39D2, 39E1, and 39E2, and FIGS. 40A, 40B1, 40B2, 40C1, 40C2, 40D1, 40D2, 40E1, and 40E2.

[0506]

FIG. 39A is a cross-sectional TEM image of Sample 06, which is the same as FIG. 37A. FIGS. 39B1, 39C1, 39D1, and 39E1 are HAADF-STEM images, which are the same as FIGS. 37B1, 37C1, 37D1, and 37E1. FIG. 39B2 shows a Mg mapping image of a region which is the same as FIG. 39B1. FIG. 39C2 shows a Mg mapping image of a region which is the same as FIG. 39C1. FIG. 39D2 shows a Mg mapping image of a region which is the same as FIG. 39D1.

FIG. 39E2 shows a Mg mapping image of a region which is the same as FIG. 39E1.

[0507]

FIG. 40A is a cross-sectional TEM image of Sample 07, which is the same as FIG. 38A. FIGS. 40B1, 40C1, 40D1, and 40E1 are HAADF-STEM images, which are the same as FIGS. 38B1, 38C1, 38D1, and 38E1. FIG. 40B2 shows a Mg mapping image of a region which is the same as FIG. 40B1. FIG. 40C2 shows a Mg mapping image of a region which is the same as FIG. 40C1. FIG. 40D2 shows a Mg mapping image of a region which is the same as FIG. 40D1. FIG. 40E2 shows a Mg mapping image of a region which is the same as FIG. 40E1.

[0508]

As shown in FIGS. 39A, 39B1, 39B2, 39C1, 39C2, 39D1, 39D2, 39E1, and 39E2, and FIGS. 40A, 40B1, 40B2, 40C1, 40C2, 40D1, 40D2, 40E1, and 40E2, in Sample 06 before heating, segregation of magnesium is not observed on the surfaces of the particles or in the crack portion. In contrast, in Sample 07 after heating, segregation of magnesium is observed both on the surfaces of the particles and in the crack portion.

[0509]

Next, to quantify titanium and magnesium, EDX point analysis was performed on the regions indicated by circles marked with 1 to 6 in FIG. 37A and the regions indicated by circles marked with 1 to 6 in FIG. 38A. In each region, two points were measured.

[0510]

FIGS. 41A and 41B show results of EDX point analysis in an atomic ratio of titanium to cobalt. FIG. 41A shows results of Sample 06 before heating. Detection points 1 to 6 in FIG. 41A correspond to the regions indicated by circles marked with 1 to 6 in FIG. 37A. FIG. 41B shows results of Sample 07 after heating. Detection points 1 to 6 in FIG. 41B correspond to the regions indicated by circles marked with 1 to 6 in FIG. 38A.

[0511]

As shown in FIGS. 41A and 41B, in the crack portion of Sample 06, Ti/Co is less than or equal to 0.01 in each measurement point; in contrast, in the crack portion of Sample 07, the amount of titanium is increased in many points, and there are measurement points where Ti/Co is greater than or equal to 0.05. Furthermore, Ti/Co on the surfaces of the particles of Sample 07 is between 0.10 and 0.18.

[0512]

Next, FIGS. 42A and 42B show results of EDX point analysis in an atomic ratio of magnesium to cobalt. The detection points are the same as those in FIGS. 41A and 41B.

[0513]

As shown in FIGS. 42A and 42B, in Sample 06, Mg/Co is less than or equal to 0.03 both

on the surfaces of the particles and in the crack portion; in contrast, in Sample 07, there are many points where the amount of magnesium is increased both on the surfaces of the particles and in the crack portion. Furthermore, Mg/Co on the surfaces of the particles is between 0.15 and 0.50, and that in the crack portion is between 0 and 0.22.

[0514]

Next, CR2032 coin-type secondary batteries were fabricated using the positive electrode active material of Sample 07 after heating. A positive electrode formed by applying slurry in which a positive electrode active material (LCO) of Sample 02, AB, and polyvinylidene fluoride (PVDF) were mixed at a weight ratio of 95:3:2 to a positive electrode current collector was used. As the positive electrode current collector, 20-μm-thick aluminum foil was used. The amount of positive electrode active material layer containing the positive electrode active material, AB, and PVDF was 7.6 mg/cm^2.

[0515]

A lithium metal was used for the counter electrode.

[0516]

An electrolyte solution formed in such a manner that 1 mol/L $LiPF_6$ was dissolved in a solution in which ethylene carbonate (EC) and diethyl carbonate (DEC) were mixed at a volume ratio of 3:7, and vinylene carbonate (VC) was added to the solution at a 2 wt% was used.

[0517]

[Initial characteristics, rate characteristics]

Initial characteristics and rate characteristics of the secondary battery using the positive electrode active material of Sample 07 formed in the above manner were measured.

[0518]

In the measurement of the initial characteristics, charging was performed at CCCV, 0.2 C, 4.6 V, and a cutoff current of 0.05 C, and discharging was performed at CC, 0.2 C, and a cutoff voltage of 3.0 V. Here, 1C was set to 160 mA/g, which was a current value per weight of the positive electrode active material. The measurement temperature was 25 °C. Table 2 shows measurement results of initial characteristics.

[0519]

[Table 2]

Initial charge capacity [mAh/g]	Initial discharge capacity [mAh/g]	Initial charge and discharge efficiency [%]
221.2	217.5	98.3

[0520]

The rate characteristics were measured after the initial characteristics were measured. The measurement was performed by changing a discharge rate in the following order: 0.2 C charge/0.2 C discharge; 0.2 C charge/0.5 C discharge; 0.2 C charge/1.0 C discharge; 0.2 C charge/2.0 C discharge; 0.2 C charge/3.0 C discharge; 0.2 C charge/4.0 C discharge; and 0.2 C charge/5.0 C discharge. Note that the conditions other than the discharge rate are the same as those of the measurement of the initial characteristics. The measurement temperature was 25 °C.

[0521]

Table 3 shows the measurement results of the initial characteristics and the rate characteristics. In addition, FIG. 43 shows discharge curves of the rates.

[0522]

[Table 3]

Rate [C]	Discharge capacity [mAh/g]	Discharge capacity retention rate [%]
0.2	218.2	100.0
0.5	215.8	98.9
1.0	213.1	97.7
2.0	207.9	95.3
3.0	204.0	93.5
4.0	198.8	91.1
5.0	184.3	84.5

[0523]

[Temperature characteristics]

Next, a cell was fabricated under conditions similar to those of a cell for evaluating the rate characteristics except that the amount of positive electrode active material layer was 8.2 mg/cm^2, and the temperature characteristics were measured. Charging was performed at 25 °C, CCCV, 0.2 C, 4.6 V, and a cutoff current of 0.05 C. Discharging was performed at 25 °C, 0 °C, −10 °C, −20 °C, and 45 °C in this order, CC, 0.2 C, and a cutoff voltage of 3.0 V. FIG. 44 shows measurement results of temperature characteristics.

[0524]

[Cycle characteristics]

Next, a cell was fabricated under conditions similar to those of the cell for measuring the temperature characteristics, and the cycle characteristics were measured. In the measurement of the cycle characteristics, charging was performed at CCCV, 1.0 C, 4.55 V, and a cutoff current of

0.05 C, and discharging was performed at CC, 1.0 C, and a cutoff voltage of 3.0 V. The measurement temperature of the cycle characteristics was 45 °C and 100 cycles were measured. The discharge capacity retention rate after 100 cycles was 86 %. FIG. 45 is a graph showing the discharge capacity retention rate of the measured cycle characteristics.

[0525]

From the measurement results, the specific surface area of the positive electrode active material of Sample 07 was 0.13 m^2/g.

[0526]

Furthermore, from the measurement results of particle size distribution of the positive electrode active material of Sample 07, the average particle diameter was 21.5 μm, 10 %D was 13.1 μm, 50 %D was 22.0 μm, and 90 %D was 34.4 μm.

[0527]

The tap density of the positive electrode active material of Sample 07 is 2.21 g/cm^3. The tap density was measured with MULTI TESTER MT-1000 (manufactured by SEISHIN ENTERPRISE Co., Ltd.).

[0528]

As described above, it is found that the positive electrode active material of Sample 07 which is one embodiment of the present invention shows excellent initial charge and discharge capacity, rate characteristics, and cycle characteristics. In particular, the initial charge and discharge capacity is high, which is 98 % or higher; thus, it is probable that a side reaction is inhibited. In addition, even at a high discharge rate of 2 C, an excellent capacity of 96.1 % is shown using 0.2 C as a reference.

[Example 3]

[0529]

In this example, a positive electrode active material including a region containing titanium and magnesium in a superficial portion was formed by changing the ratio of Li to the first transition metal of starting materials, and evaluation results of characteristics are shown.

[0530]

[Formation of positive electrode active material]

In this example, positive electrode active materials of Samples 11 to 17, Samples 21 to 28, and Samples 31 to 40 in which cobalt was used as the first transition metal were prepared. Formation methods and conditions of these samples are as follows.

[0531]

<<Samples 11 to 17>>

First, a source of lithium, a source of cobalt, a source of magnesium, and a source of

fluorine, which are to be starting materials, were individually weighed. In this example, lithium carbonate, cobalt oxide, magnesium oxide, and lithium fluoride were used as the source of lithium, the source of cobalt, the source of magnesium, and the source of fluorine, respectively.

[0532]

At that time, the starting materials of Sample 11 were weighed so that the ratio of Li to Co was 1.00. The starting materials of Sample 12 were weighed so that the ratio of Li to Co was 1.03. The starting materials of Sample 13 were weighed so that the ratio of Li to Co was 1.05. The starting materials of Sample 14 were weighed so that the ratio of Li to Co was 1.06. The starting materials of Sample 15 were weighed so that the ratio of Li to Co was 1.07. The starting materials of Sample 16 were weighed so that the ratio of Li to Co was 1.08. The starting materials of Sample 17 were weighed so that the ratio of Li to Co was 1.13.

[0533]

In addition, the starting materials of each of Samples 11 to 17 were weighed so that, when the number of atoms of cobalt contained in the starting materials was set to 1, the number of atoms of magnesium was 0.01 and the number of atoms of fluorine was 0.02.

[0534]

Next, the weighed starting materials were separately mixed with a ball mill for each sample.

[0535]

Then, the mixed starting materials were baked. The baking was performed at 1000 °C for 10 hours under the following conditions: the temperature rising rate was 200 °C/h; and the flow rate of dry air was 10 L/min.

[0536]

Through the above process, particles of a composite oxide containing lithium, cobalt, fluorine, and magnesium were synthesized.

[0537]

Next, TTIP was added to 2-propanol so that the amount of TTIP per weight of the positive electrode active material was 0.01 ml/g and then mixing was performed, so that a 2-propanol solution of tetra-i-propoxy titanium was formed.

[0538]

To the 2-propanol solution of TTIP, the particles of a composite oxide containing lithium, cobalt, fluorine, and magnesium were added and then mixing was performed.

[0539]

The above-described mixed solution was stirred with a magnetic stirrer for four hours, at 25 °C, and at a humidity of 90 %RH. Through the process, water in an atmosphere and TTIP

caused hydrolysis and polycondensation reaction, and a layer containing titanium was formed on the surface of the lithium cobalt oxide particle containing magnesium and fluorine.

[0540]

The mixed solution which had been subjected to the above process was filtered to collect the residue. As a filter for filtration, Kiriyama filter paper (No. 4) was used.

[0541]

The collected residue was dried in a vacuum at 70 °C for one hour.

[0542]

The dried particles were heated. The heating was performed in an oxygen atmosphere under the following conditions: the temperature was 800 °C (the temperature rising rate was 200 °C/h); and the retention time was two hours.

[0543]

The heated particles were cooled and subjected to crushing treatment. In the crushing treatment, the particles were made to pass through a sieve. The sieve has an aperture width of 53 μm.

[0544]

The particles which were subjected to crushing treatment were used as the positive electrode active materials of Samples 11 to 17.

[0545]

<<Samples 21 to 27>>

The starting materials of Samples 21 to 27 were the same as those of Samples 11 to 16. At that time, the starting materials of Sample 21 were weighed so that the ratio of Li to Co was 1.00. The starting materials of Sample 22 were weighed so that the ratio of Li to Co was 1.03. The starting materials of Sample 23 were weighed so that the ratio of Li to Co was 1.05. The starting materials of Sample 24 were weighed so that the ratio of Li to Co was 1.06. The starting materials of Sample 25 were weighed so that the ratio of Li to Co was 1.07. The starting materials of Sample 26 were weighed so that the ratio of Li to Co was 1.08. The starting materials of Sample 27 were weighed so that the ratio of Li to Co was 1.13.

[0546]

Samples 21 to 27 were formed in manners similar to those of Samples 11 to 17 except that the concentration of TTIP in the 2-propanol solution was adjusted so that the amount of TTIP per weight of the positive electrode active material was 0.02 ml/g.

[0547]

<<Sample 28>>

The ratio of Li to Co of the starting materials and the amount of TTIP of Sample 28 were

the same as the ratio of Li to Co of the starting materials and the amount of TTIP of Sample 23. That is, in Sample 28, the starting materials were weighed so that the ratio of Li to Co was 1.05, and the amount of TTIP per weight of the positive electrode active material was 0.02 ml/g.

[0548]

Note that in Sample 28, after the starting materials were mixed, baking was performed at 950 °C.

[0549]

Sample 28 was formed in a manner similar to that of Sample 23 except for the baking temperature.

[0550]

It is probable that Samples 11 to 17 and Samples 21 to 28 are each a positive electrode active material which contains lithium cobaltate inside and includes a region containing titanium and magnesium in a superficial portion.

[0551]

<<Samples 31 to 40>>

Samples 31 to 40 were formed as comparative examples, each of which did not include a region containing titanium.

[0552]

The starting materials of Sample 31 were weighed so that the ratio of Li to Co was 1.00. The starting materials of Sample 32 were weighed so that the ratio of Li to Co was 1.01. The starting materials of Sample 33 were weighed so that the ratio of Li to Co was 1.02. The starting materials of Sample 34 were weighed so that the ratio of Li to Co was 1.03. The starting materials of Sample 35 were weighed so that the ratio of Li to Co was 1.035. The starting materials of Sample 36 were weighed so that the ratio of Li to Co was 1.04. The starting materials of Sample 37 were weighed so that the ratio of Li to Co was 1.051. The starting materials of Sample 38 were weighed so that the ratio of Li to Co was 1.061. The starting materials of Sample 39 were weighed so that the ratio of Li to Co was 1.081. The starting materials of Sample 40 were weighed so that the ratio of Li to Co was 1.130.

[0553]

In addition, the starting materials of each of Samples 31 to 40 were weighed so that, when the number of atoms of cobalt contained in the starting materials was set to 1, the number of atoms of magnesium was 0.01 and the number of atoms of fluorine was 0.02.

[0554]

Next, the weighed starting materials were separately mixed with a ball mill for each sample.

[0555]

Then, the mixed starting materials were baked. The baking was performed at 1000 °C for 10 hours under the following conditions: the temperature rising rate was 200 °C/h; and the flow rate of dry air was 10 L/min.

[0556]

Through the above process, particles of a composite oxide containing lithium, cobalt, fluorine, and magnesium were synthesized.

[0557]

The synthesized particles were cooled and then heated. The heating was performed in an oxygen atmosphere under the following conditions: the temperature was 800 °C (the temperature rising rate was 200 °C/h); and the retention time was two hours.

[0558]

The heated particles were cooled and subjected to crushing treatment. In the crushing treatment, the particles were made to pass through a sieve. The sieve has an aperture width of 53 μm.

[0559]

The particles which were subjected to crushing treatment were used as the positive electrode active materials of Samples 31 to 40.

[0560]

Table 4 shows the formation conditions of Samples 11 to 17, Samples 21 to 28, and Samples 31 to 40.

[0561]

[Table 4]

	Li/Co	TTIP	Baking temperature
Sample 11	1.00	0.01 ml/g	1000 °C
Sample 12	1.03		
Sample 13	1.05		
Sample 14	1.06		
Sample 15	1.07		
Sample 16	1.08		
Sample 17	1.13		
Sample 21	1.00	0.02 ml/g	1000 °C
Sample 22	1.03		
Sample 23	1.05		
Sample 24	1.06		
Sample 25	1.07		
Sample 26	1.08		
Sample 27	1.13		
Sample 28	1.05	0.02 ml/g	950 °C
Sample 31	1.00	-	1000 °C
Sample 32	1.01		
Sample 33	1.02		
Sample 34	1.03		
Sample 35	1.035		
Sample 36	1.04		
Sample 37	1.051		
Sample 38	1.061		
Sample 39	1.081		
Sample 40	1.130		

[0562]

[XPS]

The positive electrode active materials of Samples 11 to 17, Samples 21 to 28, and Samples 31 to 40 were subjected to an XPS analysis. Table 5 shows results of the XPS analysis of Samples 11 to 17, Table 6 shows results of the XPS analysis of Samples 21 to 28, and Table 7 shows results of the XPS analysis of Samples 31 to 40. Tables 5 to 7 show a relative value of the concentration of each element under the condition where the concentration of cobalt is 1.

[0563]

[Table 5]

		Relative value under condition where concentration of Co is 1										
	Li/Co	Li	Co	O	C	F	Mg	Ti	Si	Ca	Na	S
Sample 11	1.00	0.82	1.00	2.87	0.25	0.37	0.57	0.15	0.00	0.04	0.27	0.01
Sample 12	1.03	0.69	1.00	2.84	0.93	0.63	0.63	0.14	0.00	0.05	0.04	0.00
Sample 13	1.05	0.96	1.00	3.22	0.66	0.47	0.61	0.14	0.00	0.03	0.09	0.03
Sample 14	1.06	1.04	1.00	3.13	0.34	0.77	0.77	0.15	0.00	0.05	0.34	0.04
Sample 15	1.07	0.84	1.00	3.42	0.58	0.60	0.23	0.18	0.00	0.08	0.27	0.00
Sample 16	1.08	1.08	1.00	3.10	0.66	0.59	0.04	0.16	0.00	0.03	0.16	0.02
Sample 17	1.13	1.29	1.00	3.70	0.83	0.87	0.00	0.19	0.00	0.04	0.20	0.02

[0564]

[Table 6]

		Relative value under condition where concentration of Co is 1										
	Li/Co	Li	Co	O	C	F	Mg	Ti	Si	Ca	Na	S
Sample 21	1.00	1.08	1.00	3.23	0.47	0.45	0.49	0.20	0.00	0.06	0.35	0.01
Sample 22	1.03	0.73	1.00	3.05	1.06	0.70	0.59	0.17	0.00	0.03	0.09	0.00
Sample 23	1.05	1.19	1.00	3.40	0.73	0.48	0.58	0.23	0.00	0.05	0.10	0.04
Sample 24	1.06	1.10	1.00	3.44	0.33	0.92	0.91	0.25	0.00	0.05	0.30	0.04
Sample 25	1.07	1.05	1.00	4.26	0.74	0.20	0.25	0.29	0.00	0.07	0.30	0.02
Sample 26	1.08	1.12	1.00	3.32	0.74	0.52	0.14	0.21	0.00	0.05	0.15	0.00
Sample 27	1.13	1.49	1.00	3.80	0.60	0.41	0.00	0.12	0.00	0.00	0.16	0.02
Sample 28	1.05	0.99	1.00	3.11	0.72	0.86	0.56	0.18	0.00	0.05	0.12	0.02

[0565]

[Table 7]

		Relative value under condition where concentration of Co is 1							
	Li/Co	Li	Co	O	C	F	Mg	Ca	Na
Sample 31	1.00	0.51	1.00	2.45	0.69	0.08	0.27	0.03	0.08
Sample 32	1.01	0.67	1.00	2.65	0.77	0.08	0.28	0.03	0.08
Sample 33	1.02	0.53	1.00	2.51	0.66	0.09	0.27	0.02	0.06
Sample 34	1.03	0.79	1.00	2.93	0.92	0.09	0.35	0.04	0.14
Sample 35	1.04	0.65	1.00	2.33	0.48	0.11	0.32	0.03	0.11
Sample 36	1.04	0.69	1.00	2.73	0.56	0.11	0.38	0.05	0.16
Sample 37	1.05	0.67	1.00	3.04	0.64	0.09	0.35	0.04	0.21
Sample 38	1.06	0.83	1.00	2.65	1.03	0.29	0.10	0.05	0.11
Sample 39	1.08	0.80	1.00	2.79	1.04	0.26	0.03	0.08	0.10
Sample 40	1.13	0.77	1.00	2.72	0.22	0.99	0.00	0.01	0.26

[0566]

FIGS. 46A and 46B are graphs in which the relative valueof magnesium and the relative valueof titanium are extracted from the analysis results of Tables 5 to 7. FIG. 46A is a graph showing the ratio of Li to Co and the relative valueof magnesium. FIG. 46B is a graph showing

the ratio of Li to Co and the relative valueof titanium.

[0567]

From the analysis results of Samples 31 to 40 in FIG. 46A, it is found that, in the case where a coating layer containing titanium is not included, the concentration of magnesium is high in samples where the ratio of Li to Co is greater than or equal to 1.00 and less than or equal to 1.05. This is probably because magnesium contained in the starting materials is segregated in a range where the element concentration can be detected by XPS by heating. In contrast, in samples where the ratio of Li to Co is greater than or equal to 1.06, the concentration of magnesium is low; thus, it is probable that the segregation of magnesium does not easily occur when the amount of lithium is too large.

[0568]

From the analysis results of Samples 11 to 16 and Samples 21 to 26 in FIG. 46A, it is found that the concentration of magnesium in a range where the element concentration can be detected by XPS is higher in the case where a region containing titanium is included in a superficial portion than in the case where the region containing titanium is not included.

[0569]

Moreover, in the case where the ratio of Li to Co is 1.06, in samples where the region containing titanium is not included, the concentration of magnesium in a range where the element concentration can be detected by XPS is low; in contrast, in samples where the region containing titanium is included, the concentration of magnesium in a range where the element concentration can be detected by XPS is high. That is, when the region containing titanium is formed in the superficial portion, magnesium is sufficiently segregated even in the case where the ratio of Li to Co is high.

[0570]

Note that even if the region containing titanium is included, the concentration of magnesium is lower in the case where the ratio of Li to Co is 1.07 than in the case where the ratio of Li to Co is 1.06. Furthermore, it is probable that, in the case where the ratio of Li to Co is greater than or equal to 1.08, the segregation of magnesium does not easily occur even if the region containing titanium is included.

[0571]

[Evaluation of cycle characteristics]

<<Energy density retention rate>>

Next, cycle characteristics were evaluated in a manner similar to that of Example 1 using positive electrode active materials of Samples 11 to 14, Sample 16, Samples 21 to 24, and Sample 26.

[0572]

The shape of the secondary battery, the materials and the mixture ratios of the positive electrode active material, the conductive additive, and the binder in the positive electrode, the counter electrode, the electrolyte solution, the exterior body, the conditions of the cycle characteristics test, and the like are the same as those in Example 1.

[0573]

FIG. 47A is a graph showing the energy density retention rates and the number of charge and discharge cycles at the time of charging at 4.6 V of secondary batteries using the positive electrode active materials of Samples 11 to 14 and Sample 16 which were formed so that the amount of TTIP per weight of the positive electrode active material was 0.01 ml/g. FIG. 47B is a graph showing the energy density retention rates and the number of charge and discharge cycles at the time of charging at 4.6 V of secondary batteries using the positive electrode active materials of Samples 21 to 24 and Sample 26 which were formed so that the amount of TTIP per weight of the positive electrode active material was 0.02 ml/g.

[0574]

As shown in FIG. 47A, in the case where TTIP is 0.01 ml/g, Samples 11 to 14, that is, the positive electrode active materials in which the ratio of Li to Co is greater than or equal to 1.00 and less than or equal to 1.06 have excellent cycle characteristics. In particular, Samples 11 and 12, that is, the positive electrode active materials in which the ratio of Li to Co is greater than or equal to 1.00 and less than or equal to 1.03 have extremely excellent cycle characteristics. In contrast, in Sample 16 in which the ratio of Li to Co is 1.08, the energy density retention rate decreases at a relatively early stage.

[0575]

As shown in FIG. 47B, in the case where TTIP is 0.02 ml/g, Samples 21 to 24, that is, the positive electrode active materials in which the ratio of Li to Co is greater than or equal to 1.00 and less than or equal to 1.06 have excellent cycle characteristics. In particular, Samples 23 and 24, that is, the positive electrode active materials in which the ratio of Li to Co is greater than or equal to 1.05 and less than or equal to 1.06 have extremely excellent cycle characteristics.

[0576]

FIG. 48 is a graph showing comparison between Sample 11 having the most excellent cycle characteristics in Samples 11 to 15 and Sample 23 having the most excellent cycle characteristics in Samples 21 to 25.

[0577]

As shown in FIG. 48, both Sample 11 and Sample 23 have excellent cycle characteristics; however, Sample 23 in which TTIP is 0.02 ml/g has more excellent cycle characteristics.

[0578]

<<Discharge capacity retention rate>>

Next, FIG. 49 shows evaluation results of a discharge capacity retention rate, which is one of the cycle characteristics of each of Samples 21 to 26 and Sample 28.

[0579]

The shape of the secondary battery, the material and the mixture ratio of the positive electrode active material, the conductive additive, and the binder in the positive electrode, the counter electrode, the electrolyte solution, the exterior body, the conditions of the cycle characteristics test, and the like of Samples 21 to 26 are the same as those in Example 1.

[0580]

The secondary battery using Sample 28 was formed in a manner similar to those of the secondary batteries using Samples 21 to 26 except that PVDF was used as a binder and the positive electrode active material (LCO), AB, and PVDF were mixed such that the weight ratio of LCO to AB and PVDF was 95:3:2, and evaluated.

[0581]

As shown in FIG. 49, Samples 21 to 24 and Sample 28 have excellent cycle characteristics. In particular, Sample 28 has extremely excellent cycle characteristics. In Sample 28, the discharge capacity retention rate after 50 cycles was higher than or equal to 85 %.

[0582]

In contrast, in Sample 25 and Sample 26 in which the ratios of Li to Co are 1.07 and 1.08, respectively, the discharge capacity retention rates decrease at a relatively early stage.

[0583]

From the above results, it is found that in the case where TTIP per weight of the positive electrode active material is 0.02 ml/g, the ratio of Li to Co preferably has a range of greater than or equal to 1.00 and less than 1.07. Moreover, it is found that a sample in which the ratio of Li to Co has a range of greater than or equal to 1.05 and less than or equal to 1.06 has extremely excellent cycle characteristics.

[0584]

FIGS. 50A to 50C show charge and discharge curves of the secondary batteries using Sample 28, which has extremely excellent cycle characteristics in FIG. 49, Sample 24, and Sample 25, in which the degradation occurs at a relatively early stage.

[0585]

FIG. 50A, FIG. 50B, and FIG. 50C show charge and discharge curves of the secondary batteries using Sample 28, Sample 24, and Sample 25, respectively. Each of the figures shows overlap of results of 50 cycles of charge and discharge. As indicated by an arrow in each figure,

the charge and discharge capacity decreases from the first cycle to the fiftieth cycle.

[0586]

As shown in FIGS. 50A and 50B, Sample 28 and Sample 24, which are positive electrode active materials of one embodiment of the present invention, have high charge and discharge capacity and excellent charge and discharge characteristics. In addition, it is found that a decrease in charge and discharge capacity of each of Sample 28 and Sample 24 in FIGS. 50A and 50B is significantly suppressed as compared with Sample 25 in FIG. 50C.

[Example 4]

[0587]

In this example, SEM observation results and SEM-EDX analysis results of the positive electrode active material of Sample 24 formed in Example 2 are described.

[0588]

Sample 24 was formed so that Li/Co was 1.06 and TTIP per weight of the positive electrode active material was 0.02 ml/g. FIG. 51A shows a SEM image of Sample 24. FIGS. 51B and 51C each show an enlarged image of a part in FIG. 51A.

[0589]

As shown in FIGS. 51A to 51C, there are a large number of projected regions in a superficial portion of the positive electrode active material.

[0590]

Next, FIGS. 52A-1, 52A-2, 52B-1, 52B-2, 52C-1, and 52C-2 show analysis results of the positive electrode active material of Sample 24 using SEM-EDX. FIG. 52A-1 shows a SEM image of a superficial portion of the positive electrode active material, FIG. 52A-2 shows a mapping image of titanium, FIG. 52B-1 shows a mapping image of magnesium, FIG. 52B-2 shows a mapping image of oxygen, FIG. 52C-1 shows a mapping image of aluminum, and FIG. 52C-2 shows a mapping image of cobalt. Note that in EDX element mapping images in FIGS. 52A-2, 52B-1, 52B-2, 52C-1, and 52C-2, a region where the number of elements is less than or equal to a lower limit of the detection is indicated in black, and as the number of elements is increased, the black region becomes white.

[0591]

The same regions in FIGS. 52A-1, 52A-2, and 52B-1 are surrounded by dotted lines. When the regions surrounded by dotted lines are compared with each other, it is found that in the projected regions in the superficial portion of the positive electrode active material, titanium and magnesium are distributed.

[0592]

Thus, it is found that Sample 24 is a positive electrode active material including the

projected fourth region 104 containing titanium and magnesium over the third region 103.

[0593]

As shown in Example 2, Sample 24 is one of the samples which show extremely excellent cycle characteristics. Thus, it is found that, even if the fourth region is provided in the superficial portion or when the fourth region is provided, the positive electrode active material with excellent cycle characteristics can be obtained.

[0594]

From the above results of Examples 1 to 3, it is found that when the region containing titanium is formed in the superficial portion, the positive electrode active material with excellent cycle characteristics can be obtained. In addition, it is found that, when the ratio of Li to Co is increased to increase the particle diameter of the positive electrode active material, the cycle characteristics might be degraded; however, the region containing titanium is formed in the superficial portion, whereby the range of the ratio of Li to Co in which excellent cycle characteristics is obtained can be widened. Furthermore, it is found that even when the fourth region containing titanium and magnesium is provided in the superficial portion of the positive electrode active material, excellent cycle characteristics is obtained.

[Example 5]

[0595]

In this example, an example of a method for producing a positive electrode active material coated with graphene oxide is shown, and observation results of the positive electrode active material produced by the method with an electron microscope are described.

[0596]

As shown in a process flow chart in FIG. 53, a process for forming a coating film on a positive electrode active material includes the steps of: weighing graphene oxide (S11); mixing and stirring the graphene oxide and pure water (S12); controlling pH (S13); adding an active material (S14); completing a suspension (S15); spraying the suspension using a spray dry apparatus (S16); and collecting particles in a container (S17).

[0597]

Note that in (S12), pure water is used as a dispersion medium; however, a dispersion medium is not particularly limited and ethanol or the like may be used. In addition, in (S14), the active material is a positive electrode active material.

[0598]

FIG. 54 is a schematic view of a spray dry apparatus 280. The spray dry apparatus 280 includes a chamber 281 and a nozzle 282. A suspension 284 is supplied to the nozzle 282 through a tube 283. The suspension 284 is supplied from the nozzle 282 to the chamber 281 in the form

of mist and dried in the chamber 281. The nozzle 282 may be heated with a heater 285. Here, a region of the chamber 281 which is close to the nozzle 282, for example, a region surrounded by dashed-two dotted line in FIG. 54, is also heated with the heater 285.

[0599]

In the case of using a suspension containing a positive electrode active material and graphene oxide as the suspension 284, powder of the positive electrode active material coated with the graphene oxide is collected in a collection container 286 through the chamber 281.

[0600]

The air in the chamber 281 may be suctioned by an aspirator or the like through a path indicated by an arrow 288.

[0601]

An example of conditions for forming the coating film is shown below.

[0602]

First, a suspension was formed by dispersing graphene oxide into a solvent.

[0603]

Although in pure water, graphene oxide is highly dispersible, pure water might react with an active material added later, so that Li might be dissolved or an active material might be damaged to change the surface structure. Thus, the graphene oxide was dispersed into a solution such that the ratio between ethanol and pure water was 4:6.

[0604]

Stirring to disperse the graphene oxide into a solution was performed under the following conditions: a stirrer and an ultrasonic wave generator were used; a rotation rate was 750 rpm; and irradiation time with ultrasonic waves was 2 minutes.

[0605]

Then, a LiOH aqueous solution was dropped to adjust pH to be pH7 (25 °C).

[0606]

The positive electrode active materials (in this example, lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-20F)) were added, and stirring was performed using a stirrer and an ultrasonic wave generator under the following conditions: a rotation rate was 750 rpm; and irradiation time with ultrasonic waves was 1 minute. Through the above process, the suspension was prepared. The above lithium cobalt oxide particles produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD. (product name: C-20F) contain at least fluorine, magnesium, calcium, sodium, silicon, sulfur, and phosphorus, and each have a diameter of approximately 20 μm.

[0607]

Next, the suspension was sprayed uniformly with a spray nozzle (having a nozzle diameter of 20 μm) of the spray dry apparatus to obtain powder. The inlet temperature was 160 °C and the outlet temperature was 40 °C as the hot-air temperature of the spray dry apparatus, and the N_2 gas flow rate was 10 L/min.

[0608]

FIG. 55 shows a cross-sectional TEM image of the obtained powder. In addition, FIG. 56 shows a SEM image of the obtained powder. When a positive electrode active material which was the same as the sprayed positive electrode active material (C-20F, produced by NIPPON CHEMICAL INDUSTRIAL CO., LTD.) was mixed with the graphene oxide with a planetary centrifugal mixer as a comparative example, coating was insufficient. FIG. 57 shows a SEM image of the comparative example.

[0609]

It is found that, as compared with FIG. 57, the coating film is uniformly formed on the surface of the powder in FIG. 56.

[0610]

FIGS. 58A and 58B illustrate a cross-sectional structure example of an active material layer 200 which is coated with graphene oxide with a spray dry apparatus and includes a graphene compound as a conductive additive.

[0611]

FIG. 58A is a longitudinal sectional view of the active material layer 200. The active material layer 200 includes positive electrode active material particles 100 coated with graphene oxide, a graphene compound 201 as a conductive additive, and a binder (not illustrated). Here, graphene or multilayer graphene may be used as the graphene compound 201, for example. The graphene compound 201 preferably has a sheet-like shape. The graphene compound 201 may have a sheet-like shape formed of a plurality of sheets of multilayer graphene and/or a plurality of sheets of graphene that partly overlap with each other.

[0612]

In the longitudinal section of the active material layer 200, as illustrated in FIG. 58B, the positive electrode active material 100 coated with a coating film 105 formed of graphene oxide is in contact with the graphene compound 201. A plurality of graphene compounds 201 are formed in such a way as to be partly in contact with the positive electrode active material 100 coated with the coating film 105 and adhere to the coating film 105 of the adjacent positive electrode active material 100, so that the graphene compounds 201 are in contact with each of the positive electrode active material 100.

[0613]

The graphene compound 201 and the coating film 105 are formed using carbon-based materials; thus, an excellent conductive path can be formed.

[0614]

The coating film 105 is effective in protecting the crystal structure of the positive electrode active material 100 so as not to be in contact with the electrolyte solution and in forming the excellent conductive path.

REFERENCE NUMERALS

[0615]

11a: positive electrode, 11b: negative electrode, 12a: lead, 12b: lead, 14: separator, 15a: bonding portion, 15b: bonding portion, 17: fixing member, 50: secondary battery, 51: exterior body, 61: folded portion, 62: seal portion, 63: seal portion, 71: crest line, 72: trough line, 73: space, 100: positive electrode active material, 101: first region, 101p: crystal plane, 102: second region, 102p: crystal plane, 103: third region, 103p: crystal plane, 104: fourth region, 105: coating film, 106: crack portion, 110: particle, 111: region, 112: layer containing titanium, 114: cobalt oxide layer, 120: particle, 121: region, 122: layer containing titanium, 124: cobalt oxide layer, 125: layer containing lithium titanate, 200: active material layer, 201: graphene compound, 214: separator, 280: spray dry apparatus, 281: chamber, 282: nozzle, 283: tube, 284: suspension, 285: heater, 286: collection container, 288: arrow, 300: coin-type secondary battery, 301: positive electrode can, 302: negative electrode can, 303: gasket, 304: positive electrode, 305: positive electrode current collector, 306: positive electrode active material layer, 307: negative electrode, 308: negative electrode current collector, 309: negative electrode active material layer, 310: separator, 500: laminated secondary battery, 501: positive electrode current collector, 502: positive electrode active material layer, 503: positive electrode, 504: negative electrode current collector, 505: negative electrode active material layer, 506: negative electrode, 507: separator, 508: electrolyte solution, 509: exterior body, 510: positive electrode lead electrode, 511: negative electrode lead electrode, 600: cylindrical secondary battery, 601: positive electrode cap, 602: battery can, 603: positive electrode terminal, 604: positive electrode, 605: separator, 606: negative electrode, 607: negative electrode terminal, 608: insulating plate, 609: insulating plate, 611: PTC element, 612: safety valve mechanism, 613: conductive plate, 614: conductive plate, 615: module, 616: wiring, 617: temperature control device, 900: circuit board, 910: label, 911: terminal, 912: circuit, 913: secondary battery, 914: antenna, 915: antenna, 916: layer, 917: layer, 918: antenna, 920: display device, 921: sensor, 922: terminal, 930: housing, 930a: housing, 930b: housing, 931: negative electrode, 932: positive electrode, 933: separator, 950: wound body, 951: terminal, 952: terminal, 980: laminated secondary battery, 981: film, 982: film, 993: wound body, 994: negative electrode,

995: positive electrode, 996: separator, 997: lead electrode, 998: lead electrode, 7100: portable display device, 7101: housing, 7102: display portion, 7103: operation button, 7104: secondary battery, 7200: portable information terminal, 7201: housing, 7202: display portion, 7203: band, 7204: buckle, 7205: operation button, 7206: input output terminal, 7207: icon, 7300: display device, 7304: display portion, 7400: mobile phone, 7401: housing, 7402: display portion, 7403: operation button, 7404: external connection port, 7405: speaker, 7406: microphone, 7407: secondary battery, 7408: lead electrode, 7409: current collector, 7500: vaporizer, 7501: atomizer, 7502: cartridge, 7504: secondary battery, 8000: display device, 8001: housing, 8002: display portion, 8003: speaker portion, 8004: secondary battery, 8021: ground-based charging apparatus, 8022: cable, 8024: secondary battery, 8100: lighting device, 8101: housing, 8102: light source, 8103: secondary battery, 8104: ceiling, 8105: wall, 8106: floor, 8107: window, 8200: indoor unit, 8201: housing, 8202: air outlet, 8203: secondary battery, 8204: outdoor unit, 8300: electric refrigerator-freezer, 8301: housing, 8302: refrigerator door, 8303: freezer door, 8304: secondary battery, 8400: automobile, 8401: headlight, 8406: electric motor, 8500: automobile, 8600: motor scooter, 8601: side mirror, 8602: secondary battery, 8603: indicator, 8604: storage unit under seat, 9600: tablet terminal, 9625: power saving mode changing switch, 9626: display mode changing switch, 9627: power switch, 9628: operation switch, 9629: fastener, 9630: housing, 9630a: housing, 9630b: housing, 9631: display portion, 9631a: display portion, 9631b: display portion, 9633: solar cell, 9634: charge and discharge control circuit, 9635: power storage unit, 9636: DCDC converter, 9637: converter, 9640: movable portion.

This application is based on Japanese Patent Application Serial No. 2016-133997 filed with Japan Patent Office on July 6, 2016, Japanese Patent Application Serial No. 2016-133143 filed with Japan Patent Office on July 5, 2016, Japanese Patent Application Serial No. 2017-002831 filed with Japan Patent Office on January 11, 2017, Japanese Patent Application Serial No. 2017-030693 filed with Japan Patent Office on February 22, 2017, Japanese Patent Application Serial No. 2017-084321 filed with Japan Patent Office on April 21, 2017,and Japanese Patent Application Serial No. 2017-119272 filed with Japan Patent Office on June 19, 2017 the entire contents of which are hereby incorporated by reference.

ABSTRACT

A positive electrode active material which can improve cycle characteristics of a secondary battery is provided. Two kinds of regions are provided in a superficial portion of a positive electrode active material such as lithium cobaltate which has a layered rock-salt crystal structure. The inner region is a non-stoichiometric compound containing a transition metal such as titanium, and the outer region is a compound of representative elements such as magnesium oxide. The two kinds of regions each have a rock-salt crystal structure. The inner layered rock-salt crystal structure and the two kinds of regions in the superficial portion are topotaxy; thus, a change of the crystal structure of the positive electrode active material generated by charging and discharging can be effectively suppressed. In addition, since the outer coating layer in contact with an electrolyte solution is the compound of representative elements which is chemically stable, the secondary battery having excellent cycle characteristics can be obtained.

FIG. 12A

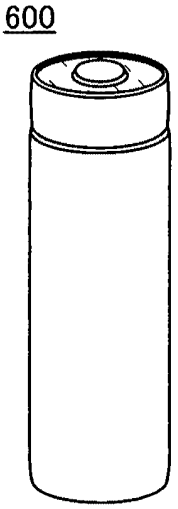


FIG. 12B

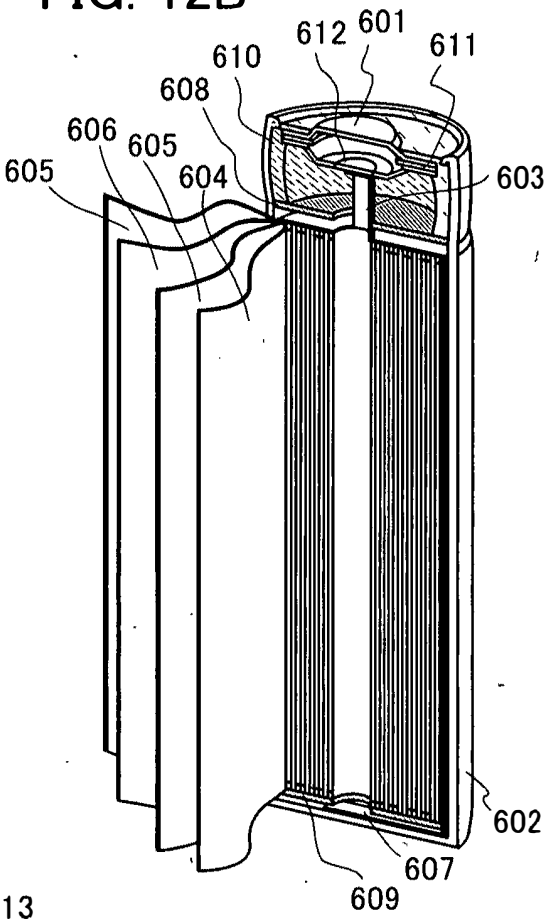


FIG. 12C

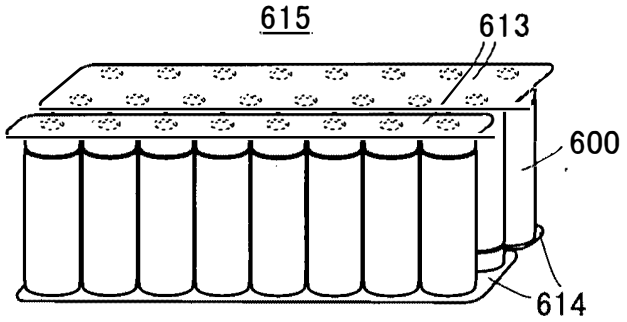


FIG. 12D

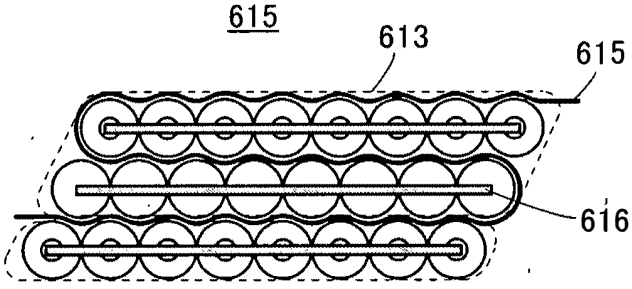


FIG. 22A

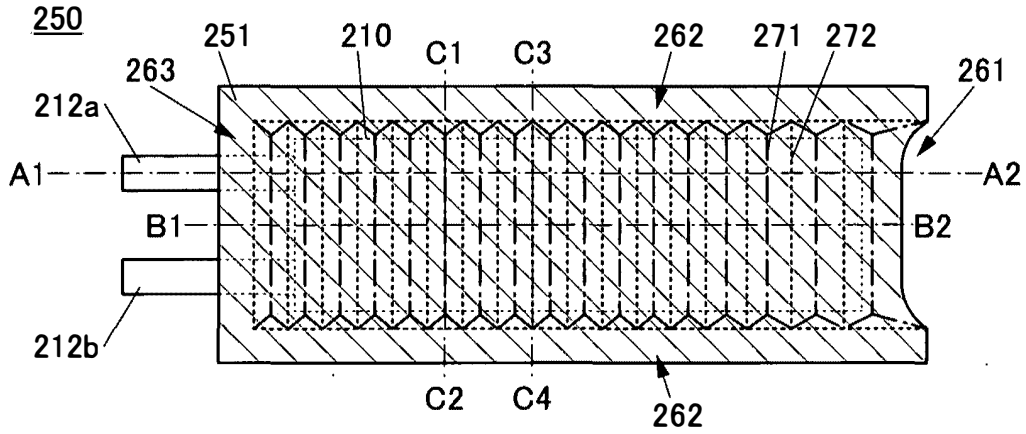


FIG. 22B1

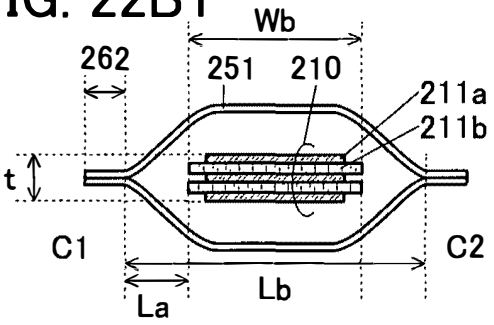


FIG. 22B2

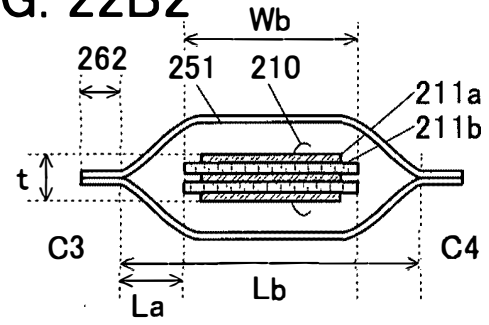


FIG. 22C

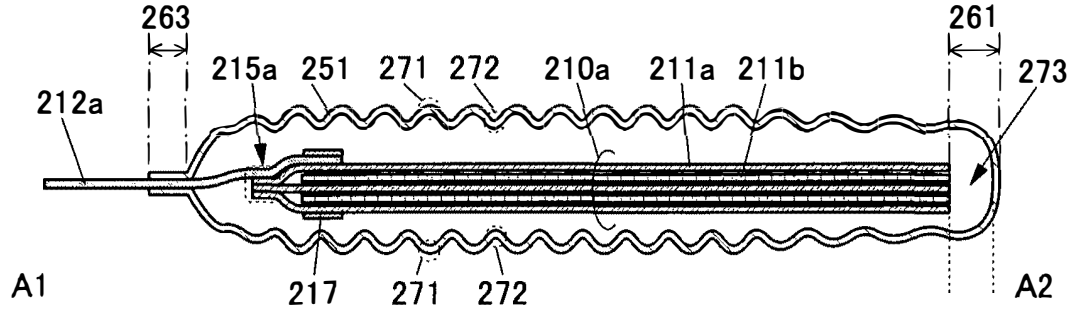


FIG. 22D

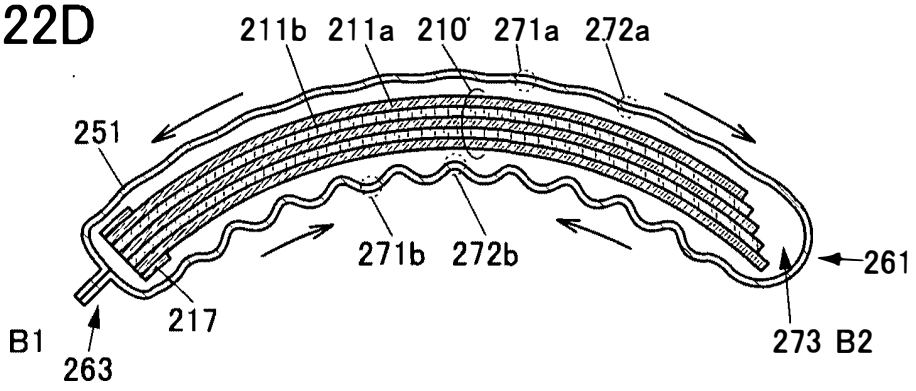


FIG. 23A

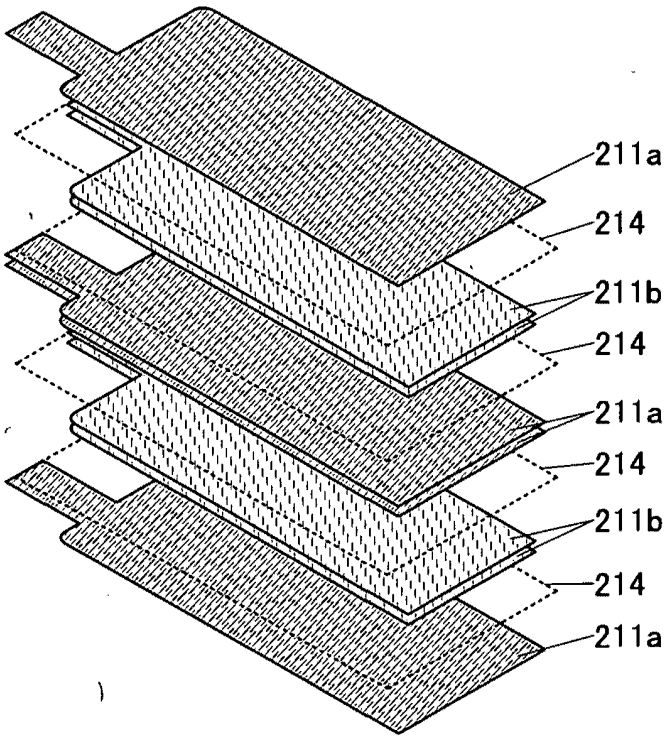


FIG. 23B

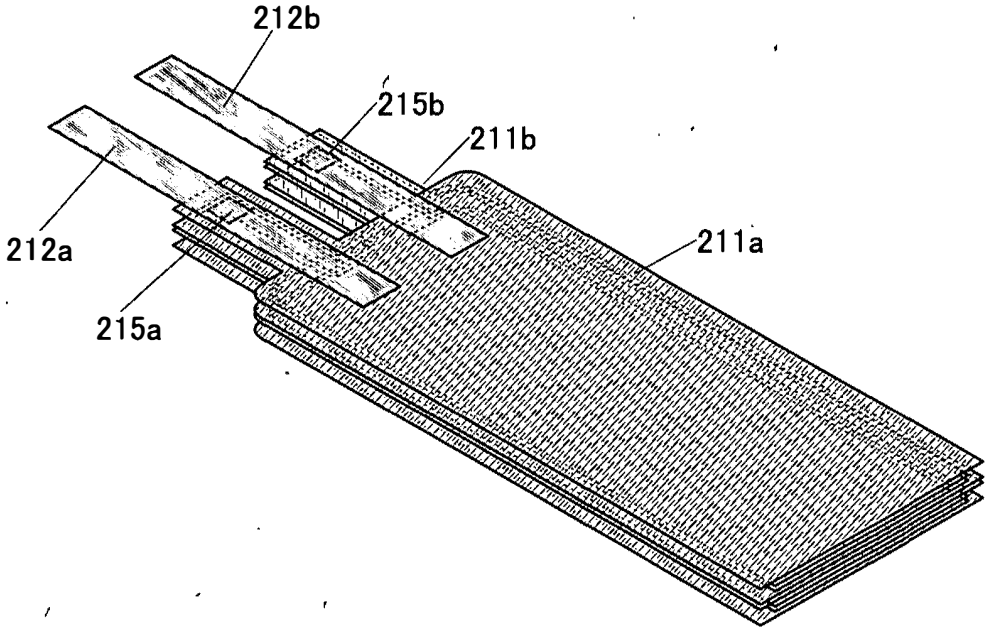


FIG. 25A

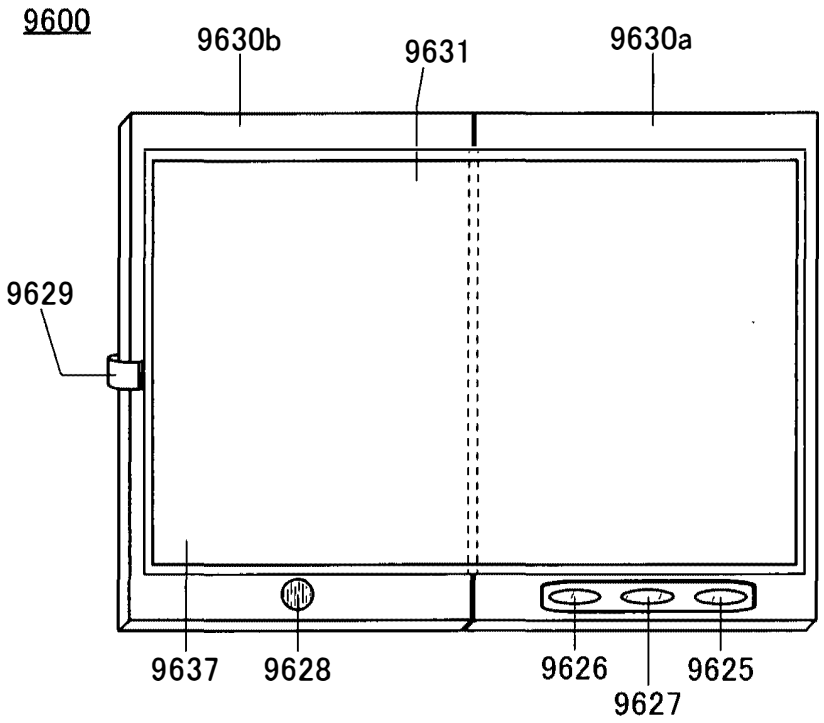


FIG. 25B

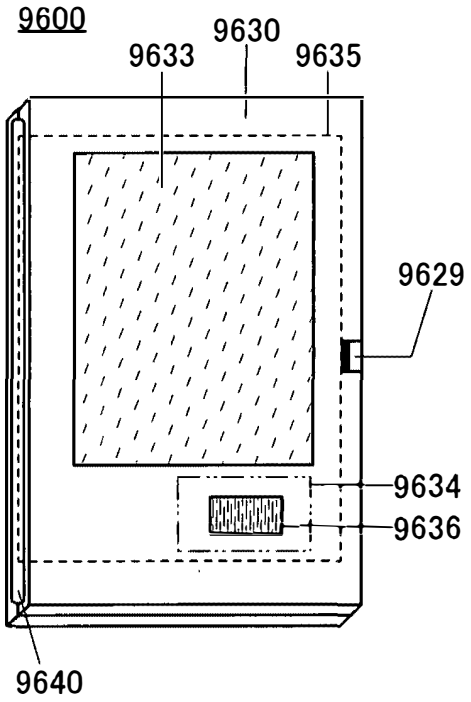


FIG. 25C

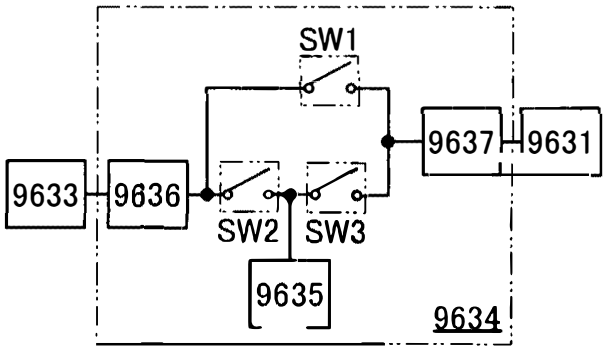


FIG. 27A

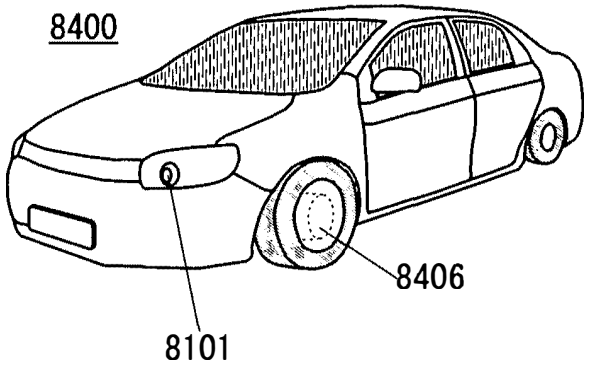


FIG. 27B

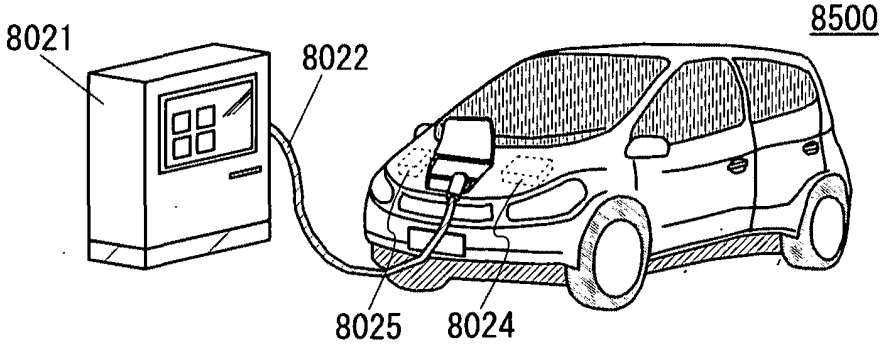
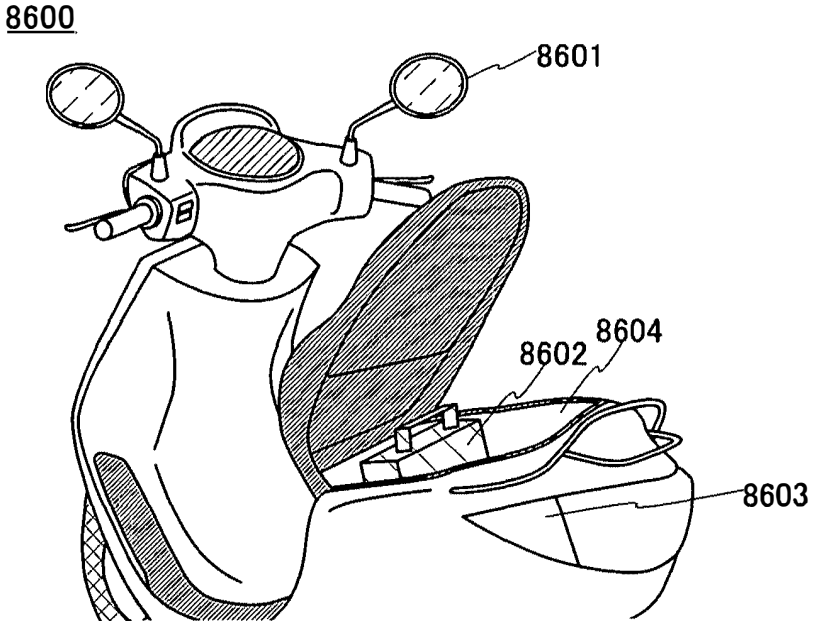


FIG. 27C



before Ti heating (Sample06)

FIG. 37A

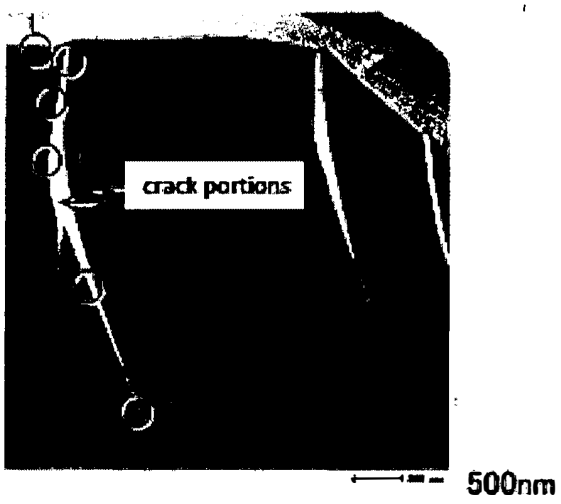
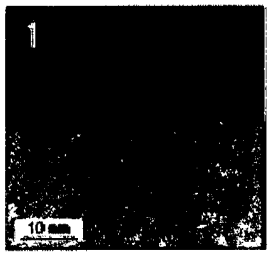


FIG. 37B1



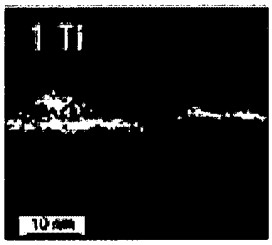
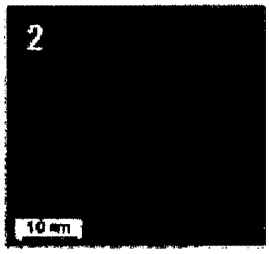


FIG. 37B2

FIG. 37C1



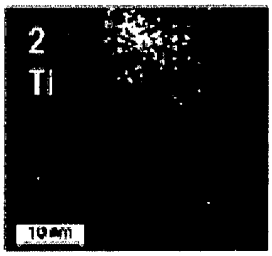
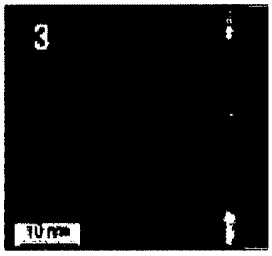


FIG. 37C2

FIG. 37D1



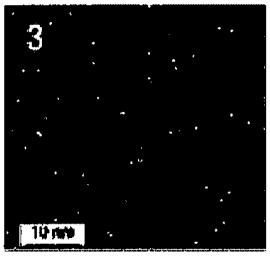
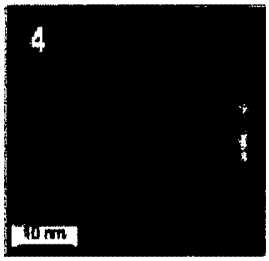


FIG. 37D2

FIG. 37E1



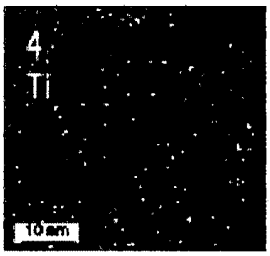


FIG. 37E2

after Ti heating (Sample07)

FIG. 38A

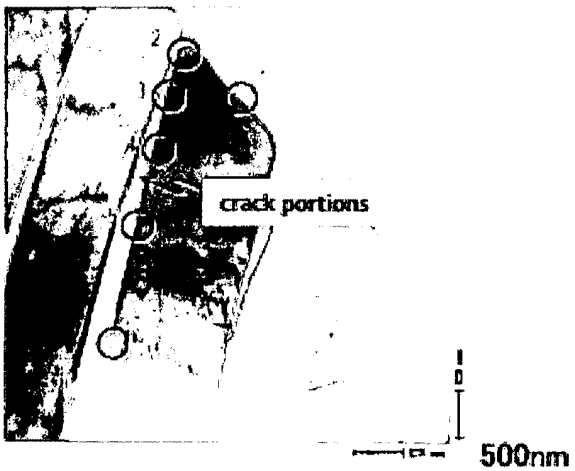
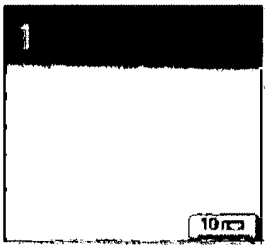


FIG. 38B1



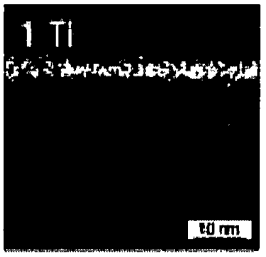
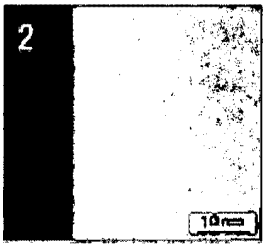


FIG. 38B2

FIG. 38C1



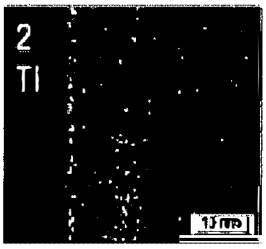
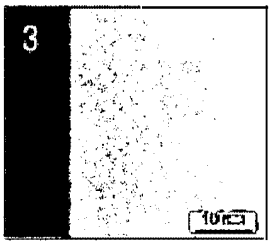


FIG. 38C2

FIG. 38D1



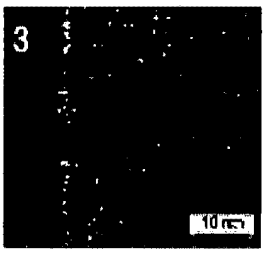
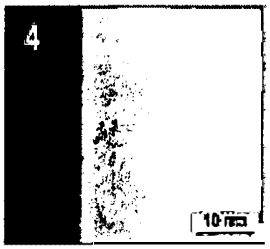


FIG. 38D2

FIG. 38E1



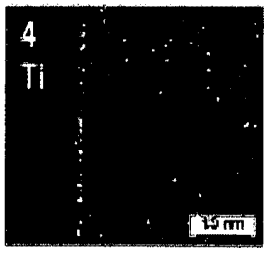


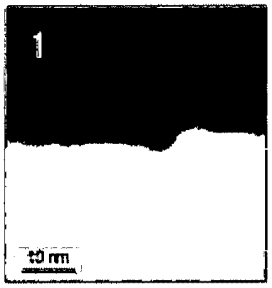
FIG. 38E2

before Mg heating (Sample06)

FIG. 39A



FIG. 39B1



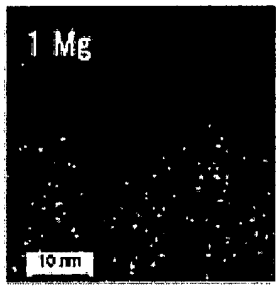
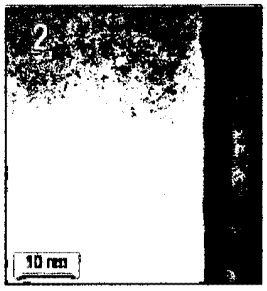


FIG. 39B2

FIG. 39C1



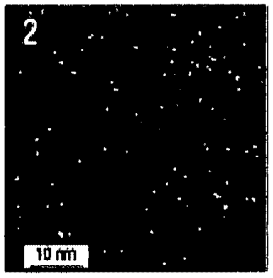
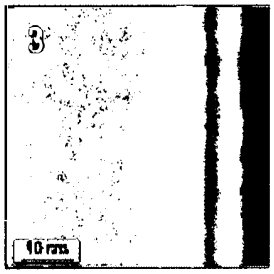


FIG. 39C2

FIG. 39D1



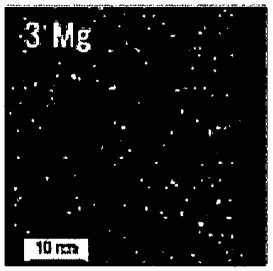
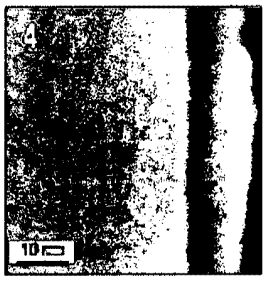


FIG. 39D2

FIG. 39E1



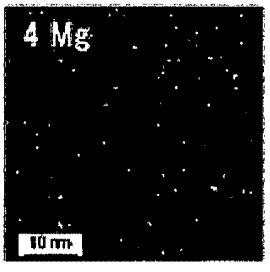


FIG. 39E2

after Mg heating (Sample07)

FIG. 40A

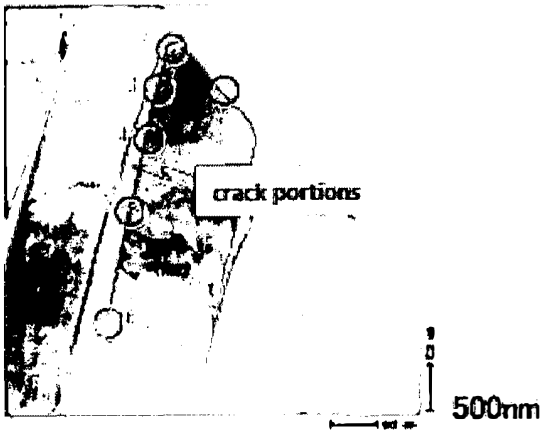
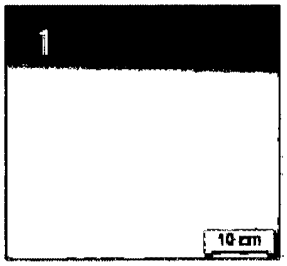


FIG. 40B1



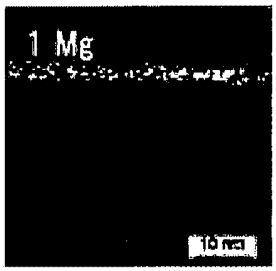
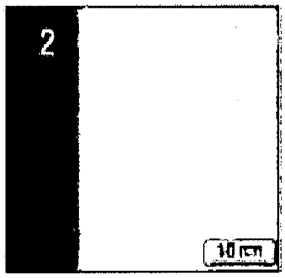


FIG. 40B2

FIG. 40C1



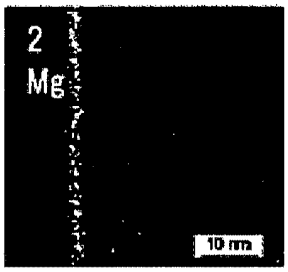
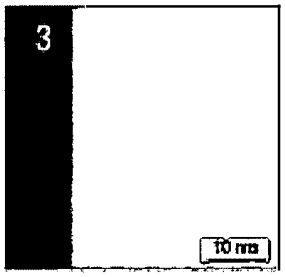


FIG. 40C2

FIG. 40D1



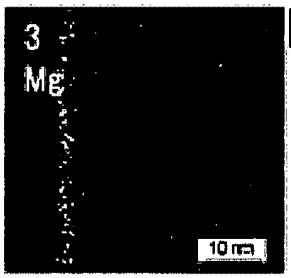


FIG. 40D2

FIG. 40E1

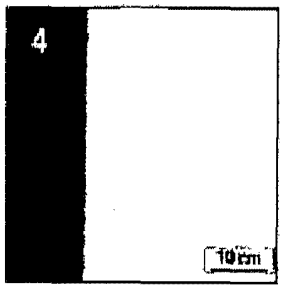
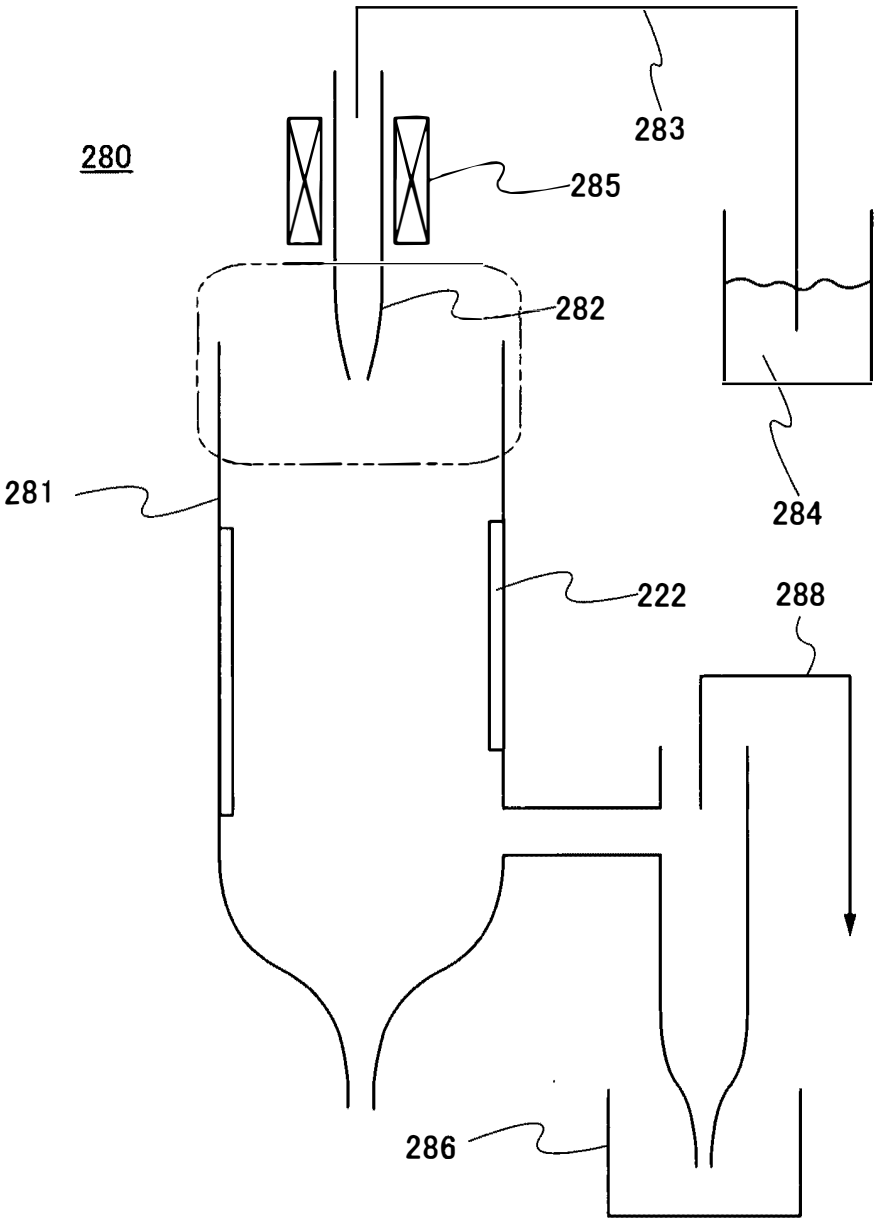




FIG. 40E2

FIG. 54



OPAP AKIMA-3
JUN 14 2019
PATENT & TRADEMARK OFFICE

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application	)	
Teruaki **OCHIAI** et al.	)	Group Art Unit: 1725
Application No.: 15/638,449	)	Examiner: Sean P. Cullen
Filed: June 30, 2017	)	Confirmation No.: 1074
For: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE **(As Currently Amended)**	)	

INFORMATION DISCLOSURE STATEMENT

Mail Stop Amendment – Via EFS
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In accordance with the duty of disclosure as set forth in 37 C.F.R. § 1.56, Applicant hereby submits the following information in conformance with 37 C.F.R. §§ 1.97 and 1.98. In accordance with the policies of the U.S. Patent and Trademark Office, only copies of newly cited foreign patents and non-patent literature are being submitted.

Applicant notes that the *Foreign Patent Documents*: JP-2002-164053; JP-08-100107; JP-07-316551 and JP-2015-527707, except the family documents expressly noted below, were previously cited by Japan Patent Office in a Japanese Office Action mailed **April 23, 2019** issued in a counterpart Japanese Application No. 2017-132227.

Applicants also wish to bring to the Examiner's attention the following family information listed on the attached PTO-1449.

1. US-6,984,469 is in the **same family** as JP-2002-164053, but was **not cited** in the above noted foreign communication.

2. US-5,604,396 and EP-0 638 625 are in the **same family** as JP-07-316551, but were **not cited** in the above noted foreign communication.

3. US-9,478,796 and WO-2014-015139 are in the **same family** as JP-2015-527707, but were **not cited** in the above noted foreign communication.

The undersigned certifies that each item of information contained in this information disclosure statement **with the exception of the family document(s) noted above and included on the attached Form PTO-1449** was first cited in any communication from a foreign patent office in connection with a counterpart foreign application not more than three (3) months prior to the filing of this statement.

It is requested that the accompanying Form PTO-1449 be considered and made of record in the above-identified application and that an Examiner-initialed copy of this form be returned to the undersigned.

The present Information Disclosure Statement is submitted before issuance of an action closing prosecution and is certified under 37 C.F.R. § 1.97(e)(1); therefore, no fee is believed to be necessary at this time.

However, if a fee becomes necessary, the Commissioner is hereby authorized to charge fees required in connection with this filing or credit any overpayment to Deposit Account No. 19-2380.

Respectfully submitted,

/Jeffrey L. Costellia, Reg. No. 35,483/
Jeffrey L. Costellia
Registration No. 35,483

NIXON PEABODY LLP
CUSTOMER NO.: 22204
799 9th Street, N.W., Suite 500
Washington, DC 20001-4501
Tel: 202-585-8000

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JUN 14 2019
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PTO/SB/08A (07-06)
Approved for use through 03/31/2007. OMB 0651-0031
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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 1

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	740756-4563

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-6,984,469	01-10-2006	Kweon.H et al.	
		US-5,604,396	02-18-1997	Watanabe.A et al.	
		US-9,478,796	10-25-2016	Li.L et al.	

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2002-164053A	06-07-2002			Abst.
		JP-08-100107A	04-16-1996			Abst.
		JP-07-316551A	12-05-1995			Abst.
		JP-2015-527707	09-17-2015			Abst.
		EP-0638625A	02-15-1995			Eng.
		WO-2014/015139	01-23-2014			Eng.

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant. 1 Applicant's unique citation designation number (optional). 2 See Kinds Codes of USPTO Patent Documents at www.uspto.gov or MPEP 901.04. 3 Enter Office that issued the document, by the two-letter code (WIPO Standard ST.3). 4 For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 5 Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. 6 Applicant is to place a check mark here if English language Translation is attached.

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 2 hours to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.

DocCode - SCORE

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Application Number: 15638449 Document Date: 06/14/2019

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PTO/SB/06 (09-11)
Approved for use through 1/31/2014. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
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PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 15/638,449	Filing Date 06/30/2017	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $80 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $420 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	06/14/2019	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 15	Minus	** 22	= 0		x $100 =	0
	Independent (37 CFR 1.16(h))	* 3	Minus	*** 4	= 0		x $460 =	0
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	0

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	SLIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/ANISSA D DEVLIN/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

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15/638,449	06/30/2017	Teruaki OCHIAI	740756-004563	1074

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EXAMINER	
CULLEN, SEAN P	

ART UNIT	PAPER NUMBER
1725	

NOTIFICATION DATE	DELIVERY MODE
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Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

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Office Action Summary	Application No. 15/638,449	Applicant(s) OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTHS FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☑ Responsive to communication(s) filed on 14 June 2019.

☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2a)☑ This action is **FINAL.** 2b)☐ This action is non-final.

3)☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

4)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims*

5) ☑ Claim(s) 1-6 and 8-16 is/are pending in the application.

5a) Of the above claim(s) _____ is/are withdrawn from consideration.

6) ☐ Claim(s) _____ is/are allowed.

7) ☑ Claim(s) 1-6 and 8-16 is/are rejected.

8) ☐ Claim(s) _____ is/are objected to.

9) ☐ Claim(s) _____ are subject to restriction and/or election requirement

* If any claims have been determined allowable, you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see http://www.uspto.gov/patents/init_events/pph/index.jsp or send an inquiry to **PPHfeedback@uspto.gov.**

Application Papers

10)☑ The specification is objected to by the Examiner.

11)☑ The drawing(s) filed on 14 June 2019 is/are: a)☐ accepted or b)☑ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

Certified copies:

a)☐ All b)☐ Some** c)☐ None of the:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

** See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☑ Notice of References Cited (PTO-892)

2) ☑ Information Disclosure Statement(s) (PTO/SB/08a and/or PTO/SB/08b) Paper No(s)/Mail Date _____.

3) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date _____.

4) ☐ Other: _____.

DETAILED ACTION

Notice of Pre-AIA or AIA Status

1. The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

Status of Claims and Other Notes

2. **Claim(s) 1-6 and 8-16** is/are pending.

3. **Claim(s) 7 and 17-22** is/are canceled.

4. The paragraph numbers cited in this Office Action in reference to the instant application are referring to the paragraph numbering of substitute specification filed on 14 June 2019.

5. The text of those sections of Title 35, U.S. Code not included in this action can be found in a prior Office action.

Information Disclosure Statement

6. The information disclosure statement (IDS) submitted on 08 March 2019 was filed before the mailing of a first Office Action on the merits. The submission complies with the provisions of 37 CFR 1.97. Accordingly, the information disclosure statement is being considered by the examiner.

7. The information disclosure statement (IDS) submitted on 14 June 2019 was filed after the mailing date of the first Office Action on the merits on 14 March 2019. The submission complies with the provisions of 37 CFR 1.97. Accordingly, the information disclosure statement is being considered by the examiner.

Drawings

8. The drawings were received on 14 June 2019. These drawings are unacceptable.

9. The drawings are objected to as failing to comply with 37 CFR 1.84(p)(4) because:

Reference character **8101** has been used to designate both a housing of a lighting device (e.g., Fig. 26) and a headlight (e.g., Fig. 27A). Reference character **8401** should be used to designate a headlight in Fig. 27A.

Reference character **615** has been used to designate both a module (e.g., Figs. 12C and 12D) and a temperature control device (e.g., Fig. 12D). Reference character **617** should be used to designate a temperature control device in Fig. 12D.

Corrected drawing sheets in compliance with 37 CFR 1.121(d) are required in reply to the Office action to avoid abandonment of the application. Any amended replacement drawing sheet should include all of the figures appearing on the immediate prior version of the sheet, even if only one figure is being amended. Each drawing sheet submitted after the filing date of an application must be labeled in the top margin as either "Replacement Sheet" or "New Sheet" pursuant to 37 CFR 1.121(d). If the changes are not accepted by the examiner, the applicant will be notified and informed of any required corrective action in the next Office action. The objection to the drawings will not be held in abeyance.

10. The drawings are objected to as failing to comply with 37 CFR 1.84(p)(5) because they do not include the following reference sign(s) mentioned in the description:

617 (e.g., [0310], [0615]),

11a (e.g., [0355]-[0360], [0362]-[0369], [0373], [0376]-[0378], [0615]),

11b (e.g., [0355]-[0360], [0362]-[0369], [0373], [0376]-[0378], [0615]),

12a (e.g., [0355], [0356], [0360], [0362], [0373], [0615]),

12b (e.g., [0355]-[0360], [0362], [0373], [0615]),

14 (e.g., [0356], [0357], [0359], [0615]),

15a (e.g., [0360], [0615]),

15b (e.g., [0360], [0615]),

17 (e.g., [0376], [0615]),

50 (e.g., [0355]-[0357], [0364], [0365], [0368], [0373]-[0378], [0615]),

51 (e.g., [0355], [0361]-[0363], [0365], [0368], [0373], [0375], [0377], [0615]),

61 (e.g., [0362], [0373], [0376], [0615]),

62 (e.g., [0362], [0363], [0365], [0366], [0368], [0369], [0615]),

71 (e.g., [0363], [0364], [0371], [0615]),

72 (e.g., [0363], [0364], [0615]),

73 (e.g., [0373], [0377], [0615]),

9631a (e.g., [0400], [0402], [0403], [0406], [0615]),

9631b (e.g., [0400], [0403], [0406], [0615]), and

8401 (e.g., [0428], [0615]).

Corrected drawing sheets in compliance with 37 CFR 1.121(d) are required in reply to the Office action to avoid abandonment of the application. Any amended replacement drawing sheet should include all of the figures appearing on the immediate prior version of the sheet, even if only one figure is being amended. Each drawing sheet submitted after the filing date of an application must be labeled in the top margin as either "Replacement Sheet" or "New Sheet" pursuant to 37 CFR 1.121(d). If the changes are not accepted by the examiner, the applicant will be notified and informed of any required corrective action in the next Office action. The objection to the drawings will not be held in abeyance.

11. The drawings are objected to as failing to comply with 37 CFR 1.84(p)(5) because they include the following reference character(s) not mentioned in the description:

210 (e.g., Figs. 22A, 22B1, 22B2, 22C, and 22D),

211a (e.g., Figs. 22B1, 22B2, 22C, 22D, 23A, and 23B),

211b (e.g., Figs. 22B1, 22B2, 22C, 22D, 23A, and 23B),

212a (e.g., Figs. 22A, 22C, and 23B),

212b (e.g., Figs. 22A and 23B),

215a (e.g., Fig. 22C),

215b (e.g., Fig. 23B),

217 (e.g., Figs. 22C and 22D),

250 (e.g., Fig. 22A),

251 (e.g., Figs. 22A, 22B1, 22B2, 22C, and 22D),

261 (e.g., Figs. 22A, 22C, and 22D),

262 (e.g., Figs. 22A, 22B1, and 22B2),

271 (e.g., Figs. 22A and 22C),

272 (e.g., Figs. 22A and 22C),

271a (e.g., Fig. 22D),

271b (e.g., Fig. 22D),

272a (e.g., Fig. 22D),

272b (e.g., Fig. 22D),

273 (e.g., Figs. 22C and 22D), and

222 (e.g., Fig. 54).

Corrected drawing sheets in compliance with 37 CFR 1.121(d), or amendment to the specification to add the reference character(s) in the description in compliance with 37 CFR 1.121(b) are required in reply to the Office action to avoid abandonment of the application. Any amended replacement drawing sheet should include all of the figures appearing on the immediate prior version of the sheet, even if only one figure is being amended. Each drawing sheet submitted after the filing date of an application must be labeled in the top margin as either "Replacement Sheet" or "New Sheet" pursuant to 37 CFR 1.121(d). If the changes are not accepted by the examiner, the applicant will be notified and informed of any required corrective action in the next Office action. The objection to the drawings will not be held in abeyance.

Specification

12. The disclosure is objected to because of the following informalities:

Reference character **617** is described in paragraphs [0310] and [0615]. However, reference character **617** is not present in the drawings.

Reference character **11a** is described in paragraphs [0355]-[0360], [0362]-[0369], [0373], [0376]-[0378], and [0615]. However, reference character **11a** is not present in the drawings.

Reference character **11b** is described in paragraphs [0355]-[0360], [0362]-[0369], [0373], [0376]-[0378], and [0615]. However, reference character **11b** is not present in the drawings.

Reference character **12a** is described in paragraphs [0355], [0356], [0360], [0362], [0373], and [0615]. However, reference character **12a** is not present in the drawings.

Reference character **12b** is described in paragraphs [0355]-[0360], [0362], [0373], and [0615]. However, reference character **12b** is not present in the drawings.

Reference character **14** is described in paragraphs [0356], [0357], [0359], and [0615]. However, reference character **14** is not present in the drawings.

Reference character **15a** is described in paragraphs [0360] and [0615]. However, reference character **15a** is not present in the drawings.

Reference character **15b** is described in paragraphs [0360] and [0615]. However, reference character **15b** is not present in the drawings.

Reference character **17** is described in paragraphs [0376], and [0615]. However, reference character **17** is not present in the drawings.

Reference character **50** is described in paragraphs [0355]-[0357], [0364], [0365], [0368], [0373]-[0378], and [0615]. However, reference character **50** is not present in the drawings.

Reference character **51** is described in paragraphs [0355], [0361]-[0363], [0365], [0368], [0373], [0375], [0377], and [0615]. However, reference character **51** is not present in the drawings.

Reference character **61** is described in paragraphs [0362], [0373], [0376], and [0615]. However, reference character **61** is not present in the drawings.

Reference character **62** is described in paragraphs [0362], [0363], [0365], [0366], [0368], [0369], and [0615]. However, reference character **62** is not present in the drawings.

Reference character **71** is described in paragraphs [0363], [0364], [0371], and [0615]. However, reference character **71** is not present in the drawings.

Reference character **72** is described in paragraphs [0363], [0364], and [0615]. However, reference character **72** is not present in the drawings.

Reference character **73** is described in paragraphs [0373], [0377], and [0615]. However, reference character **73** is not present in the drawings.

Reference character **9631a** is described in paragraphs [0400], [0402], [0403], [0406], and [0615]. However, reference character **9631a** is not present in the drawings.

Reference character **9631b** is described in paragraphs [0400], [0403], [0406], and [0615]. However, reference character **9631b** is not present in the drawings.

Reference character **8401** is described in paragraphs [0428] and [0615]. However, reference character **8401** is not present in the drawings.

Appropriate correction is required.

Claim Rejections - 35 USC § 112

13. Applicants' amendment(s) have overcome the rejection(s) of **claim(s) 1-12** under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph.

14. **Claim(s) 13-16** is/are rejected under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

Claim 13 recites the limitation "a superficial portion of the positive electrode active material particle" in lines 5-6. Claim 13 has previously recited the limitation "a superficial portion of the positive electrode active material particle" in lines 3-4. It is unclear if "a superficial portion" recited in line 5 is further limiting "a superficial portion" recited in line 3.

Claims 14 and 15 are directly or indirectly dependent from claim 13 and include all the limitations of claim 13. Therefore, claims 14 and 15 are also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

Claim 16 recites the limitation "the positive electrode active material particle according to claim 13" and includes all the limitations of claim 13. Therefore, claim 15 is also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

Claim Rejections - 35 USC § 102

15. **Claim(s) 1, 4, 6, 8, and 13-16** is/are rejected under 35 U.S.C. 102(a)(1) as being anticipated by Ooyama et al. (US 2007/0224506 A1, hereinafter Ooyama).

Regarding **claim 1**, Ooyama discloses a positive electrode active material comprising:

- a first region in the positive electrode active material (see complex oxide particles, [0022]);
- a second region (see coating layer, [0021]) in the positive electrode active material (see cathode active material, [0032]),
- the second region (see coating layer, [0021]) surrounding the first region see complex oxide particles, [0022]); and
- a third region (see surface layer, [0030]) in the positive electrode active material (see cathode active material, [0032]),
- the third region (see surface layer, [0030]) surrounding the second region (see coating layer, [0021]),
- wherein the first region comprises a first compound comprising an oxide of lithium and a first transition metal (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),
- wherein a type of a crystal structure of the first compound is a layered rock-salt crystal structure (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),

- wherein the second region comprises a second compound comprising an oxide of a second transition metal (see oxide, [0035]-[0036]),
- wherein a type of a crystal structure of the second compound is a rock-salt crystal structure (see oxide, [0035]-[0036]),
- wherein the second compound is a non-stoichiometric compound (see coating layer, [0027]),
- wherein the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride (see magnesium, [0030]), and
- wherein a type of a crystal structure of the third compound is the rock-salt crystal structure (see oxide, [0030]).

Regarding **claim 4**, Ooyama discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the first compound is a polycrystal (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]).

Regarding **claim 6**, Ooyama discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the third compound essentially consists of the representative elements (see surface layer, [0030]).

Regarding **claim 8**, Ooyama discloses a secondary battery comprising a positive electrode (21, [0044]) comprising a positive electrode active material; and a negative electrode (22, [0044]), wherein the positive electrode active material comprises:

- a first region in the positive electrode active material (see complex oxide particles, [0022]);

- a second region (see coating layer, [0021]) in the positive electrode active material (see cathode active material, [0032]),
- the second region (see coating layer, [0021]) surrounding the first region see complex oxide particles, [0022]); and
- a third region (see surface layer, [0030]) in the positive electrode active material (see cathode active material, [0032]),
- the third region (see surface layer, [0030]) surrounding the second region (see coating layer, [0021]),
- wherein the first region comprises a first compound comprising an oxide of lithium and a first transition metal (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),
- wherein a type of a crystal structure of the first compound is a layered rock-salt crystal structure (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),
- wherein the second region comprises a second compound comprising an oxide of a second transition metal (see oxide, [0035]-[0036]),
- wherein a type of a crystal structure of the second compound is a rock-salt crystal structure (see oxide, [0035]-[0036]),
- wherein the second compound is a non-stoichiometric compound (see coating layer, [0027]),
- wherein the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride (see magnesium, [0030]), and
- wherein a type of a crystal structure of the third compound is the rock-salt crystal structure (see oxide, [0030]).

Regarding **claim 13**, Ooyama discloses a positive electrode active material particle comprising:

- lithium (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]), titanium, cobalt (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]), magnesium ([0028], [0030]), oxygen (see oxide, [0030]), and fluorine (see fluoride, [0035]),
- wherein when a concentration of cobalt in a superficial portion of the positive electrode active material particle is 1 (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),
- a concentration of titanium in the superficial portion of the positive electrode active material particle is greater than or equal to 0.05 and less than or equal to 0.4 (see surface layer, [0030]),
- a concentration of magnesium in the superficial portion of the positive electrode active material particle is greater than or equal to 0.4 and less than or equal to 1.5 (see surface layer, [0030]), and
- a concentration of fluorine in the superficial portion of the positive electrode active material particle is greater than or equal to 0.05 and less than or equal to 1.5 (see coating layer, [0029]).

Regarding **claim 14**, Ooyama discloses all claim limitations set forth above and further discloses a positive electrode active material further comprising:

- a first region in the positive electrode active material particle (see complex oxide particles, [0022]);
- a second region (see coating layer, [0021]) in the positive electrode active material particle (see cathode active material, [0032]),

- the second region (see coating layer, [0021]) surrounding the first region (see complex oxide particles, [0022]); and
- a third region (see surface layer, [0030]) in the positive electrode active material (see cathode active material, [0032]),
- the third region (see surface layer, [0030]) surrounding the second region (see coating layer, [0021]),
- wherein the third region (see surface layer, [0030]) is positioned closer to a surface of the positive electrode active material particle than the second region is (see coating layer, [0030]),
- wherein the first region comprises a first compound comprising lithium and cobalt (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),
- wherein the second region comprises a second compound comprising titanium (see coating layer, [0028]),
- wherein the second compound is a non-stoichiometric compound (see coating layer, [0027]), and
- wherein the third region comprises a third compound comprising magnesium (see surface layer, [0030]) and fluorine (see fluoride, [0036]).

Regarding **claim 15**, Ooyama discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein a type of a crystal structure of the first compound is a layered rock-salt crystal structure (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),
- wherein a type of a crystal structure of the second compound is a rock-salt crystal structure (see oxide, [0035]-[0036]), and

- wherein a type of a crystal structure of the third compound is the rock-salt crystal structure (see oxide, [0030]).

Regarding **claim 16**, Ooyama discloses a secondary battery comprising a positive electrode (21, [0044]) comprising a positive electrode active material; and a negative electrode (22, [0044]), wherein the positive electrode active material particle comprises:

- lithium (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]), titanium, cobalt (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]), magnesium ([0028], [0030]), oxygen (see oxide, [0030]), and fluorine (see fluoride, [0035]),
- wherein when a concentration of cobalt in a superficial portion of the positive electrode active material particle is 1 (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),
- a concentration of titanium in the superficial portion of the positive electrode active material particle is greater than or equal to 0.05 and less than or equal to 0.4 (see surface layer, [0030]),
- a concentration of magnesium in the superficial portion of the positive electrode active material particle is greater than or equal to 0.4 and less than or equal to 1.5 (see surface layer, [0030]), and
- a concentration of fluorine in the superficial portion of the positive electrode active material particle is greater than or equal to 0.05 and less than or equal to 1.5 (see coating layer, [0029]).

Claim Rejections - 35 USC § 103

16. **Claim(s) 2 and 3** is/are rejected under 35 U.S.C. 103 as being unpatentable over Ooyama (US 2007/0224506 A1) as applied to **claim(s) 1** above, and further in view of Yura et al. (US 2012/0258358 A1, hereinafter Yura).

Regarding **claims 2 and 3**, Ooyama discloses all claim limitations set forth above, but does not explicitly disclose a positive electrode active material:

- wherein a degree of a mismatch between the crystal structure of the first compound and the crystal structure of the second compound is less than or equal to 0.12, and
- wherein a degree of a mismatch between the crystal structure of the second compound and the crystal structure of the third compound is less than or equal to 0.12; and
- wherein a degree of a mismatch between a (1-1-4) plane of the crystal structure of the first compound or a plane orthogonal to the (1-1-4) plane and a [100] plane of the crystal structure of the second compound is less than or equal to 0.12, and
- wherein a degree of a mismatch between the [100] plane of the crystal structure of the second compound and a [100] plane of the crystal structure of the third compound is less than or equal to 0.12.

Yura discloses a positive electrode active material (222) having a degree of a mismatch between a crystal structure of a first compound (222a) and a crystal structure of the second compound (222a) is less than or equal to 0.12 (see crystal orientations, [0163]), and wherein a degree of a mismatch between the crystal structure of the second compound (222a) and a crystal structure of a third compound (222a) is less than or equal to 0.12 (see crystal orientations,

[0163]); and wherein a degree of a mismatch between a (1-1-4) plane of the crystal structure of the first compound (222a) or a plane orthogonal to the (1-1-4) plane and a [100] plane of the crystal structure of the second compound (222a) is less than or equal to 0.12 (see crystal orientations, [0163]), and wherein a degree of a mismatch between the [100] plane of the crystal structure of the second compound (222a) and a [100] plane of the crystal structure of the third compound (222a) is less than or equal to 0.12 (see crystal orientations, [0163]) to improve electron and lithium ion diffusion in order to improve the cell characteristics (see cathode active material, [0038]). Ooyama and Yura are analogous art because they are directed to positive electrode active materials for lithium secondary batteries. Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the positive electrode active material of Ooyama with the degree of mismatch of Yura to improve electron and lithium ion diffusion in order to improve the cell characteristics.

17. **Claim(s) 5** is/are rejected under 35 U.S.C. 103 as being unpatentable over Ooyama (US 2007/0224506 A1) as applied to **claim(s) 1** above, and further in view of Endo et al. (US 2010/0233542 A1, hereinafter Endo).

Regarding **claim 5**, Ooyama discloses all claim limitations set forth above, but does not explicitly disclose a positive electrode active material:

- wherein an average crystallite size of the oxide of lithium and the first compound is greater than or equal to 280 nm and less than or equal to 630 nm.

Endo discloses a positive electrode active material having an average crystallite size of an oxide of lithium and a first compound greater than or equal to 280 nm and less than or equal to 630 nm to improve the reversible capacity (see crystallite size, [0144]). Ooyama and Endo are

analogous art because they are directed to positive electrode active materials for lithium secondary batteries. Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the positive electrode active material of Ooyama with the average crystallite size of Endo in order to improve the reversible capacity.

18. **Claim(s) 9** is/are rejected under 35 U.S.C. 103 as being unpatentable over Ooyama (US 2007/0224506 A1) in view of Takahashi et al. (US 2004/0229123 A1, hereinafter Takahashi).

Regarding **claim 9**, Ooyama discloses a positive electrode active material comprising:

- a first region in the positive electrode active material (see complex oxide particles, [0022]);
- a second region (see coating layer, [0021]) in the positive electrode active material (see cathode active material, [0032]),
- the second region (see coating layer, [0021]) surrounding the first region (see complex oxide particles, [0022]); and
- a third region (see surface layer, [0030]) in the positive electrode active material (see cathode active material, [0032]),
- the third region (see surface layer, [0030]) surrounding the second region (see coating layer, [0021]),
- wherein the first region comprises lithium cobaltate (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]), and
- wherein the third region comprises magnesium oxide (see magnesium, [0030]).

Ooyama does not explicitly disclose:

- wherein the second region comprises lithium titanate.

Takahashi discloses a positive electrode active material comprising a second region including lithium titanate (see titanium compound, [0074]) to improve the high rate characteristics (see titanium, [0097]). Ooyama and Takahashi are analogous art because they are directed to positive electrode active materials for lithium secondary batteries. Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the second region of Ooyama with the lithium titanate in order to improve the high rate characteristics.

19. **Claim(s) 10-12** is/are rejected under 35 U.S.C. 103 as being unpatentable over Ooyama (US 2007/0224506 A1) in view of Takahashi (US 2004/0229123 A1) as applied to **claim(s) 9** above, and further in view of Yura (US 2012/0258358 A1).

Regarding **claims 10-12**, modified Ooyama discloses all claim limitations set forth above, but does not explicitly disclose a positive electrode active material:

- wherein a crystal orientation of a part of the lithium cobaltate of the first region and a crystal orientation of a part of the lithium titanate of the second region are substantially aligned with each other;
- wherein a crystal orientation of a part of the lithium titanate of the second region and a crystal orientation of a part of the magnesium oxide of the third region are substantially aligned with each other;
- wherein a degree of a mismatch between a crystal structure of the lithium cobaltate of the first region and a crystal structure of the lithium titanate of the second region is less than or equal to 0.12, and

- wherein a degree of a mismatch between the crystal structure of the lithium titanate of the second region and a crystal structure of the magnesium oxide of the third region is less than or equal to 0.12.

Yura discloses a positive electrode active material (222) having a crystal orientation of a part of a first compound (222a) of a first region and a crystal orientation of a part of a second compound (222a) of a second region are substantially aligned with each other (see crystal orientations, [0163]); wherein a crystal orientation of a part of the second compound of the second region and a crystal orientation of a part of a third compound (222a) of a third region are substantially aligned with each other (see crystal orientations, [0163]); wherein a degree of a mismatch between a crystal structure of the first compound of the first region and a crystal structure of the second compound of the second region is less than or equal to 0.12 (see crystal orientations, [0163]), and wherein a degree of a mismatch between the crystal structure of the second compound of the second region and a crystal structure of the third compound of the third region is less than or equal to 0.12 (see crystal orientations, [0163]) to improve electron and lithium ion diffusion in order to improve the cell characteristics (see cathode active material, [0038]). Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the positive electrode active material of Ooyama with the crystal orientation and degree of mismatch of Yura to improve electron and lithium ion diffusion in order to improve the cell characteristics.

Response to Arguments

20. Applicant's arguments filed 14 June 2019 have been fully considered but they are not persuasive.

Applicants argue that the drawings have been amended as proposed by the Examiner (P9/¶3). The Office respectfully disagrees. Fig. 26 has not been amended and Fig. 27A has not been amended to change reference character **8101**. Thus, reference character **8101** has been used to designate both a housing of a lighting device (e.g., Fig. 26) and a headlight (e.g., Fig. 27A). Figs. 12C and 12D have not been amended. Thus, reference character **615** has been used to designate both a module (e.g., Figs. 12C and 12D) and a temperature control device (e.g., Fig. 12D). Therefore, the drawings have not been amended as proposed by the Examiner.

Applicants argue that the drawings have been amended to address the § 1.84(p)(5) objection (P9/¶4). The Office respectfully disagrees. The following reference character(s) are mentioned in the substitute specification: **617** (e.g., [0310], [0615]), **11a** (e.g., [0355]-[0360], [0362]-[0369], [0373], [0376]-[0378], [0615]), **11b** (e.g., [0355]-[0360], [0362]-[0369], [0373], [0376]-[0378], [0615]), **12a** (e.g., [0355], [0356], [0360], [0362], [0373], [0615]), **12b** (e.g., [0355]-[0360], [0362], [0373], [0615]), **14** (e.g., [0356], [0357], [0359], [0615]), **15a** (e.g., [0360], [0615]), **15b** (e.g., [0360], [0615]), **17** (e.g., [0376], [0615]), **50** (e.g., [0355]-[0357], [0364], [0365], [0368], [0373]-[0378], [0615]), **51** (e.g., [0355], [0361]-[0363], [0365], [0368], [0373], [0375], [0377], [0615]), **61** (e.g., [0362], [0373], [0376], [0615]), **62** (e.g., [0362], [0363], [0365], [0366], [0368], [0369], [0615]), **71** (e.g., [0363], [0364], [0371], [0615]), **72** (e.g., [0363], [0364], [0615]), **73** (e.g., [0373], [0377], [0615]), **9631a** (e.g., [0400], [0402], [0403], [0406], [0615]), **9631b** (e.g., [0400], [0403], [0406], [0615]), and **8401** (e.g., [0428], [0615]). The drawings have not been amended to include the above-mentioned reference characters. Further, the specification has not been amended to include the following reference character(s) that are present in the original and/or replacement drawings: **210** (e.g., Figs. 22A, 22B1, 22B2, 22C, and 22D), **211a** (e.g., Figs. 22B1, 22B2, 22C, 22D, 23A, and 23B), **211b** (e.g.,

Figs. 22B1, 22B2, 22C, 22D, 23A, and 23B), **212a** (e.g., Figs. 22A, 22C, and 23B), **212b** (e.g., Figs. 22A and 23B), **215a** (e.g., Fig. 22C), **215b** (e.g., Fig. 23B), **217** (e.g., Figs. 22C and 22D), **250** (e.g., Fig. 22A), **251** (e.g., Figs. 22A, 22B1, 22B2, 22C, and 22D), **261** (e.g., Figs. 22A, 22C, and 22D), **262** (e.g., Figs. 22A, 22B1, and 22B2), **271** (e.g., Figs. 22A and 22C), **272** (e.g., Figs. 22A and 22C), **271a** (e.g., Fig. 22D), **271b** (e.g., Fig. 22D), **272a** (e.g., Fig. 22D), **272b** (e.g., Fig. 22D), **273** (e.g., Figs. 22C and 22D), and **222** (e.g., Fig. 54). Therefore, the drawings have not been amended the figures to address the § 1.84(p)(5) objection.

Applicants argue amendments to the specification has been amended as requested by the Examiner (P10/¶1). The Office respectfully disagrees. The following reference character(s) are mentioned in the substitute specification: **617** (e.g., [0310], [0615]), **11a** (e.g., [0355]-[0360], [0362]-[0369], [0373], [0376]-[0378], [0615]), **11b** (e.g., [0355]-[0360], [0362]-[0369], [0373], [0376]-[0378], [0615]), **12a** (e.g., [0355], [0356], [0360], [0362], [0373], [0615]), **12b** (e.g., [0355]-[0360], [0362], [0373], [0615]), **14** (e.g., [0356], [0357], [0359], [0615]), **15a** (e.g., [0360], [0615]), **15b** (e.g., [0360], [0615]), **17** (e.g., [0376], [0615]), **50** (e.g., [0355]-[0357], [0364], [0365], [0368], [0373]-[0378], [0615]), **51** (e.g., [0355], [0361]-[0363], [0365], [0368], [0373], [0375], [0377], [0615]), **61** (e.g., [0362], [0373], [0376], [0615]), **62** (e.g., [0362], [0363], [0365], [0366], [0368], [0369], [0615]), **71** (e.g., [0363], [0364], [0371], [0615]), **72** (e.g., [0363], [0364], [0615]), **73** (e.g., [0373], [0377], [0615]), **9631a** (e.g., [0400], [0402], [0403], [0406], [0615]), **9631b** (e.g., [0400], [0403], [0406], [0615]), and **8401** (e.g., [0428], [0615]). The drawings have not been amended to include the above-mentioned reference characters. Therefore, the specification has not been amended as requested by the Examiner.

Applicants argue that a cathode active material of Ooyama is not a positive electrode active material (P11/¶3). The instant application describes an electrode with a high reaction

potential is called a positive electrode and an electrode with a low reaction potential is called a negative electrode (e.g., [0304]). The cathode of Ooyama is an electrode with a high reaction potential (see cathode active material, [0022]) and the anode is an electrode with a low reaction potential (see anode material, [0051]). The term "cathode active material" is completely consistent with definition of "positive electrode active material" of the instant specification. Therefore, the cathode active material of Ooyama is a positive electrode active material.

Applicants argue that paragraph [0022] of Ooyama does not teach "a type of crystal structure of the first compound" (P11/¶4). Paragraph [0022] discloses the chemical formula of the first compound (see complex oxide particles, [0022]). "Products of identical chemical composition can not have mutually exclusive properties." A chemical composition and its properties are inseparable. Therefore, if the prior art teaches the identical chemical structure, the properties applicant discloses and/or claims are necessarily present. *In re Spada*, 911 F.2d 705, 709, 15 USPQ2d 1655, 1658 (Fed. Cir. 1990). As Ooyama discloses the same chemical composition, Ooyama discloses the properties of the chemical composition. Therefore, paragraph [0022] of Ooyama does teach "a type of crystal structure of the first compound."

Applicants argue that paragraphs [0035] and [0036] of Ooyama does not teach "a type of crystal structure of the second compound" (P11/¶4). Paragraph [0035] and [0036] discloses the chemical compounds of the second compound. "Products of identical chemical composition can not have mutually exclusive properties." A chemical composition and its properties are inseparable. Therefore, if the prior art teaches the identical chemical structure, the properties applicant discloses and/or claims are necessarily present. *In re Spada*, 911 F.2d 705, 709, 15 USPQ2d 1655, 1658 (Fed. Cir. 1990). The instant application describes the second compound with a rock-salt crystal structure is one of titanium oxide, vanadium oxide, manganese oxide,

iron oxide, copper oxide, chromium oxide, niobium oxide, cobalt oxide, zinc oxide, and the like. Ooyama describes the second compound can be manganese oxide. As Ooyama discloses the same chemical composition, Ooyama discloses the properties of the chemical composition. Therefore, paragraphs [0035] and [0036] of Ooyama does teach "a type of crystal structure of the second compound.

Applicants argue that paragraph [0030] of Ooyama does not teach "the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride" (P11/¶4). Paragraph [0030] describes a third region including an oxide of magnesium (see surface layer, [0030]). An oxide of magnesium is a magnesium oxide. Ooyama also explicitly discloses MgO ([0083], [0085], and Table 1), which is magnesium oxide. Therefore, paragraph [0030] of Ooyama does teach "the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride."

Applicants argue that paragraph [0030] does not teach "a type of a crystal structure of the third compound" (P11/¶4). Paragraph [0030] describes an oxide of magnesium (see surface layer, [0030]). An oxide of magnesium is a magnesium oxide. Ooyama also explicitly discloses MgO ([0083], [0085], and Table 1), which is magnesium oxide. "Products of identical chemical composition can not have mutually exclusive properties." A chemical composition and its properties are inseparable. Therefore, if the prior art teaches the identical chemical structure, the properties applicant discloses and/or claims are necessarily present. *In re Spada*, 911 F.2d 705, 709, 15 USPQ2d 1655, 1658 (Fed. Cir. 1990). As Ooyama discloses the same chemical composition, Ooyama discloses the properties of the chemical composition. Therefore, paragraph [0030] does not teach "a type of a crystal structure of the third compound."

Applicants argue that paragraphs [0020], [0028], [0035], or [0029] is not related to "the surface layer" (P11/¶5). The coating layer and the surface layer of Ooyama correspond to "a superficial portion." The Office Action described part of the complex oxide particle, the coating layer and the surface layer as components of the superficial portion. See page 18, lines 3 -5, page 20, lines 20-22, and page 21, lines 11-13. Paragraphs [0020], [0028], [0035], and [0029] are related to the complex oxide particle, the coating layer and the surface layer. Therefore, paragraphs [0020], [0028], [0035], and [0029] are related to "a superficial portion."

Applicants argue that paragraph [0022] of Ooyama does not teach the element "polycrystal" (P11/¶5). Paragraph [0022] discloses the chemical formula of the first compound (see complex oxide particles, [0022]). "Products of identical chemical composition can not have mutually exclusive properties." A chemical composition and its properties are inseparable. Therefore, if the prior art teaches the identical chemical structure, the properties applicant discloses and/or claims are necessarily present. *In re Spada*, 911 F.2d 705, 709, 15 USPQ2d 1655, 1658 (Fed. Cir. 1990). As Ooyama discloses the same chemical composition, Ooyama discloses the properties of the chemical composition. Therefore, paragraph [0022] of Ooyama does teach the element "polycrystal."

Applicants argue the rejection of claims 4, 6, 8, and 14-16 should also be withdrawn at least by virtue of their dependence on claim 1 or 13 (P11/¶6). Claims 1 and 13 are not patentable as detailed above.

Applicants argue Yura, Endo, and Takahashi correct the deficiencies of Ooyama (P12/¶6). Ooyama is not deficient as detailed above.

Conclusion

21. The prior art made of record and not relied upon is considered pertinent to applicant's disclosure.

Inagaki (US 2008/0166637 A1) discloses a lithium cobalt oxide coated with a magnesium oxide layer (see positive active material, [0247]).

22. **THIS ACTION IS MADE FINAL.** Applicant is reminded of the extension of time policy as set forth in 37 CFR 1.136(a).

A shortened statutory period for reply to this final action is set to expire THREE MONTHS from the mailing date of this action. In the event a first reply is filed within TWO MONTHS of the mailing date of this final action and the advisory action is not mailed until after the end of the THREE-MONTH shortened statutory period, then the shortened statutory period will expire on the date the advisory action is mailed, and any extension fee pursuant to 37 CFR 1.136(a) will be calculated from the mailing date of the advisory action. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the mailing date of this final action.

23. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Sean P Cullen, Ph.D. whose telephone number is (571)270-1251. The examiner can normally be reached on Monday-Thursday 6:00 am-4:00 pm CDT, Friday 6:00 am-12:00 pm CDT.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Basia A Ridley can be reached on (571)272-1453. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

Notice of References Cited	Application/Control No. 15/638,449	Applicant(s)/Patent Under Reexamination OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	CPC Classification	US Classification
*	A	US-20080166637-A1	07-2008	INAGAKI; Hiroki	H01M4/366	429/329
	B					
	C					
	D					
	E					
	F					
	G					
	H					
	I					
	J					
	K					
	L					
	M					

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	CPC Classification
	N					
	O					
	P					
	Q					
	R					
	S					
	T					

NON-PATENT DOCUMENTS

*		Include as applicable: Author, Title Date, Publisher, Edition or Volume, Pertinent Pages)
	U	
	V	
	W	
	X	

*A copy of this reference is not being furnished with this Office action. (See MPEP § 707.05(a).)
Dates in MM-YYYY format are publication dates. Classifications may be US or foreign.

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

CPC - Searched*		
Symbol	**Date**	**Examiner**
H01M4/131, 1315, 1391, 13915, 625, 8657	SPC	SPC
Updated CPC search	06/27/2019	SPC

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**
429	231.3	03/07/2019	SPC
	Updated USPC search	06/27/2019	SPC

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

Search Notes		
Search Notes	**Date**	**Examiner**
H01M4/131, 1315, 1391, 13915, 625, 8657	SPC	SPC
429/231.3	03/07/2019	SPC
Inventor/applicant search, PALM	03/07/2019	SPC
EAST text search, see search history	03/07/2019	SPC
Updated inventor/applicant search, PALM	06/27/2019	SPC
Updated EAST text search, see search history	06/27/2019	SPC

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

Index of Claims	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

✓	Rejected	-	Cancelled	N	Non-Elected	A	Appeal
=	Allowed	÷	Restricted	I	Interference	O	Objected

CLAIMS

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIM		DATE								
Final	Original	12/20/2018	03/07/2019	06/27/2019						
	1	÷	✓	✓						
	2	÷	✓	✓						
	3	÷	✓	✓						
	4	÷	✓	✓						
	5	÷	✓	✓						
	6	÷	✓	✓						
	7	÷	✓	-						
	8	÷	✓	✓						
	9	÷	✓	✓						
	10	÷	✓	✓						
	11	÷	✓	✓						
	12	÷	✓	✓						
	13	÷	✓	✓						
	14	÷	✓	✓						
	15	÷	✓	✓						
	16	÷	✓	✓						
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	19	÷	-	-						
	20	÷	-	-						
	21	÷	-	-						
	22	÷	-	-						

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
S76	17	(US-6984469-$\|US-5604396-$|US-9478796-$\|JP-2002164053-$|JP-08100107-$\|JP-07316551-$|JP-2015527707-$\|EP-0638625-$|WO-2014015139-$\|WO-2012132387-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/06/27 12:18
S77	2	US-20070224506-$.did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/06/27 13:25
L5	16	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) same (lithium adj titanate "LiTiO.sub.2") same (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:09
L4	14	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) with (lithium adj titanate "LiTiO.sub.2") with (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:09
L3	16	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) same (lithium adj titanate "LiTiO.sub.2") same (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:09
L7	2	l5 not L6	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:10
L6	14	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) with (lithium adj titanate "LiTiO.sub.2") with (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:10
L9	15	l8 not l5	US-PGPUB;	OR	ON	2019/07/08

			USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB			10:11
L8	21	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) and (lithium adj titanate "LiTiO.sub.2") with (shell coating coat coated layer film) and (magnesium adj oxide MgO) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:11
L10	16	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.sub.2" LiCo2 lithium adj cobalt adj oxide) same (lithium adj titanate "LiTiO.sub.2") same (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:12
L13	25	l12 not l5	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:13
L12	31	H01M.cpcl. and (lithium adj cobaltate "LiCoO.sub.2" LiCoO2 lithium adj cobalt adj oxide) and (lithium adj titanate "LiTiO.sub.2") with (shell coating coat coated layer film) and (magnesium adj oxide MgO) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:13
L11	16	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.sub.2" LiCoO2 lithium adj cobalt adj oxide) same (lithium adj titanate "LiTiO.sub.2") same (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:13

EAST Search History (Interference)

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7/ 8/ 2019 10:19:28 AM
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Substitute for form 1449A/PTO				*Complete if Known*	
INFORMATION DISCLOSURE STATEMENT BY APPLICANT *(use as many sheets as necessary)*				Application Number	15/638,449
				Filing Date	June 30, 2017
				First Named Inventor	Teruaki OCHIAI et al.
				Art Unit	1725
				Examiner Name	Sean P. Cullen
Sheet	1	of	1	Attorney Docket Number	740756-004563

U.S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	U.S. Patent Document Number - Kind Code[2] *(if known)*	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] Number[4]	Kind Code[5] *(if known)*	Publication Date MM-DD-YYYY	Name of Patentee or Application of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		WO-2012-132387		10-04-2012			Full

OTHER PRIOR ART – NON PATENT LITERATURE DOCUMENTS

Examiner Initials*	Cite No.[1]	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published.	T[2]

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/27/2019

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

[1] Applicant's unique citation designation number (optional). [2] See Kinds Codes of USPTO Patent Documents at www.uspto.gov or MPEP 901.04. [3] Enter Office that issued the document, by the two-letter code (WIPO Standard ST.3). [4] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [5] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [6] Applicant is to place a check mark here if English language Translation is attached.

[1] Applicant's unique citation designation number (optional). [2] Applicant is to place a check mark here if English language Translation is attached.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /SPC/

OPAP AKIMA-3 JUN 14 2019 PATENT & TRADEMARK OFFICE

Docket No.: 740756-004563

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application	)	
Teruaki **OCHIAI** et al.	)	Group Art Unit: 1725
Application No.: 15/638,449	)	Examiner: Sean P. Cullen
Filed: June 30, 2017	)	Confirmation No.: 1074
For: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE **(As Currently Amended)**	)	

INFORMATION DISCLOSURE STATEMENT

Mail Stop Amendment – Via EFS
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In accordance with the duty of disclosure as set forth in 37 C.F.R. § 1.56, Applicant hereby submits the following information in conformance with 37 C.F.R. §§ 1.97 and 1.98. In accordance with the policies of the U.S. Patent and Trademark Office, only copies of newly cited foreign patents and non-patent literature are being submitted.

Applicant notes that the *Foreign Patent Documents*: JP-2002-164053; JP-08-100107; JP-07-316551 and JP-2015-527707, except the family documents expressly noted below, were previously cited by Japan Patent Office in a Japanese Office Action mailed **April 23, 2019** issued in a counterpart Japanese Application No. 2017-132227.

Applicants also wish to bring to the Examiner's attention the following family information listed on the attached PTO-1449.

1. US-6,984,469 is in the **same family** as JP-2002-164053, but was **not cited** in the above noted foreign communication.

2. US-5,604,396 and EP-0 638 625 are in the **same family** as JP-07-316551, but were **not cited** in the above noted foreign communication.

3. US-9,478,796 and WO-2014-015139 are in the **same family** as JP-2015-527707, but were **not cited** in the above noted foreign communication.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /SPC/

The undersigned certifies that each item of information contained in this information disclosure statement **with the exception of the family document(s) noted above and included on the attached Form PTO-1449** was first cited in any communication from a foreign patent office in connection with a counterpart foreign application not more than three (3) months prior to the filing of this statement.

It is requested that the accompanying Form PTO-1449 be considered and made of record in the above-identified application and that an Examiner-initialed copy of this form be returned to the undersigned.

The present Information Disclosure Statement is submitted before issuance of an action closing prosecution and is certified under 37 C.F.R. § 1.97(e)(1); therefore, no fee is believed to be necessary at this time.

However, if a fee becomes necessary, the Commissioner is hereby authorized to charge fees required in connection with this filing or credit any overpayment to Deposit Account No. 19-2380.

Respectfully submitted,

/Jeffrey L. Costellia, Reg. No. 35,483/
Jeffrey L. Costellia
Registration No. 35,483

NIXON PEABODY LLP
CUSTOMER NO.: 22204
799 9th Street, N.W., Suite 500
Washington, DC 20001-4501
Tel: 202-585-8000

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JUN 14 2019 U.S. PATENT AND TRADEMARK OFFICE

PTO/SB/08A (07-06)
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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 1

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	740756-4563

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-6,984,469	01-10-2006	Kweon.H et al.	
		US-5,604,396	02-18-1997	Watanabe.A et al.	
		US-9,478,796	10-25-2016	Li.L et al.	

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2002-164053A	06-07-2002			Abst.
		JP-08-100107A	04-16-1996			Abst.
		JP-07-316551A	12-05-1995			Abst.
		JP-2015-527707	09-17-2015			Abst.
		EP-0638625A	02-15-1995			Eng.
		WO-2014/015139	01-23-2014			Eng.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/27/2019

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant. 1 Applicant's unique citation designation number (optional). 2 See Kinds Codes of USPTO Patent Documents at www.uspto.gov or MPEP 901.04. 3 Enter Office that issued the document, by the two-letter code (WIPO Standard ST.3). 4 For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 5 Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. 6 Applicant is to place a check mark here if English language Translation is attached.

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 2 hours to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /SPC/

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:	)	
Teruaki **OCHIAI** et al.	)	Group Art Unit: 1725
Application No. 15/638,449	)	Examiner: Sean P. Cullen
Filed: June 30, 2017	)	Confirmation No.: 1074
For POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE	)	

AMENDMENT

Mail Stop Amendment – Via EFS
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the **Final Office Action** mailed **July 12, 2019**, please amend the above-identified application and consider Applicant's remarks as follows.

Amendments to the Drawings begin on **page 2** of this paper and include attached replacement sheets.

Amendments to the Claims begins on **page 3** of this paper.

Remarks begin on **page 7** of this paper.

FIG. 12A

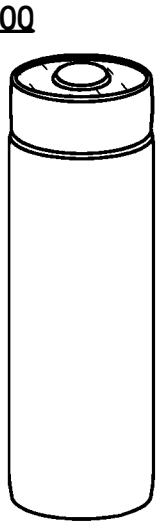


FIG. 12B

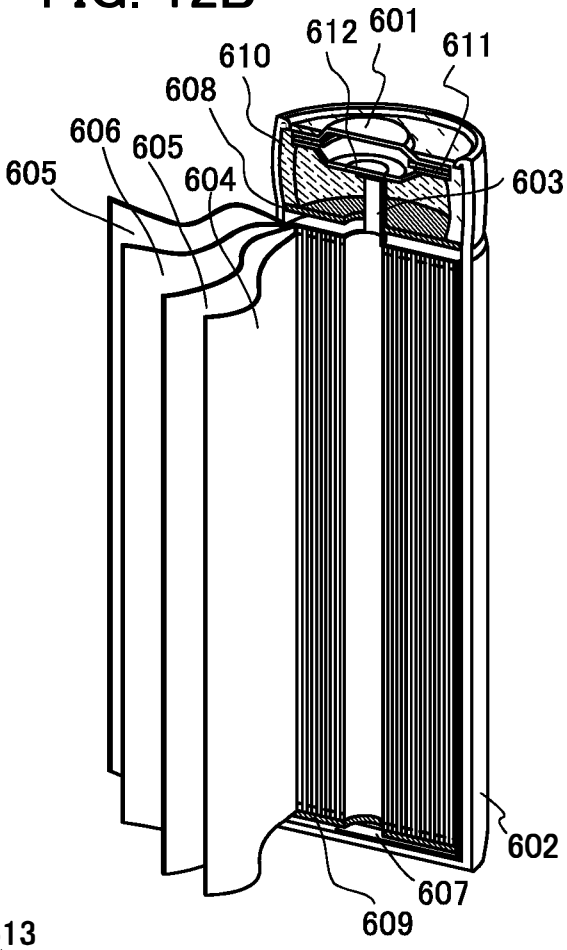


FIG. 12C

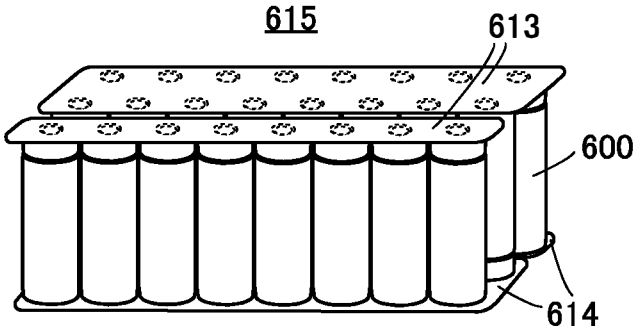


FIG. 12D

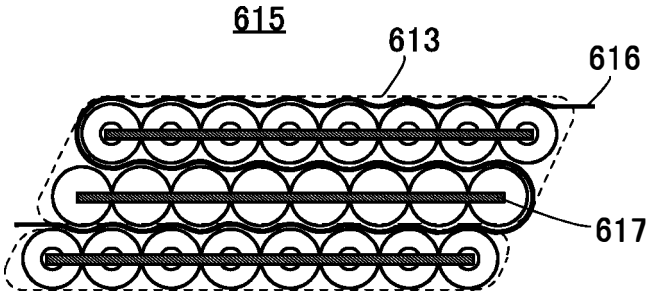


FIG. 22A

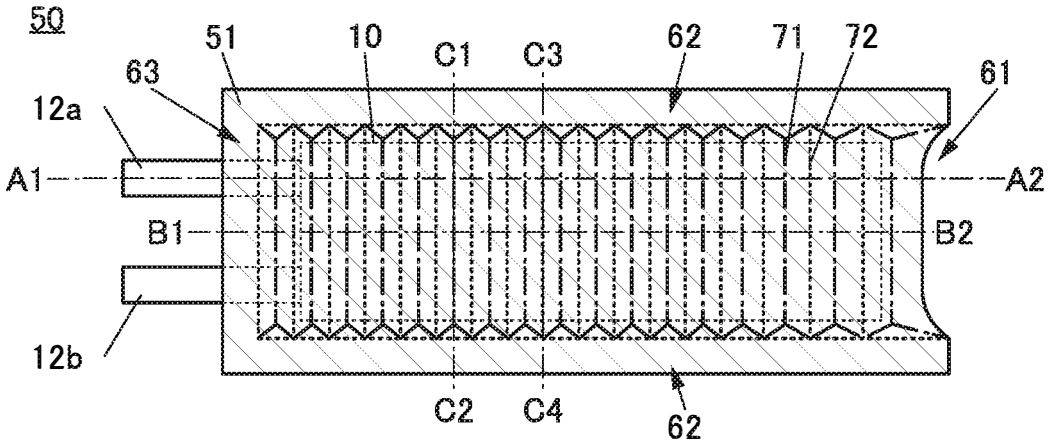


FIG. 22B1

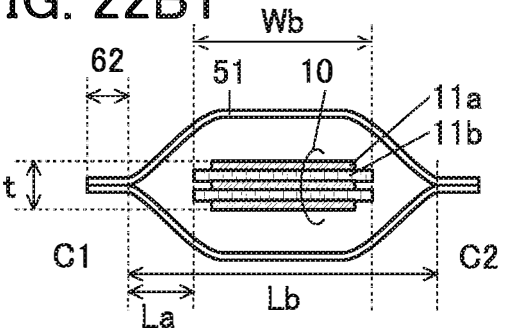


FIG. 22B2

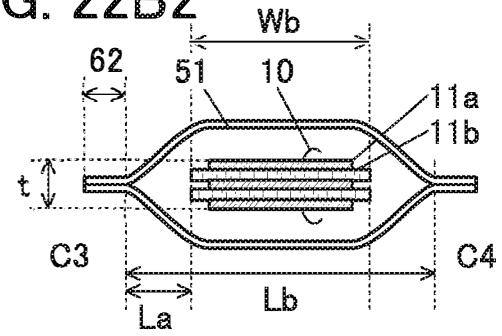


FIG. 22C

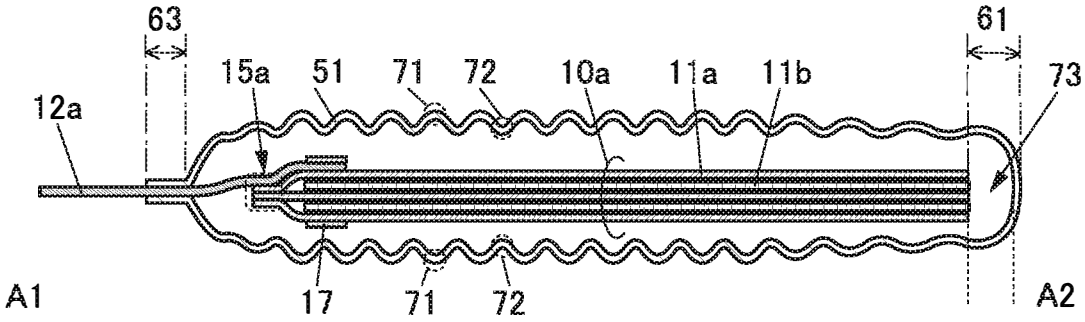


FIG. 22D

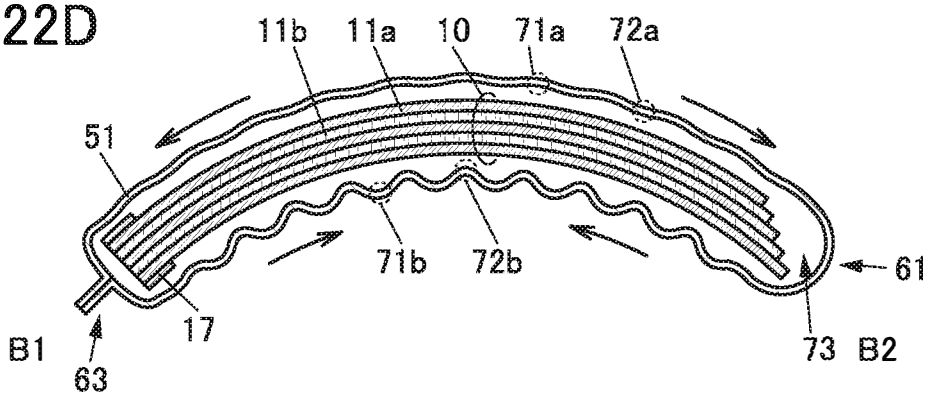


FIG. 23A

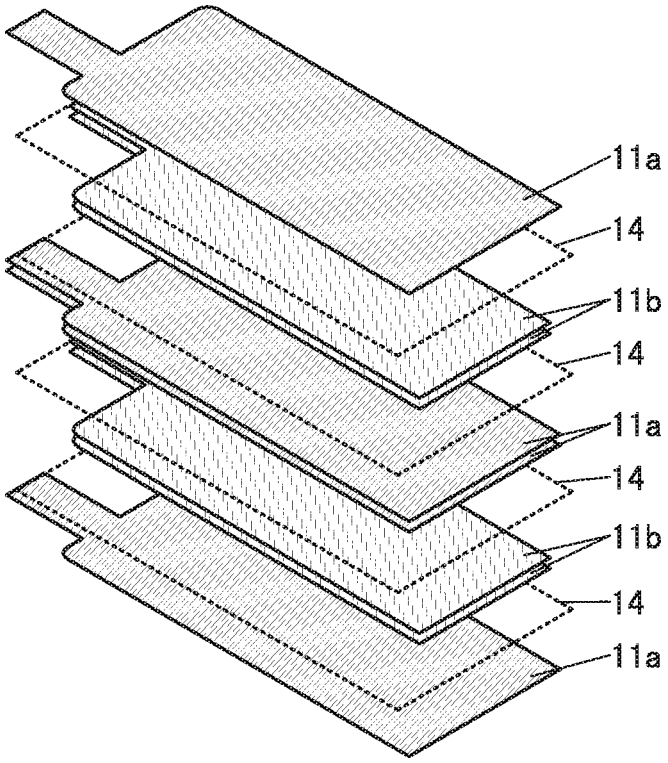


FIG. 23B

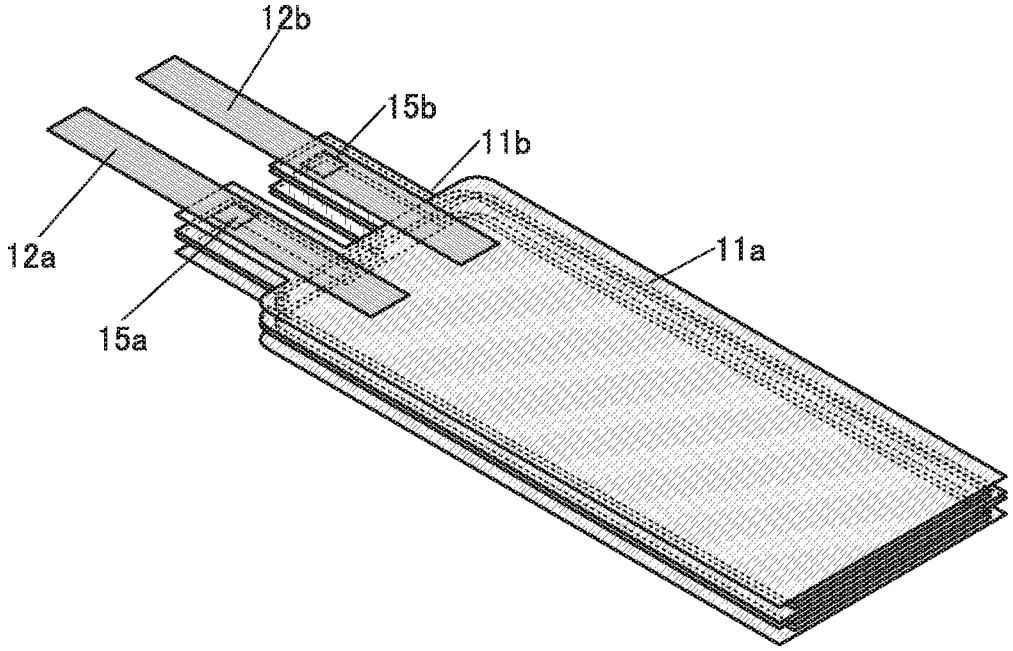


FIG. 25A

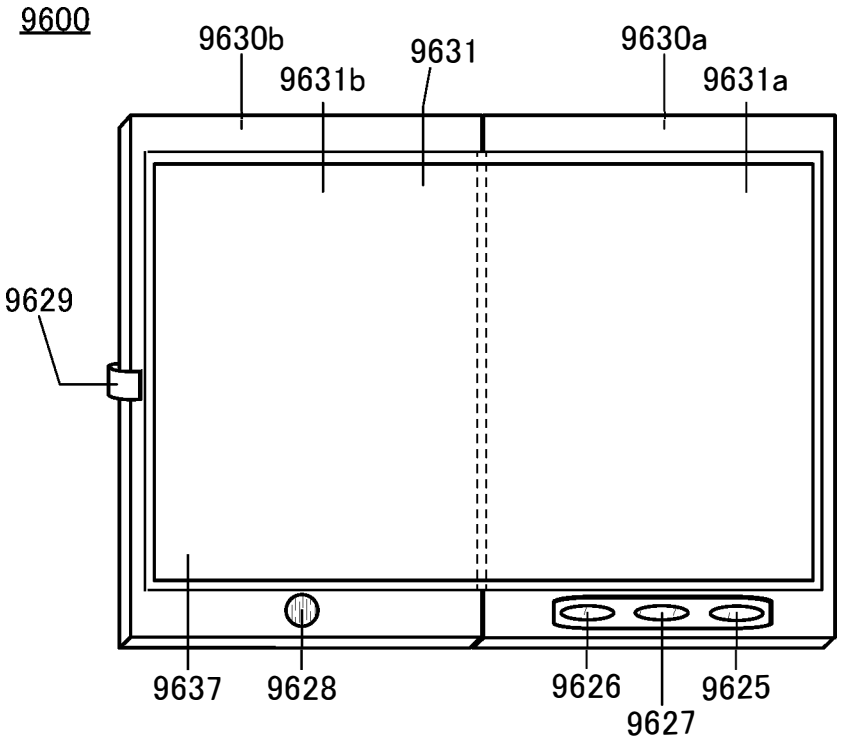


FIG. 25B

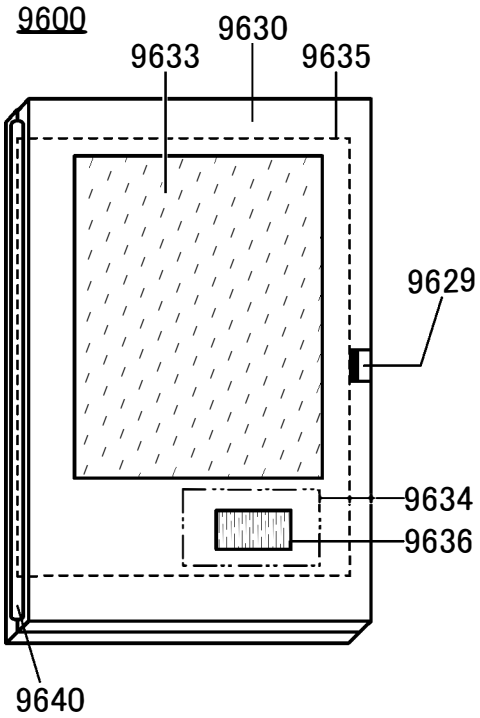


FIG. 25C

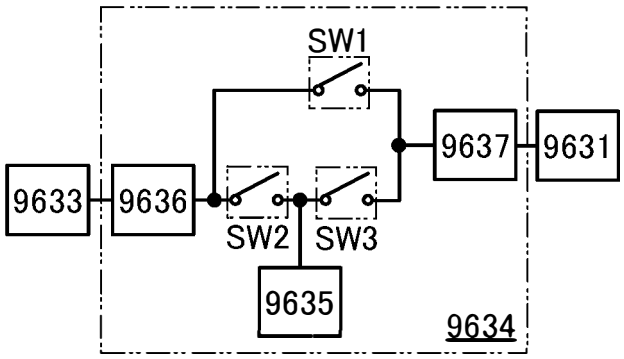


FIG. 27A

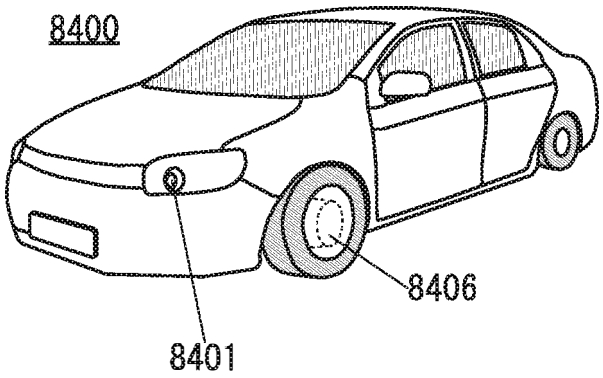


FIG. 27B

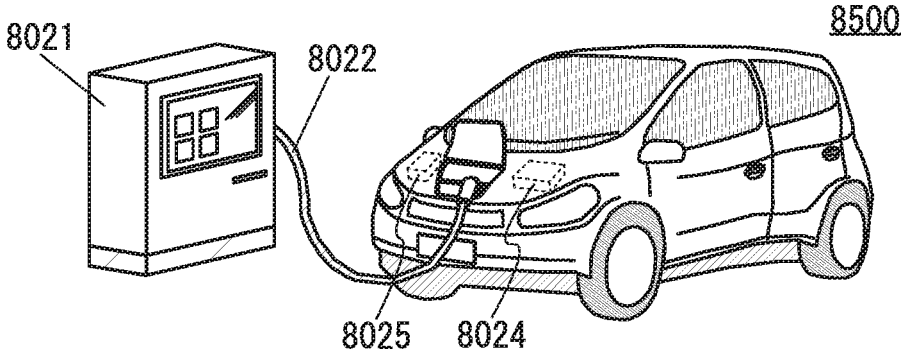
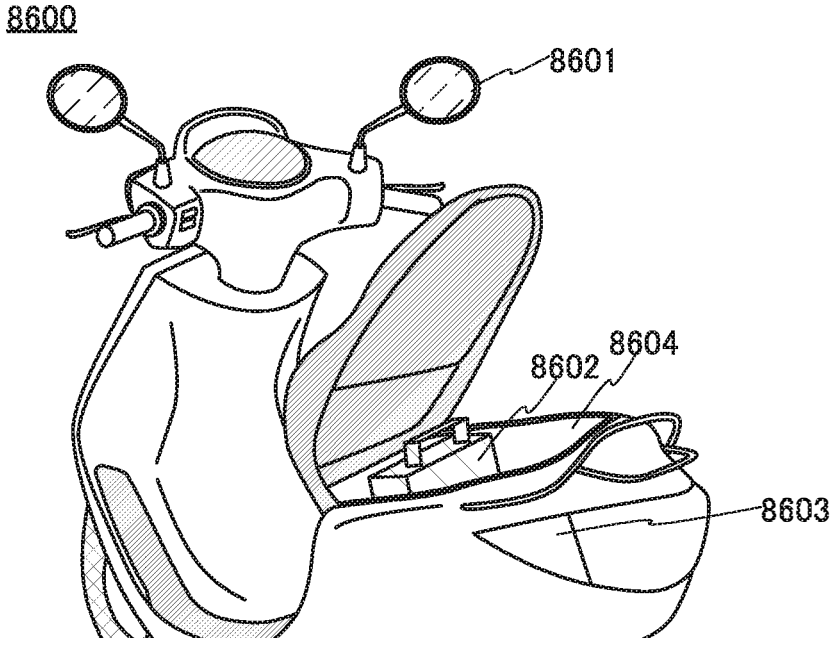


FIG. 27C



before Ti heating (Sample06)

FIG. 37A

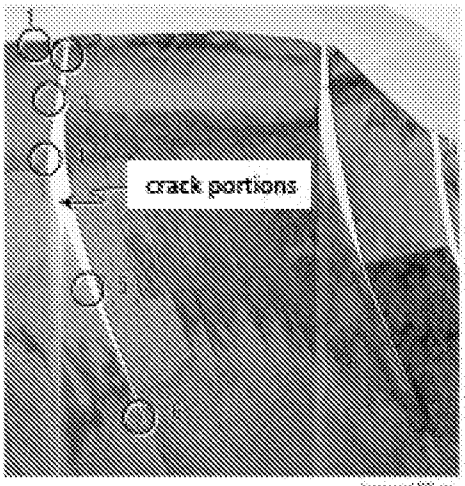
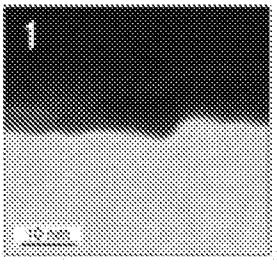


FIG. 37B1



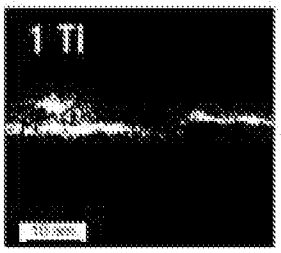
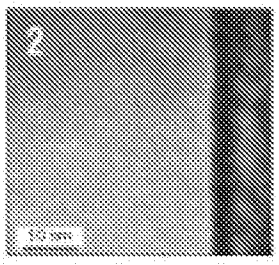


FIG. 37B2

FIG. 37C1



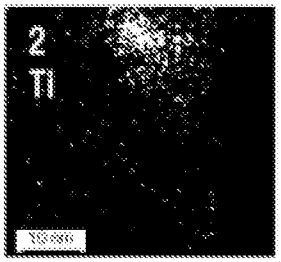
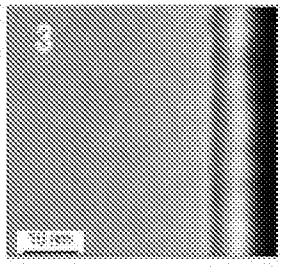


FIG. 37C2

FIG. 37D1



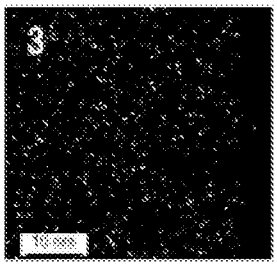
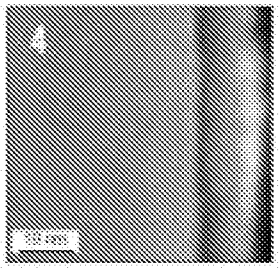


FIG. 37D2

FIG. 37E1



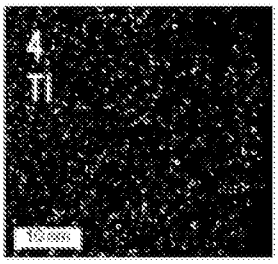


FIG. 37E2

after Ti heating (Sample07)

FIG. 38A

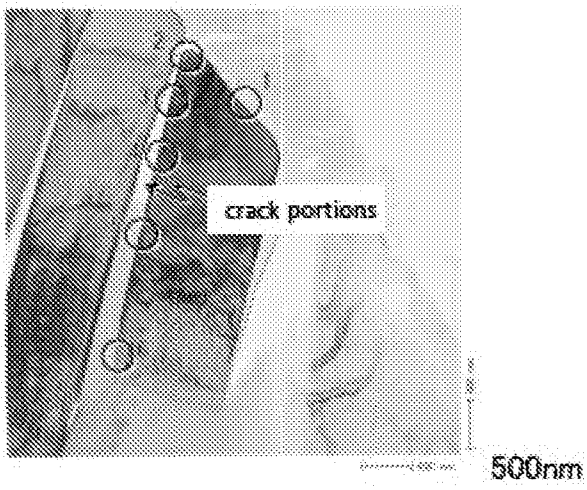
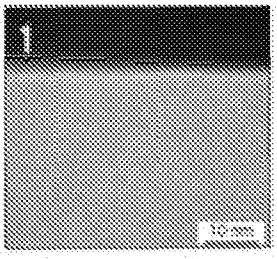


FIG. 38B1



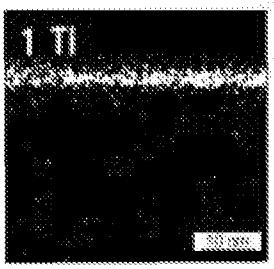
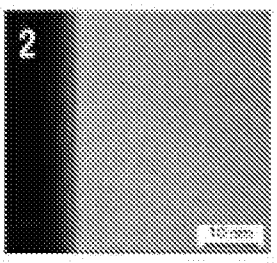


FIG. 38B2

FIG. 38C1



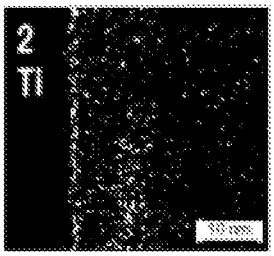
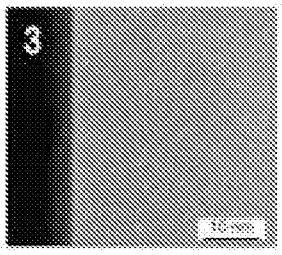


FIG. 38C2

FIG. 38D1



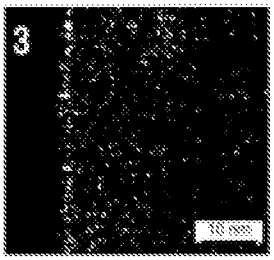
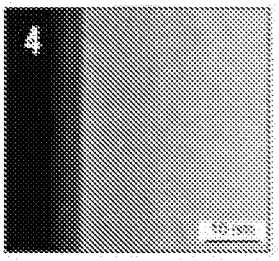


FIG. 38D2

FIG. 38E1



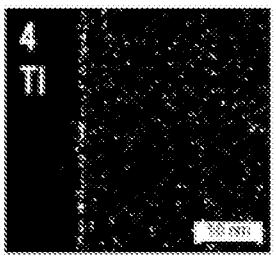


FIG. 38E2

before Mg heating (Sample06)

FIG. 39A

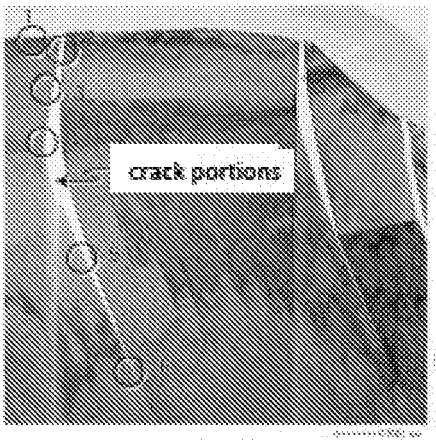
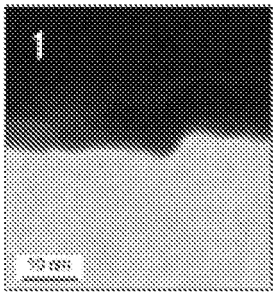


FIG. 39B1



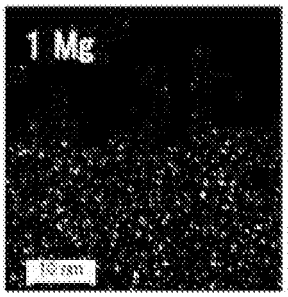
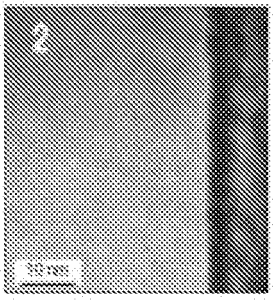


FIG. 39B2

FIG. 39C1



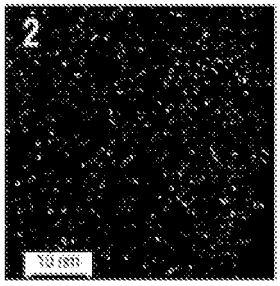
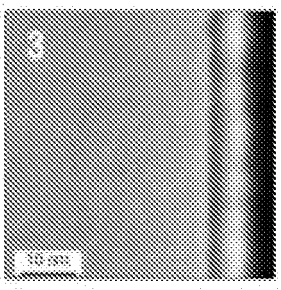


FIG. 39C2

FIG. 39D1



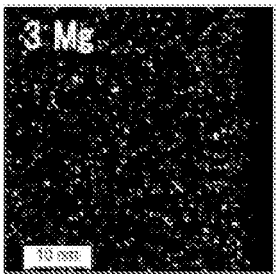
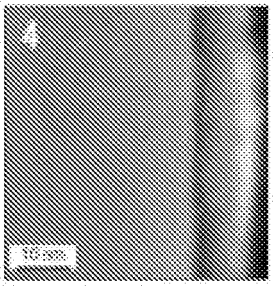


FIG. 39D2

FIG. 39E1



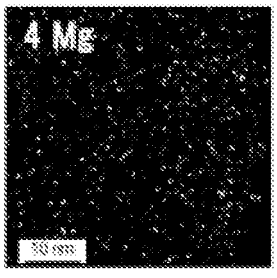


FIG. 39E2

after Mg heating (Sample07)

FIG. 40A

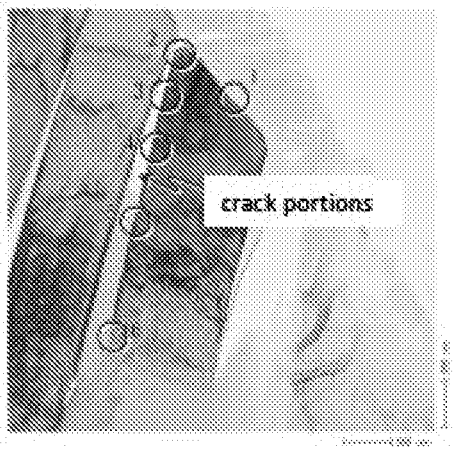


FIG. 40B1

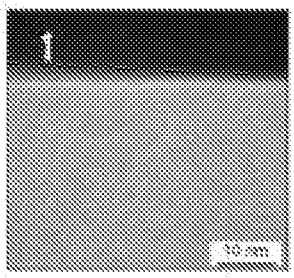
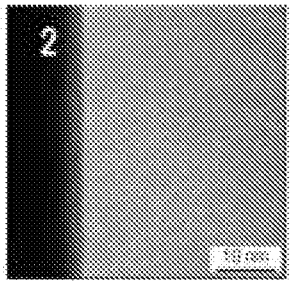




FIG. 40B2

FIG. 40C1



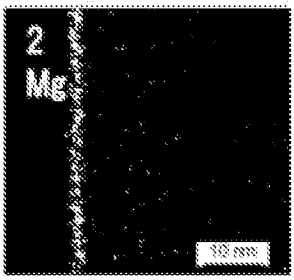
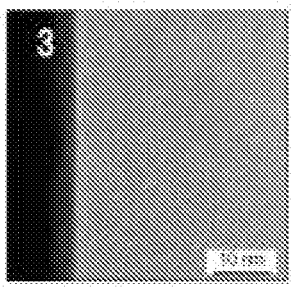


FIG. 40C2

FIG. 40D1



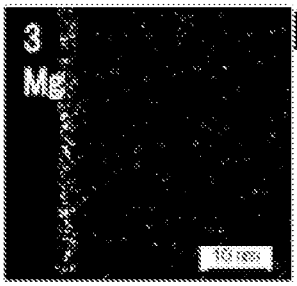
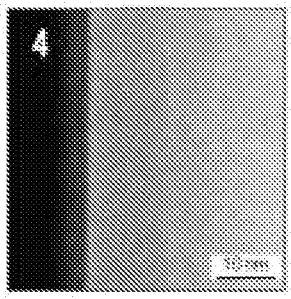


FIG. 40D2

FIG. 40E1



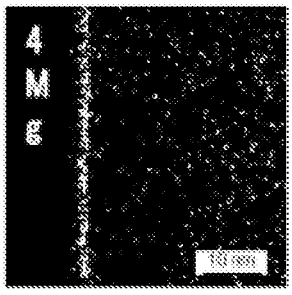
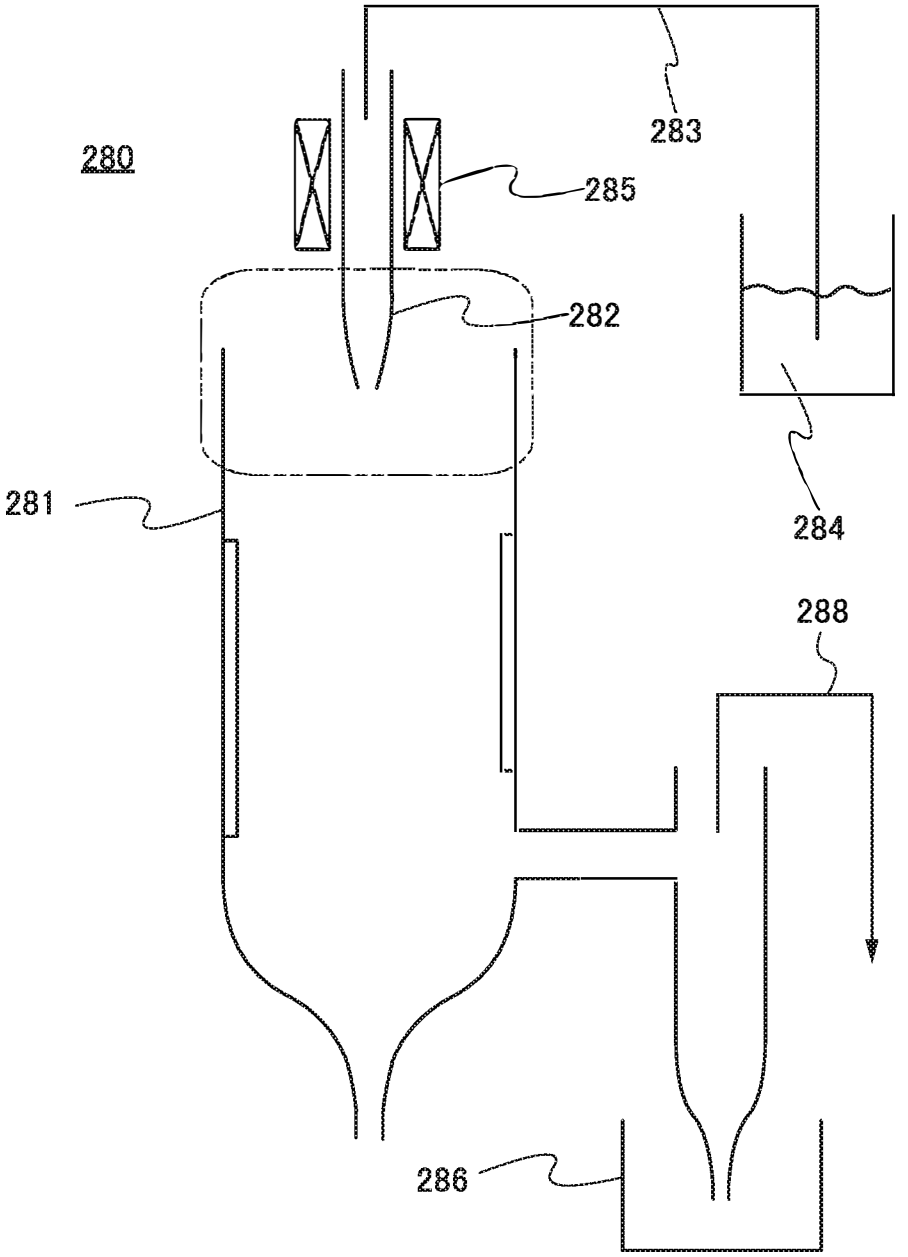


FIG. 40E2

FIG. 54



Electronic Acknowledgement Receipt

EFS ID:	37466573
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	22204
Filer:	Jeffrey Costellia/George Hinton
Filer Authorized By:	Jeffrey Costellia
Attorney Docket Number:	740756-004563
Receipt Date:	15-OCT-2019
Filing Date:	30-JUN-2017
Time Stamp:	22:12:19
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		740756-004563_Amendment.pdf	210972 d579dffdddf95ad4dcf7b86ddc013cf8e8bd0488	yes	10

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Applicant Arguments/Remarks Made in an Amendment	7	10
	Claims	3	6
	Applicant Arguments/Remarks Made in an Amendment	2	2
	Response After Final Action	1	1

Warnings:					
Information:					
2	Drawings-only black and white line drawings	740756-4563_Replacement_Sheets.pdf	1188765 99306ef737d8f832380d8f1a8818d785bc33367d	no	10
Warnings:					
Information:					
		Total Files Size (in bytes):	1399737		

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New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

REMARKS

The Final Office Action mailed **July 12, 2019** was received and its contents carefully noted. This response is filed within three months of the mailing date of the Office Action and therefore is believed to be timely without extension of time. Accordingly, Applicant respectfully submits that this response is being timely filed.

Status of Claims:

Claims 1–6 and 8–16 are pending in the present application, of which claims 1, 9 and 13 are independent. Claim 13 is amended to correct antecedence to a feature.

No new matter is introduced by the present amendment.

For the reasons set forth in detail below, all claims are believed to be in condition for allowance. Favorable reconsideration is requested.

Objection to the Drawings

The drawings are objected to as allegedly failing to comply with 37 CFR 1.84(p)(4) because:

A. *Reference character **8101** has been used to designate both a housing of a lighting device (e.g., Fig. 26) and a headlight (e.g., Fig. 27A). Examiner proposes reference character **8401** should be used to designate a headlight in Fig. 27A.*

B. *Reference character **615** has been used to designate both a module (e.g., Figs. 12C and 12D) and a temperature control device (e.g., Fig. 12D). Examiner proposes reference character **617** should be used to designate a temperature control device in Fig. 12D.*

C. *Reference character **9631a/9631b** are not present in the drawings.*

The drawings are also objected to as failing to comply with 37 CFR 1.84(p)(5) because they do not include the following reference sign(s) mentioned in the description: ***210***, ***211a/211b***, ***212a/212b***, ***215a/215b***, ***217, 250***, ***251***, ***261***, ***262***, ***271***, ***272***, ***271a/271b***, ***272a/272b***, ***273*** *and* ***222.***

Regarding the §1.84(p)(4) objection, Items A, B, and C above, Applicant amends the drawings as proposed by the Examiner and shown in the Replacement Sheets.

Regarding the §1.84(p)(5) objection, Applicant amends the figures as reflected in the Replacement Sheets to address this objection.

In addition, Replacement Figures 37-40 (Sheets 37/58-40/58)are also provided herein since unclear versions of these figures were inadvertently included with the filing of June 14, 2019.

For these reasons Applicant submits the Objection to the drawings should be withdrawn.

Objection to the Specification

The disclosure stands objected to because it includes the following reference characters not described in the written specification and/or drawings *: **617**, **11a**, **11b**, **12a**, **12b**, **14**, **15a**, **15b**, **17**, **50**, **51**, **61**, **62**, **71**, **72**, **73**, **9631b**, **9638**, **9639**, and **8401**.*

In response, Applicant submits that the drawings are amended in this response to address this objection. Accordingly, Applicant requests that this objection be withdrawn.

Claim Rejections – 35 USC § 112

Claims 13–16 stand rejected under AIA 35 U.S.C. 112(b), as allegedly being indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention. Applicant traverses this rejection.

The Examiner on page 8 notes that superficial portion appears twice in claim 13 and that is not clear if both instances refer to the same element. The clarifying amendment shows that the second instance refers to the same element and obviates this rejection.

Applicant respectfully accordingly requests withdrawal of this §112 rejection of independent claim 13 and dependent claims 14–16.

Claim Rejections Under 35 U.S.C. § 102

Claims 1, 4, 6, 8, and 13-16 stand rejected under AIA 35 U.S.C. 102(a)(1) as allegedly being anticipated by Ooyama et al. (US 2007/0224506 Al, hereinafter Ooyama). Applicant traverses this rejection.

Applicant again submits that Ooyama does not disclose each and every element of the base claims and therefore cannot support this § 102 rejection. The cathode active material in Ooyama is not the same as a positive electrode active material of the instant invention. In addition, Ooyama does not teach the type of crystal structure as recited in the claims. The Examiner is referred to the response filed June 14, 2019 ("previous response") and supplements that response with the following.

As noted in the previous response, Ooyama describes "a cathode active material", while the instant claims recite "positive electrode active material", not only in the preamble but throughout the body of the claims. In the Office Action on page 22, the Examiner asserts that "The term "cathode active material" is completely consistent with definition of "positive electrode active material" of the instant specification." Applicant disagrees because paragraph

[0022] of Ooyama recites a "high discharge potential," not "high reaction potential" as recited in the instant application paragraph [0304]. Accordingly, the "cathode active material" of Ooyama is NOT consistent with definition of "positive electrode active material" as instantly claimed.

Ooyama does not teach the crystal structure as instantly claimed. [0022] of Ooyama does not teach "a type of a crystal structure of the first compound" and [0035]/[0036] of Ooyama does not teach "a type of a crystal structure of the second compound." In the referenced paragraphs, Ooyama merely recites "complex oxide particles" with chemical formula I. A person of ordinary skill in the art understands that a material having one chemical formula can have several different structures such as having different crystal structures. For example, silicon, can have polycrystalline structure, single crystal structure, or an amorphous structure. Paragraph [0030] of Ooyama also does not teach "the third region comprises a third compound comprising representative elements at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride" or "a type of a crystal structure of the third compound." Rather, Ooyama only recites in this paragrah "oxide including magnesium", which does not teach a type of a crystal.

For at least these reasons, this §102 rejection of base claims 1 and 13 should be withdrawn. The rejection of claims 4, 6-8, and 14-16 should also be withdrawn at least by virtue of their dependence on claim 1 or 13.

Claim Rejections Under 35 U.S.C. §103

Claims 2 and 3 stand rejected under AIA 35 U.S.C. 103 as allegedly being unpatentable over Ooyama as applied to claim 1 above, and further in view of Yura et al. (US 2012/0258358 Al, hereinafter Yura).

Claim 5 stand rejected under AIA 35 U.S.C. 103 as allegedly being unpatentable over Ooyama as applied to claim 1 above, and further in view of Endo et al. (US 2010/0233542 Al, hereinafter Endo).

Claim 9 stand rejected under AIA 35 U.S.C. 103 as allegedly being unpatentable over Ooyama in view of Takahashi et al. (US 2004/0229123 Al, hereinafter Takahashi).

Claims 10-12 stand rejected under AIA 35 U.S.C. 103 as allegedly being unpatentable over Ooyama in view of Takahashi as applied to claim 9 above, and further in view of Yura.

Applicant traverses these §103 rejections for the following reasons.

Applicant submits that no combination of Yura, Endo, Takahashi and Ooyama renders claims 2, 3, 5, 9–12 obvious. None of these references corrects the deficiencies identified above by supplying or rendering obvious the missing elements and to support *prima facia* obviousness.

For at least these reasons, this rejection §103 rejection of claims 2, 3, 5, 9–12 should be withdrawn.

CONCLUSION

In view of the foregoing, Applicant respectfully requests allowance of the instant application. If a conference would be helpful in expediting prosecution of the instant application, the Examiner is invited to telephone the undersigned to arrange such a conference.

Except for issue fees payable under 37 C.F.R. § 1.18, the Commissioner is hereby authorized by this paper to charge any additional fees during the entire pendency of this application including fees due under 37 C.F.R. §§ 1.16 and 1.17 which may be required, including any required extension of time fees, or credit any overpayment to Deposit Account No. 19-2380.

Respectfully submitted,

/Jeffrey L. Costellia, Reg. No. 35,483/
Jeffrey L. Costellia
Registration No. 35,483

NIXON PEABODY LLP
CUSTOMER NO.: 22204
799 9th Street, N.W., Suite 500
Washington, DC 20001-4501
Tel: 202-585-8000

Amendments to the Claims:

The listing of claims will replace all prior versions, and listings, of claims in the application:

1. (Previously Presented) A positive electrode active material comprising:

a first region in the positive electrode active material;

a second region in the positive electrode active material, the second region surrounding the first region; and

a third region in the positive electrode active material, the third region surrounding the second region,

wherein the first region comprises a first compound comprising an oxide of lithium and a first transition metal,

wherein a type of a crystal structure of the first compound is a layered rock-salt crystal structure,

wherein the second region comprises a second compound comprising an oxide of a second transition metal,

wherein a type of a crystal structure of the second compound is a rock-salt crystal structure,

wherein the second compound is a non-stoichiometric compound,

wherein the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride, and

wherein a type of a crystal structure of the third compound is the rock-salt crystal structure.

2. (Original) The positive electrode active material according to claim 1,

wherein a degree of a mismatch between the crystal structure of the first compound and the crystal structure of the second compound is less than or equal to 0.12, and

wherein a degree of a mismatch between the crystal structure of the second compound and the crystal structure of the third compound is less than or equal to 0.12.

3. (Original) The positive electrode active material according to claim 1,

wherein a degree of a mismatch between a (1-1-4) plane of the crystal structure of the first compound or a plane orthogonal to the (1-1-4) plane and a {100} plane of the crystal structure of the second compound is less than or equal to 0.12, and

wherein a degree of a mismatch between the {100} plane of the crystal structure of the second compound and a {100} plane of the crystal structure of the third compound is less than or equal to 0.12.

4. (Original) The positive electrode active material according to claim 1,

wherein the first compound is a polycrystal.

5. (Original) The positive electrode active material according to claim 4,

wherein an average crystallite size of the oxide of lithium and the first compound is greater than or equal to 280 nm and less than or equal to 630 nm.

6. (Previously Presented) The positive electrode active material according to claim 1,

wherein the third compound essentially consists of the one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride.

7. (Canceled)

8. (Original) A secondary battery comprising:

a positive electrode comprising the positive electrode active material according to claim 1; and

a negative electrode.

9. (Previously Presented) A positive electrode active material comprising:

a first region in the positive electrode active material;

a second region in the positive electrode active material, the second region surrounding the first region; and

a third region in the positive electrode active material, the third region surrounding the second region,

wherein the first region comprises lithium cobaltate,

wherein the second region comprises lithium titanate, and

wherein the third region comprises magnesium oxide.

10. (Original) The positive electrode active material according to claim 9,

wherein a crystal orientation of a part of the lithium cobaltate of the first region and a crystal orientation of a part of the lithium titanate of the second region are substantially aligned with each other.

11. (Original) The positive electrode active material according to claim 9,

wherein a crystal orientation of a part of the lithium titanate of the second region and a crystal orientation of a part of the magnesium oxide of the third region are substantially aligned with each other.

12. (Original) The positive electrode active material according to claim 9,

wherein a degree of a mismatch between a crystal structure of the lithium cobaltate of the first region and a crystal structure of the lithium titanate of the second region is less than or equal to 0.12, and

wherein a degree of a mismatch between the crystal structure of the lithium titanate of the second region and a crystal structure of the magnesium oxide of the third region is less than or equal to 0.12.

13. (Currently Amended) A positive electrode active material particle comprising lithium, titanium, cobalt, magnesium, oxygen, and fluorine,

wherein when a concentration of cobalt in a superficial portion of the positive electrode active material particle is 1, a concentration of titanium in [[a]] the superficial portion of the positive electrode active material particle is greater than or equal to 0.05 and less than or equal to 0.4, a concentration of magnesium in the superficial portion of the positive electrode active material particle is greater than or equal to 0.4 and less than or equal to 1.5, and a concentration of fluorine in the superficial portion of the positive electrode active material particle is greater than or equal to 0.05 and less than or equal to 1.5.

14. (Previously Presented) The positive electrode active material particle according to claim 13, further comprising:

a first region in the positive electrode active material particle; a second region in the positive electrode active material particle, the second region surrounding the first region; and

a third region in the positive electrode active material, the third region surrounding the second region,

wherein the first region comprises a first compound comprising lithium and cobalt,

wherein the second region comprises a second compound comprising titanium,

wherein the second compound is a non-stoichiometric compound, and

wherein the third region comprises a third compound comprising magnesium and fluorine.

15. (Original) The positive electrode active material particle according to claim 14,

wherein a type of a crystal structure of the first compound is a layered rock-salt crystal structure,

wherein a type of a crystal structure of the second compound is a rock-salt crystal structure, and

wherein a type of a crystal structure of the third compound is the rock-salt crystal structure.

16. (Original) A secondary battery comprising:

a positive electrode comprising the positive electrode active material particle according to claim 13; and

a negative electrode.

17-22. (Canceled)

Amendments to the Drawings:

The attached sheets of drawings include changes to Figures 12A-12D, 22A-22D, 23A-23B, 25C, 27A-27C and 54. These sheets, which include Figures 12A-12D, 22A-22D, 23A-23B, 25C, 27A-27C and 54, replace the original sheets including Figures 12A-12D, 22A-22D, 23A-23B, 25C, 27A-27C and 54.

No new matter is introduced by the present amendment.

Attachment: Replacement Sheets

PTO/SB/06 (09-11)
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PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 15/638,449	Filing Date 06/30/2017	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $80 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $420 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	10/15/2019	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 15	Minus	** 22	= 0		x $100 =	0
	Independent (37 CFR 1.16(h))	* 3	Minus	*** 4	= 0		x $460 =	0
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	0

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	LIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/VERONICA EVERETTE/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	740756-004563	1074

22204 7590 10/23/2019

NIXON PEABODY, LLP
799 Ninth Street, NW
SUITE 500
WASHINGTON, DC 20001

EXAMINER	
CULLEN, SEAN P	

ART UNIT	PAPER NUMBER
1725	

NOTIFICATION DATE	DELIVERY MODE
10/23/2019	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

ipairlink@nixonpeabody.com
nppatent@nixonpeabody.com

DETAILED ACTION

Drawings

1. The drawings were received on 15 October 2019. These drawings are acceptable.

2. Applicants' amendment(s) have overcome the objection(s) to the drawing(s).

Specification

3. Applicants' amendment(s) have overcome the objection(s) to the specification.

Claim Rejections - 35 USC § 112

4. Applicants' amendment(s) have overcome the rejection(s) of **claim(s) 13-16** under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph.

Response to Arguments

5. Applicant's arguments filed 15 October 2019 have been fully considered but they are not persuasive.

Applicants argue the cathode active material of Ooyama is not consistent with the definition of "positive electrode active material" as instantly claimed (P9/¶1). The "negative electrode" described in the instant application (e.g., [0313]-[0325]) is consistent with the anode of Ooyama (e.g., [0048]-[0055]). The "positive electrode" described in the instant application (e.g., [0289]-[0311]) is consistent with the cathode of Ooyama (e.g., [0022]-[0047]). Therefore, the cathode active material of Ooyama is consistent with the definition of "positive electrode active material" as instantly claimed.

Applicants argue Ooyama does not teach a type of a crystal structure of the first compound and a type of a crystal structure of the second compound (P9/¶2). Paragraph [0022] discloses the chemical formula of the first compound (see complex oxide particles, [0022]). "Products of identical chemical composition can not have mutually exclusive properties." A chemical composition and its properties are inseparable. Therefore, if the prior art teaches the identical chemical structure, the properties applicant discloses and/or claims are necessarily present. *In re Spada*, 911 F.2d 705, 709, 15 USPQ2d 1655, 1658 (Fed. Cir. 1990). As Ooyama discloses the same chemical composition, Ooyama discloses the properties of the chemical composition. Paragraph [0035] and [0036] discloses the chemical compounds of the second compound. " The instant application describes the second compound with a rock-salt crystal structure is one of titanium oxide, vanadium oxide, manganese oxide, iron oxide, copper oxide, chromium oxide, niobium oxide, cobalt oxide, zinc oxide, and the like. Ooyama describes the second compound can be manganese oxide. As Ooyama discloses the same chemical composition, Ooyama discloses the properties of the chemical composition. Therefore, Ooyama does teach a type of a crystal structure of the first compound and a type of a crystal structure of the second compound.

Applicants argue a material having one chemical formula can have several different structures (P9/¶2). Ooyama discloses specific chemical compositions that are identical to the claimed chemical composition. "Products of identical chemical composition can not have mutually exclusive properties." A chemical composition and its properties are inseparable. Therefore, if the prior art teaches the identical chemical structure, the properties applicant discloses and/or claims are necessarily present. *In re Spada*, 911 F.2d 705, 709, 15 USPQ2d

1655, 1658 (Fed. Cir. 1990). Therefore, the specific chemical composition of Ooyama does not have several different structures.

Applicants argue silicon can have polycrystalline structure, single crystal structure, or an amorphous structure (P9/¶2). The claimed material is drawn to metal oxides and/or metal fluorides. The material of Ooyama is drawn to metal oxides. The property of silicon, which is not a metal oxide, is not applicable to the properties of metal oxides. Therefore, the applicant has not demonstrated that metal oxides can have polycrystalline structure, single crystal structure, or an amorphous structure.

Applicants argue that Ooyama does not teach "the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride" or "a type of a crystal structure of the third compound" (P9/¶2). Paragraph [0030] describes a third region including an oxide of magnesium (see surface layer, [0030]). An oxide of magnesium is a magnesium oxide. Ooyama also explicitly discloses MgO ([0083], [0085], and Table 1), which is magnesium oxide. Paragraph [0030] describes an oxide of magnesium (see surface layer, [0030]). An oxide of magnesium is a magnesium oxide. Ooyama also explicitly discloses MgO ([0083], [0085], and Table 1), which is magnesium oxide. "Products of identical chemical composition can not have mutually exclusive properties." A chemical composition and its properties are inseparable. Therefore, if the prior art teaches the identical chemical structure, the properties applicant discloses and/or claims are necessarily present. *In re Spada*, 911 F.2d 705, 709, 15 USPQ2d 1655, 1658 (Fed. Cir. 1990). As Ooyama discloses the same chemical composition, Ooyama discloses the properties of the chemical composition. Therefore, Ooyama does teach "the third region comprises a third compound

comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride" and "a type of a crystal structure of the third compound."

Applicants argue that the rejection of claim 13 should be withdrawn based on the above reasons (P9/¶3). Claim 13 does not recite a type of a crystal structure of the first compound, a type of a crystal structure of the second compound, the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride, or a type of a crystal structure of the third compound. As such, the arguments with respect to claim 1 are not applicable to claim 13. Therefore, the rejection of claim 13 should not be withdrawn.

Applicants argue the rejection of claims 4, 6-8, and 14-16 should be withdrawn by virtue of their dependence on claims 1 or 13 (P9/¶3). Claim 1 and 13 are not patentable as detailed above.

Applicants argue Yura, Endo, Takahashi do not correct the deficiencies of Ooyama (P10/¶1). Ooyama is not deficient as detailed above. The Office further notes that independent claim 9 does not recite a type of a crystal structure of the first compound, a type of a crystal structure of the second compound, the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride, or a type of a crystal structure of the third compound. As such, the arguments with respect to claim 1 are not applicable to claim 9.

Conclusion

6. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Sean P Cullen, Ph.D. whose telephone number is (571)270-1251.

The examiner can normally be reached on Monday-Thursday 6:00 am-4:00 pm CDT, Friday 6:00 am-12:00 pm CDT.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Basia A Ridley can be reached on (571)272-1453. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

Advisory Action Before the Filing of an Appeal Brief	**Application No.** 15/638,449	**Applicant(s)** OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	**Art Unit** 1725	**AIA (FITF) Status** Yes

--The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

THE REPLY FILED 15 October 2019 FAILS TO PLACE THIS APPLICATION IN CONDITION FOR ALLOWANCE.

NO NOTICE OF APPEAL FILED

1. ☑ The reply was filed after a final rejection. No Notice of Appeal has been filed. To avoid abandonment of this application, applicant must timely file one of the following replies: (1) an amendment, affidavit, or other evidence, which places the application in condition for allowance; (2) a Notice of Appeal (with appeal fee) in compliance with 37 CFR 41.31; or (3) a Request for Continued Examination (RCE) in compliance with 37 CFR 1.114 if this is a utility or plant application. Note that RCEs are not permitted in design applications. The reply must be filed within one of the following time periods:
   a) ☑ The period for reply expires 3 months from the mailing date of the final rejection.
   b) ☐ The period for reply expires on: (1) the mailing date of this Advisory Action; or (2) the date set forth in the final rejection, whichever is later. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the mailing date of the final rejection.
   c) ☐ A prior Advisory Action was mailed more than 3 months after the mailing date of the final rejection in response to a first after-final reply filed within 2 months of the mailing date of the final rejection.The current period for reply expires ____ months from the mailing date of *the prior Advisory Action* or SIX MONTHS from the mailing date of the final rejection, whichever is earlier.
   Examiner Note: If box 1 is checked, check either box (a), (b) or (c). ONLY CHECK BOX (b) WHEN THIS ADVISORY ACTION IS THE FIRST RESPONSE TO APPLICANTS FIRST AFTER-FINAL REPLY WHICH WAS FILED WITHIN TWO MONTHS OF THE FINAL REJECTION. ONLY CHECK BOX (c) IN THE LIMITED SITUATION SET FORTH UNDER BOX (c). See MPEP 706.07(f).

Extensions of time may be obtained under 37 CFR 1.136(a). The date on which the petition under 37 CFR 1.136(a) and the appropriate extension fee have been filed is the date for purposes of determining the period of extension and the corresponding amount of the fee. The appropriate extension fee under 37 CFR 1.17(a) is calculated from: (1) the expiration date of the shortened statutory period for reply originally set in the final Office action; or (2) as set forth in (b) or (c) above, if checked. Any reply received by the Office later than three months after the mailing date of the final rejection, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

NOTICE OF APPEAL

2. ☐ The Notice of Appeal was filed on _____. A brief in compliance with 37 CFR 41.37 must be filed within two months of the date of filing the Notice of Appeal (37 CFR 41.37(a)), or any extension thereof (37 CFR 41.37(e)), to avoid dismissal of the appeal. Since a Notice of Appeal has been filed, any reply must be filed within the time period set forth in 37CFR 41.37(a).

AMENDMENTS

3. ☐ The proposed amendments filed after a final rejection, but prior to the date of filing a brief, will not be entered because
   a) ☐ They raise new issues that would require further consideration and/or search (see NOTE below);
   b) ☐ They raise the issue of new matter (see NOTE below);
   c) ☐ They are not deemed to place the application in better form for appeal by materially reducing or simplifying the issues for appeal; and/or
   d) ☐ They present additional claims without canceling a corresponding number of finally rejected claims.
   NOTE: _____ (See 37CFR 1.116 and 41.33(a)).
4. ☐ The amendments are not in compliance with 37CFR 1.121. See attached Notice of Non-Compliant Amendment (PTOL-324).
5. ☑ Applicants reply has overcome the following rejection(s): See attached Detailed Action.
6. ☐ Newly proposed or amended claim(s) _____ would be allowable if submitted in a separate, timely filed amendment canceling the non-allowable claim(s).
7. ☑ For purposes of appeal, the proposed amendment(s):(a) ☐ will not be entered, or (b) ☑ will be entered, and an explanation of how the new or amended claims would be rejected is provided below or appended.

AFFIDAVIT OR OTHER EVIDENCE

8. ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____
9. ☐ The affidavit or other evidence filed after final action, but before or on the date of filing a Notice of Appeal will not be entered because applicant failed to provide a showing of good and sufficient reasons why the affidavit or other evidence is necessary and was not earlier presented. See 37 CFR 1.116(e).
10. ☐ The affidavit or other evidence filed after the date of filing the Notice of Appeal, but prior to the date of filing a brief, will not be entered because the affidavit or other evidence failed to overcome all rejections under appeal and/or appellant fails to provide a showing of good and sufficient reasons why it is necessary and was not earlier presented. See 37 CFR 41.33(d)(1).
11. ☐ The affidavit or other evidence is entered. An explanation of the status of the claims after entry is below or attached.

REQUEST FOR RECONSIDERATION/OTHER

12. ☑ The request for reconsideration has been considered but does NOT place the application in condition for allowance because: see attached Detailed Action.
13. ☐ Note the attached Information *Disclosure Statement*(s). (PTO/SB/08) Paper No(s). _____
14. ☐ Other: _____

STATUS OF CLAIMS

15. The status of the claim(s) is (or will be) as follows:
    Claim(s) allowed:_____.
    Claim(s) objected to:_____.
    Claim(s) rejected:1-6 and 8-16.
    Claim(s) withdrawn from consideration:_____.

/Sean P Cullen, Ph.D./ Primary Examiner, Art Unit 1725	

OK TO ENTER: /SPC/

10/17/2019

Docket No: 740756-004563

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:	)	
Teruaki **OCHIAI** et al.	)	Group Art Unit: 1725
Application No. 15/638,449	)	Examiner: Sean P. Cullen
Filed: June 30, 2017	)	Confirmation No.: 1074
For POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE	)	

AMENDMENT

Mail Stop Amendment – Via EFS
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the **Final Office Action** mailed **July 12, 2019**, please amend the above-identified application and consider Applicant's remarks as follows.

Amendments to the Drawings begin on **page 2** of this paper and include attached replacement sheets.

Amendments to the Claims begins on **page 3** of this paper.

Remarks begin on **page 7** of this paper.

PTO/SB/30EFS (02-18)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

REQUEST FOR CONTINUED EXAMINATION(RCE)TRANSMITTAL
(Submitted Only via EFS-Web)

Application Number	15/638,449	Filing Date	2017-06-30	Docket Number (if applicable)	740756-004563	Art Unit	1725
First Named Inventor	Teruaki OCHIAI et al.			Examiner Name	Sean P Cullen		

This is a Request for Continued Examination (RCE) under 37 CFR 1.114 of the above-identified application.
Request for Continued Examination (RCE) practice under 37 CFR 1.114 does not apply to any utility or plant application filed prior to June 8, 1995, or to any design application. The Instruction Sheet for this form is located at WWW.USPTO.GOV

SUBMISSION REQUIRED UNDER 37 CFR 1.114

Note: If the RCE is proper, any previously filed unentered amendments and amendments enclosed with the RCE will be entered in the order in which they were filed unless applicant instructs otherwise. If applicant does not wish to have any previously filed unentered amendment(s) entered, applicant must request non-entry of such amendment(s).

☐ Previously submitted. If a final Office action is outstanding, any amendments filed after the final Office action may be considered as a submission even if this box is not checked.

- ☐ Consider the arguments in the Appeal Brief or Reply Brief previously filed on _____
- ☐ Other _____

☒ Enclosed

- ☒ Amendment/Reply
- ☐ Information Disclosure Statement (IDS)
- ☐ Affidavit(s)/ Declaration(s)
- ☐ Other _____

MISCELLANEOUS

☐ Suspension of action on the above-identified application is requested under 37 CFR 1.103(c) for a period of months _____
(Period of suspension shall not exceed 3 months; Fee under 37 CFR 1.17(i) required)

☐ Other _____

FEES

The RCE fee under 37 CFR 1.17(e) is required by 37 CFR 1.114 when the RCE is filed.

☒ The Director is hereby authorized to charge any underpayment of fees, or credit any overpayments, to Deposit Account No 192380

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT REQUIRED

☒ Patent Practitioner Signature

☐ Applicant Signature

PTO/SB/30EFS (02-18)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Signature of Registered U.S. Patent Practitioner			
Signature	/Jeffrey L. Costellia, Reg. No. 35483/	Date (YYYY-MM-DD)	2019-11-12
Name	Jeffrey L. Costellia	Registration Number	35483

This collection of information is required by 37 CFR 1.114. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450.

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these records.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:	)	
Teruaki **OCHIAI** et al.	)	Group Art Unit: 1725
Application No.: 15/638,449	)	Examiner: Sean P Cullen
Filed: June 30, 2017	)	Confirmation No.: 1074
For: POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY	)	

AMENDMENT UNDER 37 C.F.R. § 1.114

Mail Stop RCE - Via EFS
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the **Final Office Action** dated **July 12, 2019** and the **Advisory Action** dated **October 23, 2019**, please consider the following amendments and remarks in connection with the above-identified application.

A Request for Continued Examination (RCE) accompanies this reply. Any fees required for the RCE are hereby authorized to be charged to Deposit Account No. 19-2380.

Amendments to the Claims are begins on **page 2** of this paper.

Remarks begin on **page 7** of this paper.

Amendments to the Claims:

The listing of claims will replace all prior versions, and listings, of claims in the application:

1. (Currently Amended) A positive electrode active material comprising:

a first region in the positive electrode active material;

a second region in the positive electrode active material, the second region surrounding the first region; and

a third region in the positive electrode active material, the third region surrounding the second region,

wherein the first region comprises a first compound comprising an oxide of lithium and a first transition metal,

wherein a type of a crystal structure of the first compound is a layered rock-salt crystal structure,

wherein the second region comprises a second compound comprising an oxide of a second transition metal,

wherein a type of a crystal structure of the second compound is a rock-salt crystal structure,

wherein the second compound is a non-stoichiometric compound,

wherein the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride, ~~and~~

wherein a type of a crystal structure of the third compound is the rock-salt crystal structure,

wherein a degree of a mismatch between the crystal structure of the first compound and the crystal structure of the second compound is less than or equal to 0.12, and

wherein a degree of a mismatch between the crystal structure of the second compound and the crystal structure of the third compound is less than or equal to 0.12.

2. (Canceled)

3. (Original) The positive electrode active material according to claim 1,

wherein a degree of a mismatch between a (1-1-4) plane of the crystal structure of the first compound or a plane orthogonal to the (1-1-4) plane and a {100} plane of the crystal structure of the second compound is less than or equal to 0.12, and

wherein a degree of a mismatch between the {100} plane of the crystal structure of the second compound and a {100} plane of the crystal structure of the third compound is less than or equal to 0.12.

4. (Original) The positive electrode active material according to claim 1,

wherein the first compound is a polycrystal.

5. (Original) The positive electrode active material according to claim 4,

wherein an average crystallite size of the oxide of lithium and the first compound is greater than or equal to 280 nm and less than or equal to 630 nm.

6. (Previously Presented) The positive electrode active material according to claim 1,

wherein the third compound essentially consists of the one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride.

7. (Canceled)

8. (Original) A secondary battery comprising:

a positive electrode comprising the positive electrode active material according to claim 1; and

a negative electrode.

9. (Previously Presented) A positive electrode active material comprising:

a first region in the positive electrode active material;

a second region in the positive electrode active material, the second region surrounding the first region; and

a third region in the positive electrode active material, the third region surrounding the second region,

wherein the first region comprises lithium cobaltate,

wherein the second region comprises lithium titanate, ~~and~~

wherein the third region comprises magnesium oxide.

wherein a degree of a mismatch between a crystal structure of the lithium cobaltate of the first region and a crystal structure of the lithium titanate of the second region is less than or equal to 0.12, and

wherein a degree of a mismatch between the crystal structure of the lithium titanate of the second region and a crystal structure of the magnesium oxide of the third region is less than or equal to 0.12.

10. (Original) The positive electrode active material according to claim 9,

wherein a crystal orientation of a part of the lithium cobaltate of the first region and a crystal orientation of a part of the lithium titanate of the second region are substantially aligned with each other.

11. (Original) The positive electrode active material according to claim 9,

wherein a crystal orientation of a part of the lithium titanate of the second region and a crystal orientation of a part of the magnesium oxide of the third region are substantially aligned with each other.

12-22. (Canceled)

23. (New) A positive electrode active material comprising:

a first region in the positive electrode active material;

a second region closer to a surface of the positive electrode active material than the first region; and

a third region closer to the surface of the positive electrode active material than the second region,

wherein the first region includes a first oxide of lithium and a first transition metal,

wherein the second region includes a second oxide of lithium and a second transition metal,

wherein the third region includes representative elements and oxygen,

wherein a second distance between the lithium and the second transition metal through oxygen in the second region is longer than a first distance between the lithium and the first transition metal through oxygen in the first region, and

wherein a third distance between the representative elements through the oxygen in the third region is longer than the second distance.

24. (New) The positive electrode active material according to claim 23,

wherein a degree of a mismatch f between the first distance and the second distance is less than or equal to 0.12.

25. (New) The positive electrode active material according to claim 23,

wherein a degree of a mismatch f between the second distance and the third distance is less than or equal to 0.12.

26. (New) The positive electrode active material according to claim 23,

wherein each of the first distance, the second distance, and the third distance is measured by STEM-FFT.

27. (New) The positive electrode active material according to claim 23,

wherein the second region include the first transition metal.

28. (New) The positive electrode active material according to claim 23,

wherein the third region include the first transition metal and the second transition metal.

29. (New) A positive electrode active material comprising:

a first region in the positive electrode active material;

a second region closer to a surface of the positive electrode active material than the first region; and

a third region closer to the surface of the positive electrode active material than the second region,

wherein the first region includes a first oxide of lithium and a first transition metal, a type of a crystal structure of the first oxide being a layered rock-salt crystal structure,

wherein the second region includes a non-stoichiometric compound including a second transition metal, a type of a crystal structure of the non-stoichiometric compound being a rock-salt crystal structure, and

wherein the third region includes a compound including a representative element, a type of a crystal structure of the compound being a rock-salt crystal structure.

30. (New) The positive electrode active material according to claim 29,

wherein the second region include the first transition metal.

31. (New) The positive electrode active material according to claim 29,

wherein the third region include the first transition metal and the second transition metal.

32. (New) A positive electrode active material comprising:

a first region in the positive electrode active material;

a second region closer to a surface of the positive electrode active material than the first region; and

a third region closer to the surface of the positive electrode active material than the second region,

wherein the first region includes a first oxide including lithium and at least one of cobalt, manganese, and nickel, a type of a crystal structure of the first oxide being a layered rock-salt crystal structure,

wherein the second region includes a second oxide including at least one of titanium, vanadium, manganese, iron, chromium, niobium, cobalt, zinc, zirconium, and nickel, a type of a crystal structure of the second oxide being a rock-salt crystal structure, and

wherein the third region includes a compound including a representative element, a type of a crystal structure of the compound being a rock-salt crystal structure.

33. (New) The positive electrode active material according to claim 32,

wherein the second region includes at least one of cobalt, manganese, and nickel.

34. (New) The positive electrode active material according to claim 32,

wherein the third region includes the one of cobalt, manganese, and nickel, and

wherein the third region includes the one of titanium, vanadium, manganese, iron, chromium, niobium, cobalt, zinc, zirconium, and nickel.

REMARKS

The Final Office Action, herein "the Office Action." mailed July 12, 2019 and Advisory Action, herein "the Advisory Action," mailed October 23, 2019 were received and their contents carefully noted. This response is filed with a petition for a one-month Extension of Time, a Request for continued Examination and required fees. Accordingly, Applicant respectfully submits that this response is being timely filed.

Status of Claims:

Prior to these amendments, claims 1-6 and 8–16 were pending. By this amendment, claims 1 and 8 are amended, claims 2 and 12–16 are cancelled, and claims 23–34 are newly added. Accordingly, after entry of this amendment claims 1, 3–6, 8–11, 13, 14 and 23–34 are pending of which claims 1, 9, 23 and 34 are independent.

For the reasons set forth in detail below, all claims are believed to be in condition for allowance. Favorable reconsideration is requested.

Claim Amendments

Claims 1 and 2 are amended to recite the features in claims 2 and 12 respectively. Claims 2 and 12 are accordingly cancelled.

New claim 23–34 find support in the originally filed specification, for example in paragraph [113] and FIGs. 1A–1C describing a positive electrode active material having a first, second and third regions and their organization. Details regarding the compositions of the composition are also found in the as filed specification, for example in paragraphs 123, 132, and 142. A description regarding the degree of mismatch and bonding distances can also be found, for example in paragraph 169 and FIG. 3.

No new matter is added by this amendment.

Objection to the Drawings

The drawings were objected to as allegedly failing to comply with 37 CFR 1.84(p)(4) for various reasons enumerated in the Office Action. Applicant notes with appreciation that this Objection was withdrawn by the Examiner as stated in the Advisory Action.

Objection to the Specification

The disclosure was objected to for various reasons stated in the Office Action. Applicant notes with appreciation that this objection was withdrawn in the Advisory Action.

Claim Rejections – 35 USC § 112

In the Office Action, claims 13-16 were rejected under AIA 35 U.S.C. 112(b). Applicant notes with appreciation that in the Advisory Action, this rejection was withdrawn by the Examiner. Applicant has also cancelled these claims rendering this issue moot with respect to this application.

Claim Rejections Under 35 U.S.C. § 102

Claims 1, 4, 6, 8, and 13-16 stand rejected under AIA 35 U.S.C. 102(a)(1) as allegedly being anticipated by Ooyama et al. (US 2007/0224506 Al, hereinafter Ooyama). Applicant traverses this rejection.

Without acquiescing to this rejection and in order to expedite the prosecution and allowance of this application, independent claim 1 is amended to recite the limitation from claims 2: "wherein a degree of a mismatch between a crystal structure of the lithium cobaltate of the first region and a crystal structure of the lithium titanate of the second region is less than or equal to 0.12, and wherein a degree of a mismatch between the crystal structure of the lithium titanate of the second region and a crystal structure of the magnesium oxide of the third region is less than or equal to 0.12." Ooyama neither expressly or inherently describes at least this element of the instant claims. Examiner by not rejecting claim 2 under this rejection and by acknowledgment that Ooyama does not explicitly disclose a positive electrode active material with these features (page 15 of the Office Action) appears to agree with Applicant.

For at least this reason, the §102 rejection of claim 1 should be withdrawn. The rejection of claims 4, 6 and 8 should be withdrawn at least by virtue of their dependence on claim 1. The rejection of claims 13–16 is rendered moot by their being cancelled by this amendment.

Claim Rejections Under 35 U.S.C. §103

Claims 2 and 3 stand rejected under AIA 35 U.S.C. 103 as allegedly being unpatentable over Ooyama as applied to claim 1 above, and further in view of Yura et al. (US 2012/0258358 Al, hereinafter Yura).

Claim 5 stands rejected under AIA 35 U.S.C. 103 as allegedly being unpatentable over Ooyama as applied to claim 1 above, and further in view of Endo et al. (US 2010/0233542 Al, hereinafter Endo).

Claim 9 stands rejected under AIA 35 U.S.C. 103 as allegedly being unpatentable over Ooyama in view of Takahashi et al. (US 2004/0229123 Al, hereinafter Takahashi).

Claims 10-12 stands rejected under AIA 35 U.S.C. 103 as allegedly being unpatentable over Ooyama in view of Takahashi as applied to claim 9 above, and further in view of Yura.

Applicant traverses these §103 rejections for the following reasons.

In addition to amending claim 1 as described above, Applicant herein amends claim 9 to recite the elements from claim 12, namely "wherein a degree of a mismatch between a crystal structure of the lithium cobaltate of the first region and a crystal structure of the lithium titanate of the second region is less than or equal to 0.12, and wherein a degree of a mismatch between the crystal structure of the lithium titanate of the second region and a crystal structure of the magnesium oxide of the third region is less than or equal to 0.12." Applicant submits that none of the prior art teaches or renders obvious the instant claims in view of these amendments.

The Office Action on page 16 asserts that "Yura discloses a positive electrode active material (222) having a degree of a mismatch between a crystal structure of a first compound (222a) and a crystal structure of the second compound (222a) is less than or equal to 0.12 (see crystal orientations, [0163]), and wherein a degree of a mismatch between the crystal structure of the second compound (222a) and a crystal structure of a third compound (222a) is less than or equal to 0.12 (see crystal orientations, [0163])..." Applicant submits that this assertion is flawed because Yura teaches only one cathode active material layer 22 and within this cathode active material layer 22, a cathode active material particle 222 plural primary grains 222a in particle 222, that is, an aggregate of the primary grains or an assembly of a plural monocrystalline primary grains (see for example Yura paragraph 27 and 58–61 and FIGs. 2–3). That is, compounds 222a are *all the same kind of particle* because orientations of different crystal structure cannot be compared. Yura focuses on electron conduction and lithium ion diffusion pathways, and does not focus on the crystal directions, *e.g.* a-, b-, or c-axes, set for convenience sake in another crystal. Now, the Examiner identified the claimed second region with the coating layer of Ooyama, which would correspond to surface region of the primary grain of Yura. The combination of Ooyama and Yura merely teaches the

coated particle being aggregated with their crystal orientations being aligned similarly. The combination of Ooyama and Yura does not teach a degree of a mismatch between the crystal structures of *different compounds in the same primary grain* as recited in the instant claims.

For at least these reasons, the §103 rejections of the independent claims 1 and 9 cannot be maintained and should be withdrawn. The rejection of the dependent claims 3, 5, 10 and 11 should be withdrawn at least by virtue of their dependence on one of claims 1 or 9.

New Claims

Claim 23–34 are new and claim further aspects of the disclosure Applicant is entitled to seek protection for. Consideration and allowance of this claims is respectfully requested.

CONCLUSION

In view of the foregoing, Applicant respectfully requests allowance of the instant application. If a conference would be helpful in expediting prosecution of the instant application, the Examiner is invited to telephone the undersigned to arrange such a conference.

Except for issue fees payable under 37 C.F.R. § 1.18, the Commissioner is hereby authorized by this paper to charge any additional fees during the entire pendency of this application including fees due under 37 C.F.R. §§ 1.16 and 1.17 which may be required, including any required extension of time fees, or credit any overpayment to Deposit Account No. 19-2380.

Respectfully submitted,

/Jeffrey L. Costellia, Reg. No. 35483/
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Electronic Patent Application Fee Transmittal

Application Number:	15638449
Filing Date:	30-Jun-2017
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Filer:	Jeffrey Costellia/George Hinton
Attorney Docket Number:	740756-004563

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Extension - 1 month with $0 paid	1251	1	200	200
Miscellaneous:				
RCE- 1ST REQUEST	1801	1	1300	1300
		Total in USD ($)		**1500**

Electronic Acknowledgement Receipt

EFS ID:	37732179
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	22204
Filer:	Jeffrey Costellia/George Hinton
Filer Authorized By:	Jeffrey Costellia
Attorney Docket Number:	740756-004563
Receipt Date:	12-NOV-2019
Filing Date:	30-JUN-2017
Time Stamp:	19:40:59
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	DA
Payment was successfully received in RAM	$1500
RAM confirmation Number	E2019ABJ41144765
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Request for Continued Examination (RCE)	740756-004563_RCE_Transmittal.pdf	42096 3204fd04d00fb57d0d5a3374b87bc52dc780a540	no	3
Warnings:					
This is not a USPTO supplied RCE SB30 form.					
Information:					
2		740756-004563_Amendment_in_RCE.pdf	208647 6514d8aa3fefb9fb87d7478b1df126a15b5989d7	yes	10

Multipart Description/PDF files in .zip description

Document Description	Start	End
Amendment Submitted/Entered with Filing of CPA/RCE	1	1
Claims	2	6
Applicant Arguments/Remarks Made in an Amendment	7	10

Warnings:					
Information:					
3	Fee Worksheet (SB06)	fee-info.pdf	32708 894446a2b4191cac2b3bbdcee3a141dbd06d86b9	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	283451		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

PTO/SB/06 (09-11)
Approved for use through 1/31/2014. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
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PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 15/638,449	Filing Date 06/30/2017	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $80 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $420 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	11/12/2019	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 21	Minus	** 22	= 0		x $100 =	0
	Independent (37 CFR 1.16(h))	* 5	Minus	*** 4	= 1		x $460 =	460
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	460

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	LIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/BRENDA MURPHY/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

United States Patent and Trademark Office

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Document Code:WFEE

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Sale Accounting Date:11/18/2019

Sale Item Reference Number	Effective Date
15638449	11/12/2019

Document Number	Fee Code	Fee Code Description	Amount Paid	Payment Method
I2019AF847020014	1201	INDEPENDENT CLAIMS IN EXCESS OF 3	$460.00	Deposit Account

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	740756-004563	1074

22204 7590 12/09/2019

NIXON PEABODY, LLP
799 Ninth Street, NW
SUITE 500
WASHINGTON, DC 20001

EXAMINER
CULLEN, SEAN P

ART UNIT	PAPER NUMBER
1725	

NOTIFICATION DATE	DELIVERY MODE
12/09/2019	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

ipairlink@nixonpeabody.com
nppatent@nixonpeabody.com

Office Action Summary	Application No. 15/638,449	Applicant(s) OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTHS FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☑ Responsive to communication(s) filed on 12 November 2019.

☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2a)☐ This action is **FINAL.** 2b) ☑ This action is non-final.

3)☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

4)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims*

5) ☑ Claim(s) 1,3-6,8-11 and 23-34 is/are pending in the application.

5a) Of the above claim(s) _____ is/are withdrawn from consideration.

6) ☐ Claim(s) _____ is/are allowed.

7) ☑ Claim(s) 1,3-6,8-11 and 23-34 is/are rejected.

8) ☐ Claim(s) _____ is/are objected to.

9) ☐ Claim(s) _____ are subject to restriction and/or election requirement

* If any claims have been determined allowable, you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see http://www.uspto.gov/patents/init_events/pph/index.jsp or send an inquiry to **PPHfeedback@uspto.gov.**

Application Papers

10)☐ The specification is objected to by the Examiner.

11)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

Certified copies:

a)☐ All b)☐ Some** c)☐ None of the:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. ____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

** See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☑ Notice of References Cited (PTO-892)

2) ☐ Information Disclosure Statement(s) (PTO/SB/08a and/or PTO/SB/08b) Paper No(s)/Mail Date _____.

3) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date _____.

4) ☐ Other: _____.

DETAILED ACTION

Notice of Pre-AIA or AIA Status

1. The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

Continued Examination Under 37 CFR 1.114

2. A request for continued examination under 37 CFR 1.114, including the fee set forth in 37 CFR 1.17(e), was filed in this application after final rejection. Since this application is eligible for continued examination under 37 CFR 1.114, and the fee set forth in 37 CFR 1.17(e) has been timely paid, the finality of the previous Office action has been withdrawn pursuant to 37 CFR 1.114. Applicant's submission filed on 12 November 2019 has been entered.

Status of Claims and Other Notes

3. **Claim(s) 1, 3-6, 8-11, and 23-34** is/are pending.

4. **Claim(s) 2, 7, and 12-22** is/are canceled.

5. The text of those sections of Title 35, U.S. Code not included in this action can be found in a prior Office action.

Claim Rejections - 35 USC § 112

6. **Claim(s) 23-34** is/are rejected under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

The term "representative" in **claim 23** is a relative term, which renders the claim indefinite. The term "representative" is not defined by the claim, the specification does not provide a standard for ascertaining the requisite degree, and one of ordinary skill in the art would not be reasonably apprised of the scope of the invention. Therefore, the limitation "representative elements" is indefinite.

Claims 24-28 is directly dependent from claim 23 and include all the limitations of claim 23. Therefore, claims 24-28 are also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

The term "representative" in **claim 29** is a relative term, which renders the claim indefinite. The term "representative" is not defined by the claim, the specification does not provide a standard for ascertaining the requisite degree, and one of ordinary skill in the art would not be reasonably apprised of the scope of the invention. Therefore, the limitation "a representative element" is indefinite.

Claims 30 and 31 is directly dependent from claim 29 and include all the limitations of claim 29. Therefore, claims 30 and 31 are also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

The term "representative" in **claim 32** is a relative term, which renders the claim indefinite. The term "representative" is not defined by the claim, the specification does not provide a standard for ascertaining the requisite degree, and one of ordinary skill in the art would not be reasonably apprised of the scope of the invention. Therefore, the limitation "a representative element" is indefinite.

Claims 33 and 34 is directly dependent from claim 32 and include all the limitations of claim 32. Therefore, claims 33 and 34 are also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

Claim Rejections - 35 USC § 102

7. **Claim(s) 29-34** is/are rejected under 35 U.S.C. 102(a)(1) as being anticipated by Yanagihara et al. (WO 2016/017077 A1; see English language equivalent, US 2017/0207444 A1; hereinafter Yanagihara).

Regarding **claim 29**, Yanagihara discloses a positive electrode active material (Fig. 1, [0061]) comprising:

- a first region (1) in the positive electrode active material (Fig. 1, [0061]);
- a second region (2) closer to a surface of the positive electrode active material than the first region (1, [0061]); and
- a third region (3) closer to the surface of the positive electrode active material than the second region (2, [0061]),
- wherein the first region (1) includes a first oxide of lithium and a first transition metal (see lithium composite oxide, [0063]),
- a type of a crystal structure of the first oxide being a layered rock-salt crystal structure (see lithium composite oxide, [0064]),
- wherein the second region (2) includes a non-stoichiometric compound including a second transition metal ([0064], [0068]),

- a type of a crystal structure of the non-stoichiometric compound being a rock-salt crystal structure ([0064], [0068]), and
- wherein the third region (3) includes a compound including a representative element (see lithium transition metal composite oxide, [0081]),
- a type of a crystal structure of the compound being a rock-salt crystal structure ([0064], [0081]).

Regarding **claim 30**, Yanagihara discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the second region (2) include the first transition metal (see lithium composite oxide, [0068]).

Regarding **claim 31**, Yanagihara discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the third region (3) include the first transition metal and the second transition metal ([0064]-[0066], [0068], [0079]-[0082]).

Regarding **claim 32**, Yanagihara discloses a positive electrode active material (Fig. 1, [0063]) comprising:

- a first region (1) in the positive electrode active material (Fig. 1, [0061]);
- a second region (2) closer to a surface of the positive electrode active material than the first region (1, [0061]); and
- a third region (3) closer to the surface of the positive electrode active material than the second region (2, [0061]),
- wherein the first region (1) includes a first oxide including lithium and at least one of cobalt, manganese, and nickel (see lithium composite oxide, [0064]),

- a type of a crystal structure of the first oxide being a layered rock-salt crystal structure (see lithium composite oxide, [0064]),
- wherein the second region includes a second oxide including at least one of titanium, vanadium, manganese, iron, chromium, niobium, cobalt, zinc, zirconium, and nickel (see lithium composite oxide, [0068]),
- a type of a crystal structure of the second oxide being a rock-salt crystal structure ([0064], [0068]), and
- wherein the third region includes a compound including a representative element (see lithium transition metal composite oxide, [0081]),
- a type of a crystal structure of the compound being a rock-salt crystal structure ([0064], [0081]).

Regarding **claim 33**, Yanagihara discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the second region (2) includes at least one of cobalt, manganese, and nickel ([0064], [0068]).

Regarding **claim 34**, Yanagihara discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the third region (3) includes the one of cobalt, manganese, and nickel (see lithium transition metal composite oxide, [0081]), and
- wherein the third region (3) includes the one of titanium, vanadium, manganese, iron, chromium, niobium, cobalt, zinc, zirconium, and nickel (see lithium transition metal composite oxide, [0081]).

Claim Rejections - 35 USC § 103

8. **Claim(s) 1, 3, 4, 6, 8-11, and 23-28** is/are rejected under 35 U.S.C. 103 as being unpatentable over Yanagihara (WO 2016/017077 A1; see English language equivalent, US 2017/0207444 A1) in view of Kawakami (US 2011/0300441 A1).

Regarding **claims 1 and 3**, Yanagihara discloses a positive electrode active material (Fig. 1, [0061]) comprising:

- a first region (1) in the positive electrode active material (Fig. 1, [0061]);
- a second region (2) in the positive electrode active material (Fig. 1, [0061]),
- the second region (2) surrounding the first region (Fig. 1, [0061]); and
- a third region (3) in the positive electrode active material (Fig. 1, [0061]),
- the third region (3) surrounding the second region (2, [0061]),
- wherein the first region (1) comprises a first compound comprising an oxide of lithium and a first transition metal (see lithium composite oxide, [0064]),
- wherein a type of a crystal structure of the first compound is a layered rock-salt crystal structure (see lithium composite oxide, [0064]),
- wherein the second region (2) comprises a second compound comprising an oxide of a second transition metal (see lithium composite oxide, [0068]),
- wherein a type of a crystal structure of the second compound is a rock-salt crystal structure ([0064], [0068]),
- wherein the second compound is a non-stoichiometric compound ([0064], [0068]),

- wherein the third region (3) comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide (see oxide, [0079]), lithium fluoride, and sodium fluoride (see fluoride, [0082]), and
- wherein a type of a crystal structure of the third compound is the rock-salt crystal structure ([0064], [0084]).

Yanagihara does not explicitly disclose:

- wherein a degree of a mismatch between the crystal structure of the first compound and the crystal structure of the second compound is less than or equal to 0.12, and
- wherein a degree of a mismatch between the crystal structure of the second compound and the crystal structure of the third compound is less than or equal to 0.12; and
- wherein a degree of a mismatch between a (1-1-4) plane of the crystal structure of the first compound or a plane orthogonal to the (1-1-4) plane and a {100} plane of the crystal structure of the second compound is less than or equal to 0.12, and
- wherein a degree of a mismatch between the {100} plane of the crystal structure of the second compound and a {100} plane of the crystal structure of the third compound is less than or equal to 0.12.

Kawakami discloses a positive electrode active material (100) having a degree of a mismatch between a crystal structure of a first compound in a first region (102) and a crystal structure of a second compound in a second region (104) is less than or equal to 0.12 (see aligned, [0087]), and wherein a degree of a mismatch between the crystal structure of the second compound (102) and a crystal structure of a third compound in a third region (104) is less than or

equal to 0.12 (see aligned, [0087]); and wherein a degree of a mismatch between a (1-1-4) plane of the crystal structure of the first compound or a plane orthogonal to the (1-1-4) plane and a {100} plane of the crystal structure of the second compound is less than or equal to 0.12 (see aligned, [0087]), and wherein a degree of a mismatch between the {100} plane of the crystal structure of the second compound and a {100} plane of the crystal structure of the third compound is less than or equal to 0.12 (see aligned, [0087]) to improve the diffusion of lithium into and out of the positive electrode active material ([0087]-[0088]). Yanagihara and Kawakami are analogous art because they are directed to positive electrode active materials for lithium secondary batteries. Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the modify the degree of mismatch between the first and second regions and between the second and third regions of Yanagihara as taught by Kawakami in order to improve the diffusion of lithium into and out of the positive electrode active material.

Regarding **claim 4**, modified Yanagihara discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the first compound is a polycrystal (see polycrystalline, [0063]).

Regarding **claim 6**, modified Yanagihara discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the third compound essentially consists of the one of magnesium oxide, calcium oxide, beryllium oxide (see oxide, [0079]), lithium fluoride, and sodium fluoride (see fluoride, [0082]).

Regarding **claim 8**, Yanagihara discloses a secondary battery (Fig. 3, [0095]) comprising a positive electrode (21) comprising a positive electrode active material (Fig. 3, [0101]); and a negative electrode (22), wherein the positive electrode active material (Fig. 1, [0061]) comprises:

- a first region (1) in the positive electrode active material (Fig. 1, [0061]);
- a second region (2) in the positive electrode active material (Fig. 1, [0061]),
- the second region (2) surrounding the first region (1, [0061]); and
- a third region (3) in the positive electrode active material (Fig. 1, [0061]),
- the third region (3) surrounding the second region (2, [0061]),
- wherein the first region (1) comprises a first compound comprising an oxide of lithium and a first transition metal (see lithium composite oxide, [0064]),
- wherein a type of a crystal structure of the first compound is a layered rock-salt crystal structure (see lithium composite oxide, [0064]),
- wherein the second region (2) comprises a second compound comprising an oxide of a second transition metal (see lithium composite oxide, [0068]),
- wherein a type of a crystal structure of the second compound is a rock-salt crystal structure ([0064], [0068]),
- wherein the second compound is a non-stoichiometric compound ([0064], [0068]),
- wherein the third region (3) comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide (see oxide, [0079]), lithium fluoride, and sodium fluoride (see fluoride, [0082]), and
- wherein a type of a crystal structure of the third compound is the rock-salt crystal structure ([0064], [0084]).

Yanagihara does not explicitly disclose:

- wherein a degree of a mismatch between the crystal structure of the first compound and the crystal structure of the second compound is less than or equal to 0.12, and
- wherein a degree of a mismatch between the crystal structure of the second compound and the crystal structure of the third compound is less than or equal to 0.12.

Kawakami discloses a positive electrode active material (100) having a degree of a mismatch between a crystal structure of a first compound in a first region (102) and a crystal structure of a second compound in a second region (104) is less than or equal to 0.12, and wherein a degree of a mismatch between the crystal structure of the second compound (102) and a crystal structure of a third compound (104) in a third region is less than or equal to 0.12 to improve the diffusion of lithium into and out of the positive electrode active material ([0087]-[0088]). Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the modify the degree of mismatch between the first and second regions and between the second and third regions of Yanagihara as taught by Kawakami in order to improve the diffusion of lithium into and out of the positive electrode active material.

Regarding **claim 9-11**, Yanagihara discloses a positive electrode active material (Fig. 1, [0061]) comprising:

- a first region (1) in the positive electrode active material (Fig. 1, [0061]);
- a second region (2) in the positive electrode active material (Fig. 1, [0061]),
- the second region (2) surrounding the first region (1, [0061]); and
- a third region (3) in the positive electrode active material (Fig. 1, [0061]),

- the third region (3) surrounding the second region (2, [0061]),
- wherein the first region (see lithium composite oxide, [0063]) comprises lithium cobaltate (see Co, [0064]),
- wherein the second region (see lithium composite oxide, [0068]) comprises lithium titanate (see Ti, [0064]), and
- wherein the third region comprises magnesium oxide (see Mg, [0078]),

Yanagihara does not explicitly disclose:

- wherein a degree of a mismatch between a crystal structure of the lithium cobaltate of the first region and a crystal structure of the lithium titanate of the second region is less than or equal to 0.12, and
- wherein a degree of a mismatch between the crystal structure of the lithium titanate of the second region and a crystal structure of the magnesium oxide of the third region is less than or equal to 0.12; and
- wherein a crystal orientation of a part of the lithium cobaltate of the first region and a crystal orientation of a part of the lithium titanate of the second region are substantially aligned with each other;
- wherein a crystal orientation of a part of the lithium titanate of the second region and a crystal orientation of a part of the magnesium oxide of the third region are substantially aligned with each other.

Kawakami discloses a positive electrode active material (100) having a degree of a mismatch between a crystal structure of a first compound in a first region (102) and a crystal structure of a second compound in a second region (102) is less than or equal to 0.12 (see aligned, [0087]), and wherein a degree of a mismatch between the crystal structure of the second

compound (102) and a crystal structure of a third compound (104) in a third region is less than or equal to 0.12 (see aligned, [0087]); and wherein a crystal orientation of a part of the first compound of the first region (102) and a crystal orientation of a part of the second compound of the second region (104) are substantially aligned with each other (see aligned, [0087]); wherein a crystal orientation of a part of the second compound of the second region (102) and a crystal orientation of a part of the third compound of the third region (104) are substantially aligned with each other (see aligned, [0087]) to improve the diffusion of lithium into and out of the positive electrode active material ([0087]-[0088]). Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the modify the degree of mismatch between the first and second regions and between the second and third regions of Yanagihara as taught by Kawakami in order to improve the diffusion of lithium into and out of the positive electrode active material.

Regarding **claim 23-26**, Yanagihara discloses a positive electrode active material (Fig. 1, [0061]) comprising:

- a first region (1) in the positive electrode active material (Fig. 1, [0061]);
- a second region (2) closer to a surface of the positive electrode active material than the first region (1, [0061]); and
- a third region (3) closer to the surface of the positive electrode active material than the second region (2, [0061]),
- wherein the first region (1) includes a first oxide of lithium and a first transition metal (see lithium composite oxide, [0064]),
- wherein the second region includes a second oxide of lithium and a second transition metal (see lithium composite oxide, [0068]),

- wherein the third region includes representative elements and oxygen (see oxide, [0079]).

Yanagihara does not explicitly disclose:

- wherein a second distance between the lithium and the second transition metal through oxygen in the second region is longer than a first distance between the lithium and the first transition metal through oxygen in the first region, and
- wherein a third distance between the representative elements through the oxygen in the third region is longer than the second distance;
- wherein a degree of a mismatch f between the first distance and the second distance is less than or equal to 0.12;
- wherein a degree of a mismatch f between the second distance and the third distance is less than or equal to 0.12; and
- wherein each of the first distance, the second distance, and the third distance is measured by STEM-FFT.

Kawakami discloses a positive electrode active material (100) comprising a second distance between the lithium and the second transition metal through oxygen in a second region (104) is longer than a first distance between the lithium and the first transition metal through oxygen in a first region (102, [0088]), and wherein a third distance between representative elements through the oxygen in a third region (104) is longer than the second distance (see lattice mismatch, [0088]); wherein a degree of a mismatch f between the first distance and the second distance is less than or equal to 0.12 (see aligned, [0087]); wherein a degree of a mismatch f between the second distance and the third distance is less than or equal to 0.12; and wherein each of the first distance (see aligned, [0087]), the second distance, and the third distance is measured

by STEM-FFT (see lattice mismatch, [0088]) to improve the diffusion of lithium into and out of the positive electrode active material ([0087]-[0088]). Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the modify the degree of mismatch between the first and second regions and between the second and third regions of Yanagihara as taught by Kawakami in order to improve the diffusion of lithium into and out of the positive electrode active material.

Regarding **claim 27**, modified Yanagihara discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the second region (2) include the first transition metal (see lithium composite oxide, [0068]).

Regarding **claim 28**, modified Yanagihara discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the third region (3) include the first transition metal and the second transition metal ([0064]-[0066], [0068], [0079]-[0082]).

9. **Claim(s) 5** is/are rejected under 35 U.S.C. 103 as being unpatentable over Yanagihara (WO 2016/017077 A1; see English language equivalent, US 2017/0207444 A1) in view of Kawakami (US 2011/0300441 A1) as applied to **claim(s) 1** above, and further in view of Endo et al. (US 2010/0233542 A1, hereinafter Endo).

Regarding **claim 5**, modified Yanagihara discloses all claim limitations set forth above, but does not explicitly disclose a positive electrode active material:

- wherein an average crystallite size of the oxide of lithium and the first compound is greater than or equal to 280 nm and less than or equal to 630 nm.

Endo discloses a positive electrode active material having an average crystallite size of an oxide of lithium and a first compound greater than or equal to 280 nm and less than or equal to 630 nm to improve the reversible capacity (see crystallite size, [0144]). Yanagihara and Endo are analogous art because they are directed to positive electrode active materials for lithium secondary batteries. Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the first region of modified Yanagihara with the crystallite size of Endo in order to improve the reversible capacity.

Response to Arguments

10. Applicant's arguments with respect to **claim(s) 1, 3-6, 8-11, and 23-34** have been considered but are moot because the arguments do not apply to any of the references being used in the current rejection.

Conclusion

11. The prior art made of record and not relied upon is considered pertinent to applicant's disclosure.

Yanagihara (WO 2016/017079 A1, US 2017/0187035 A1) discloses a positive electrode active material (Fig. 1B, [0068]) comprising a first region (3) in the positive electrode active material (Fig. 1B, [0068]); a second region (4) closer to a surface of the positive electrode active material than the first region (3, [0068]); and a third region (5) closer to the surface of the positive electrode active material than the second region (4, [0068]), wherein the first region (3) includes a first oxide of lithium and a first transition metal (see lithium composite oxide, [0046]), a type of a crystal structure of the first oxide being a layered rock-salt crystal structure (see

lithium composite oxide, [0046]), wherein the second region (4) includes a non-stoichiometric compound including a second transition metal ([0046], [0054]), a type of a crystal structure of the non-stoichiometric compound being a rock-salt crystal structure ([0046], [0054]), and wherein the third region (3) includes a compound including a representative element (see lithium composite oxide, [0072]), a type of a crystal structure of the compound being a rock-salt crystal structure ([0046], [0072]).

Kawakami (US 2011/0269023 A1) discloses a positive electrode active material (100) having a degree of a mismatch between a crystal structure of a first compound in a first region (102) and a crystal structure of a second compound in a second region (104) is less than or equal to 0.12 (see aligned, [0057]), and wherein a degree of a mismatch between the crystal structure of the second compound (102) and a crystal structure of a third compound in a third region (104) is less than or equal to 0.12 (see aligned, [0057]).

12. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Sean P Cullen, Ph.D. whose telephone number is (571)270-1251. The examiner can normally be reached on Monday-Thursday 6:00 am-4:00 pm CDT, Friday 6:00 am-12:00 pm CDT.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Basia A Ridley can be reached on (571)272-1453. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

Notice of References Cited	Application/Control No. 15/638,449	Applicant(s)/Patent Under Reexamination OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	CPC Classification	US Classification
*	A	US-20170207444-A1	07-2017	YANAGIHARA; Asuki	C01G23/005	1/1
*	B	US-20170187035-A1	06-2017	YANAGIHARA; ASUKI	B60L53/20	1/1
*	C	US-20110300441-A1	12-2011	KAWAKAMI; Takahiro	H01M4/136	429/211
*	D	US-20110269023-A1	11-2011	KAWAKAMI; Takahiro	H01M4/136	429/221
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	N	WO-2016017077-A1	02-2016	WO	YANAGIHARA; Asuki	C01B35/1027
	O	WO-2016017079-A1	02-2016	WO	YANAGIHARA; Asuki	H02J7/0068
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NON-PATENT DOCUMENTS

*		Include as applicable: Author, Title Date, Publisher, Edition or Volume, Pertinent Pages)
	U	
	V	
	W	
	X	

*A copy of this reference is not being furnished with this Office action. (See MPEP § 707.05(a).)
Dates in MM-YYYY format are publication dates. Classifications may be US or foreign.

Index of Claims	Application/Control No.	Applicant(s)/Patent Under Reexamination
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	Sean P Cullen, Ph.D.	1725

✓	Rejected	-	Cancelled	N	Non-Elected	A	Appeal
=	Allowed	÷	Restricted	I	Interference	O	Objected

CLAIMS

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIM		DATE								
Final	Original	12/20/2018	03/07/2019	06/27/2019	12/04/2019					
	1	÷	✓	✓	✓					
	2	÷	✓	✓	-					
	3	÷	✓	✓	✓					
	4	÷	✓	✓	✓					
	5	÷	✓	✓	✓					
	6	÷	✓	✓	✓					
	7	÷	✓	-	-					
	8	÷	✓	✓	✓					
	9	÷	✓	✓	✓					
	10	÷	✓	✓	✓					
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	31				✓					
	32				✓					
	33				✓					
	34				✓					

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

CPC - Searched*		
Symbol	Date	Examiner
H01M4/131, 1315, 1391, 13915, 625, 8657	03/07/2019	SPC
Updated CPC search	06/27/2019	SPC
Updated CPC search	12/04/2019	SPC

CPC Combination Sets - Searched*		
Symbol	Date	Examiner

US Classification - Searched*			
Class	Subclass	Date	Examiner
429	231.3	03/07/2019	SPC
	Updated USPC search	06/27/2019	SPC
	Updated USPC search	12/04/2019	SPC

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

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	Examiner	Art Unit
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Search Notes		
Search Notes	**Date**	**Examiner**
H01M4/131, 1315, 1391, 13915, 625, 8657	03/07/2019	SPC
429/231.3	03/07/2019	SPC
Inventor/applicant search, PALM	03/07/2019	SPC
EAST text search, see search history	03/07/2019	SPC
Updated inventor/applicant search, PALM	06/27/2019	SPC
Updated EAST text search, see search history	06/27/2019	SPC
Updated inventor/applicant search, PALM	12/04/2019	SPC
Updated EAST text search, see search history	12/04/2019	SPC

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
S1	1	15/638449.app.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2018/12/20 12:25
S2	166	(US-3172388-$ | US-3959333-$ | US-4302518-$ | US-4668595-$ | US-5240794-$ | US-5443929-$ | US-5783333-$ | US-5824278-$ | US-5834139-$ | US-5871866-$ | US-5910382-$ | US-6085015-$ | US-6346348-$ | US-6458487-$ | US-6514640-$ | US-6582814-$ | US-6680143-$ | US-6846592-$ | US-7138209-$ | US-7303840-$ | US-7368071-$ | US-7393476-$ | US-7635536-$ | US-7709148-$ | US-7736807-$ | US-7790308-$ | US-7892679-$ | US-7927506-$ | US-7935270-$ | US-8003256-$ | US-8080340-$ | US-8470477-$ | US-8476510-$ | US-8557440-$ | US-8685569-$ | US-8685570-$ | US-8709654-$ | US-8877381-$ | US-8883351-$ | US-8927148-$ | US-8945772-$ | US-9225003-$ | US-9227850-$ | US-9391322-$ | US-20010010807-$ | US-20020102459-$ | US-20020164156-$ | US-20020195591-$ | US-20030134186-$ | US-20040234857-$ | US-20060051671-$ | US-20060051673-$ | US-20060121352-$ | US-20060188780-$ | US-20060275664-$ | US-20060286459-$ | US-20070026315-$ | US-20070099086-$ | US-20070122712-$ | US-20070148546-$ | US-20070160906-$ | US-20070212609-$ | US-20090011335-$ | US-20090087731-$ | US-20090123813-$ | US-20090123842-$ | US-20090220862-$ | US-20100035147-$ | US-20100129714-$ | US-20100129715-$ | US-20100143784-$ | US-20100143799-$ | US-20100159330-$ | US-20100178464-$ | US-20100216024-$ | US-20100233542-$ | US-20100248033-$ | US-20110076564-$ | US-20110200879-$ | US-20110229757-$ | US-20120064409-$ | US-20120088151-$ | US-20120088156-$ | US-20120118149-$ | US-20120256337-$ | US-20120258358-$ | US-20120258365-$ | US-20120258369-$ | US-20120308891-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/06 10:02
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S6	261	S2 S3 S4 S5	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/06 10:28
S7	166	(US-3172388-$ | US-3959333-$ | US-4302518-$ | US-4668595-$ | US-5240794-$ | US-5443929-$ | US-5783333-$ | US-5824278-$ | US-5834139-$ | US-5871866-$ | US-5910382-$ | US-6085015-$ | US-6346348-$ | US-6458487-$ | US-6514640-$ | US-6582814-$ | US-6680143-$ | US-6846592-$ | US-7138209-$ | US-7303840-$ | US-7368071-$ | US-7393476-$ | US-7635536-$ | US-7709148-$ | US-7736807-$ | US-7790308-$ | US-7892679-$ | US-7927506-$ | US-7935270-$ | US-8003256-$ | US-8080340-$ | US-8470477-$ | US-8476510-$ | US-8557440-$ | US-8685569-$ | US-8685570-$ | US-8709654-$ | US-8877381-$ | US-8883351-$ | US-8927148-$ | US-8945772-$ | US-9225003-$ | US-9227850-$ | US-9391322-$ | US-20010010807-$ | US-20020102459-$ | US-20020164156-$ | US-20020195591-$ | US-20030134186-$ | US-20040234857-$ | US-20060051671-$ | US-20060051673-$ | US-20060121352-$ | US-20060188780-$ | US-20060275664-$ | US-20060286459-$ | US-20070026315-$ | US-20070099086-$ | US-20070122712-$ | US-20070148546-$ | US-20070160906-$ | US-20070212609-$ | US-20090011335-$ | US-	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44

		20090123813-$ | US-20090123842-$ | US-20090220862-$ | US-20100035147-$ | US-20100129714-$ | US-20100129715-$ | US-20100143784-$ | US -20100143799-$\| US-20100159330-$ | US-20100178464-$ | US-20100216024-$ | US-20100233542-$ | US-20100248033-$ | US-20110076564-$ | US-20110200879-$ | US-20110229757-$ | US-20120064409-$ | US-20120088151-$ | US-20120088156-$ | US-20120118149-$ | US-20120256337-$ | US-20120258358-$ | US-20120258365-$ | US-20120258369-$ | US-20120308891-$).DID.				
S8	55	(CN-101148263-$ | JP-08037007-$ | JP-11025983-$ | JP-2003331824-$ | JP-2004196604-$ | JP-2005225734-$ | JP-2005332629-$ | JP-2006318928-$ | JP-2006318929-$ | JP-2008166156-$ | JP-2009179501-$ | JP-2012066944-$ | JP-2012074366-$ | JP-2012084257-$ | JP-2012169217-$ | JP-2013012410-$ | JP-2013062082-$ | JP-2013091581-$ | JP-2013100197-$ | JP-2013246936-$ | JP-2015082374-$ | JP-2015201432-$ | JP-3172388-$ | JP-3959333-$ | VA-223131450-$ | WO-2011141486-$ | WO-2012124242-$ | WO-2012132387-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S9	35	(US-0033749-$ | US-0034516-$ | US-0131780-$ | US-0944137-$ | US-20080131780-$ | US-20110033749-$ | US-20120034516-$).DID. (CN-101208269-$ | CN-101675547-$ | CN-102210045-$ | EP-2352190-$ | IB-2017053896-$ | JP-2011181527-$ | JP-2012508444-$ | KR-100944137-$ | KR-20080024105-$ | KR-20090086440-$ | KR-20100052419-$ | VA-223131450-$ | WO-2007052712-$ | WO-2009057722-$ | WO-2010053328-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S10	6	(JP-2004103566-$ | JP-2004342554-$ | VA-223131450-$ | WO-2012005180-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S11	261	S7 S8 S9 S10	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S12	3	S11 and mismatch	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S13	0	S11 and mis-match	US-PGPUB; USPAT; USOCR;	OR	ON	2019/03/07 09:45

			FPRS; EPO; JPO; DERWENT; IBM_TDB			
S14	1	S11 and crystal$4 near2 align$4	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:45
S15	4	S12 S14	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:45
S16	2	US-20110300441-$.did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:49
S17	1	S16 and mismatch	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:49
S18	9	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) with (lithium adj titanate "LiTiO.sub.2") with (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:03
S19	1	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) with (TTIP (Ti titanium) adj (tetraisopropoxide tetra adj isopropoxide tetra adj (isoprop isopropyl) adj oxide))	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:06
S20	3	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) same (TTIP (Ti titanium) adj (tetraisopropoxide tetra adj isopropoxide tetra adj (isoprop isopropyl) adj oxide))	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:07
S21	4	(JP-2004103566-$\|WO-2012005180-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:11
S22	6	(JP-2004103566-$\|WO-2012005180-$|JP-	US-PGPUB;	OR	ON	2019/03/07

		2004342554-$).did.	USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB			10:12
S23	18	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) with (lithium adj titanate "LiTiO.sub.2") near2 (coating layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:21
S24	35	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide LiCoO2) with (lithium adj titanate "LiTiO.sub.2" LiTiO2) near2 (coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:22
S25	6	(WO-20014032405-$\|JP-2004103566-$|JP-2003007299-$\|JP-2003331846-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:26
S26	8	(WO-2014032405-$\|JP-2004103566-$|JP-2003007299-$\|JP-2003331846-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:26
S27	4	(US-20030211391-$\|US-20030082448-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:29
S28	35	H01M.cpcl. and ((Li lithium) adj cobaltate "LiCo.2" (Li lithium) adj (Co cobalt) adj oxide LiCoO2) with ((Li lithium) adj titanate "LiTiO.sub.2" LiTiO2) near2 (coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:31
S29	35	H01M.cpcl. and ((Li lithium) adj cobaltate "LiCo.2" (Li lithium) adj (Co cobalt) adj oxide LiCoO2) with ((Li lithium) adj titanate "LiTiO.sub.2" LiTiO2) near2 (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:33
S30	0	H01M.cpcl. and ((Li lithium) adj cobaltate "LiCo.2" (Li lithium) adj (Co cobalt) adj oxide LiCoO2) same ((Li lithium) adj titanate "LiTiO.sub.2" LiTiO2) near2 (shell coating coat coated layer film) same ((Mg magnesium) adj oxide MgO) near2 (shell	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT;	OR	ON	2019/03/07 10:33

		coating coat coated layer film)	IBM_TDB			
S31	122	H01M.cpcl. and ((Li lithium) adj cobaltate "LiCo.2" (Li lithium) adj (Co cobalt) adj oxide LiCoO2) with ((Li lithium) adj titanate "LiTiO.sub.2" LiTiO2) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:33
S32	87	S31 not S29	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:34
S33	1	H01M.cpcl. and ((Li lithium) adj cobaltate "LiCo.2" (Li lithium) adj (Co cobalt) adj oxide LiCoO2) same ((Li lithium) adj titanate "LiTiO.sub.2" LiTiO2) with (shell coating coat coated layer film) same ((Mg magnesium) adj oxide MgO) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:34
S34	2	H01M.cpcl. and (LCO (Li lithium) adj cobaltate "LiCo.2" (Li lithium) adj (Co cobalt) adj oxide LiCoO2) same (LTO (Li lithium) adj titanate "LiTiO.sub.2" LiTiO2) with (shell coating coat coated layer film) same ((Mg magnesium) adj oxide MgO) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:35
S35	152	H01M.cpcl. and (LCO (Li lithium) adj cobaltate "LiCo.2" (Li lithium) adj (Co cobalt) adj oxide LiCoO2) with (LTO (Li lithium) adj titanate "LiTiO.sub.2" LiTiO2) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:37
S36	117	S35 not S29	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:37
S37	6	(JP-10333573-$\|JP-10372470-$|JP-2987358-$\|JP-2004227869-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:46
S38	2	(JP-10236826-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:47
S39	4	(JP-2002151078-$|JP-2005123111-$$).did.	US-PGPUB; USPAT; USOCR; FPRS;	OR	ON	2019/03/07 10:50

			EPO; JPO; DERWENT; IBM_TDB			
S40	2	(US-20040229123-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:52
S41	77	(US-20040229123-$ | US-20060105240-$ | US-20060105241-$ | US-20070298324-$ | US-20080182170-$ | US-20090081554-$ | US-20100173197-$ | US-20110033749-$ | US-20110037037-$ | US-20110287321-$ | US-20120273737-$ | US-20130244113-$ | US-20150194655-$).DID. (JP-08053776-$ | JP-08183605-$ | JP-2000164214-$ | JP-2000200605-$ | JP-2004002066-$ | JP-2007080680-$ | JP-2007103306-$ | JP-2008103244-$ | JP-2008159543-$ | JP-2011028999-$ | JP-2011181527-$ | JP-2012030988-$ | JP-2012113823-$ | JP-2013073707-$ | JP-2013114786-$ | JP-2014096281-$ | JP-2014170656-$ | JP-2018206747-$ | JP-2018508116-$ | KR-20060085085-$ | KR-20090086440-$ | WO-2004070863-$ | WO-2006114047-$ | WO-2007004590-$ | WO-2009057722-$ | WO-2010090185-$ | WO-2013034821-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:59
S42	1	15/638449.app.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 11:00
S43	64	S41 not (S35 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 11:00
S44	7	(KR-20060085085-$\|JP-2014096281-$|JP-2012113823-$\|JP-2000200605-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 11:54
S45	1	(KR-2006085085-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 11:55
S46	10	(US-20070202411-$ | US-20120021284-$ | US-20120156565-$ | US-20160149215-$).DID. (KR-20070082892-$ | KR-	US-PGPUB; USPAT; USOCR;	OR	ON N	2019/03/07 12:11

		20120069398-$).DID.	FPRS; EPO; JPO; DERWENT; IBM_TDB			
S47	10	S46 not (S41 S35 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:11
S48	4	(US-20140057173-$\|US-20150017550-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:20
S49	17	(US-20030087155-$ | US-20160149215-$ | US-9343745-$).DID. (EP-3086389-$ | JP-2017216243-$ | KR-20080045855-$ | KR-20180064196-$ | WO-2015093725-$ | WO-2018038501-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:24
S50	15	S49 not (S46 l36l30 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:24
S51	2	US-20160372739-$.did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:30
S52	153	(US-20010053480-$ | US-20020071990-$ | US-20030082448-$ | US-20040005265-$ | US-20060154146-$ | US-20060166096-$ | US-20060239883-$ | US-20070048615-$ | US-20080131778-$ | US-20080160414-$ | US-20080318131-$ | US-20090068561-$ | US-20090087746-$ | US-20090220862-$ | US-20090263707-$ | US-20100086853-$ | US-20100086854-$ | US-20100151332-$ | US-20100255384-$ | US-20110017528-$ | US-20110052981-$ | US-20110052989-$ | US-20110076556-$ | US-20110111298-$ | US-20110136019-$ | US-20110236751-$ | US-20110244331-$ | US-20120028105-$ | US-20120056590-$ | US-20120070725-$ | US-20120105007-$ | US-20120107680-$ | US-20120156565-$ | US-20120263987-$ | US-20130043843-$ | US-20130202953-$ | US-20130216701-$ | US-20130216900-$ | US-20140027670-$ | US-20140050972-$ | US-20140050993-$ | US-20140065464-$ | US-20140131616-$ | US-20140178760-$ | US-20140308572-$ | US-20150030927-$ |	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:44

		US-20150030928-$ | US-20150037678-$ | US-20150132651-$ | US-20150364748-$ | US-5292601-$ | US-5705291-$).DID. (CN-101894938-$ | CN-103354958-$ | CN-103975466-$ | CN-104521040-$ | CN-105378985-$ | EP-2814093-$ | EP-2869369-$ | EP-3007253-$ | JP-2008311132-$ | JP-2009016302-$ | JP-2009206047-$ | JP-2011082150-$ | JP-2014238957-$ | JP-2015529958-$ | JP-2016041659-$ | JP-2016131156-$ | KR-20070018293-$ | KR-20070097923-$ | KR-20100059023-$ | KR-20120069398-$ | KR-20120079801-$ | KR-20130080565-$ | WO-2005048380-$ | WO-2007114557-$ | WO-2010147179-$ | WO-2012093797-$ | WO-2012093798-$ | WO-2013132663-$ | WO-2014073833-$ | WO-2017047021-$).DID.				
S53	146	S52 not (S49 S46 S41 S35 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:44
S54	169	(US-20020071991-$ | US-20020076613-$ | US-20030082448-$ | US-20050196674-$ | US-20070224506-$ | US-20090200509-$ | US-20100015516-$ | US-20110042609-$ | US-20110042610-$ | US-20110059367-$ | US-20110200880-$ | US-20110274965-$ | US-20120003538-$ | US-20120027929-$ | US-20120028118-$ | US-20120034503-$ | US-20120156565-$ | US-20120258373-$ | US-20130323596-$ | US-20140127582-$ | US-20140154552-$ | US-20140154581-$ | US-20140170494-$ | US-20140212745-$ | US-20150171424-$ | US-20150228974-$ | US-20150349339-$ | US-20160020487-$ | US-20160056457-$ | US-20160172663-$ | US-20160308202-$ | US-20170179481-$ | US-20170263929-$ | US-20180062156-$ | US-20180083278-$).DID. (CN-107251306-$ | EP-2940762-$ | EP-3419086-$ | FR-2937185-$ | JP-08138670-$ | JP-09265984-$ | JP-1116566-$ | JP-1167209-$ | JP-2000231919-$ | JP-2001313034-$ | JP-2002075367-$ | JP-2002124262-$ | JP-2002164053-$ | JP-2002279991-$ | JP-2003007299-$ | JP-2003017053-$ | JP-2003217572-$ | JP-2003221234-$ | JP-2004002066-$ | JP-2004014405-$ | JP-2004175609-$ | JP-2005243301-$ | JP-2005276454-$ | JP-2006073482-$ | JP-2006173099-$ | JP-2006503789-$ | JP-2007066839-$ | JP-2007258095-$ | JP-2009146739-$ | JP-2009218198-$ | JP-2010092706-$ | JP-2010118312-$ | JP-2011003354-$ | JP-2011124118-$ | JP-2011204661-$ | JP-2012018796-$ | JP-2012028231-$ | JP-2012142154-$ | JP-2013125732-$ | JP-2014082219-$ | JP-2014120202-$ | JP-2017033641-$ | JP-	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:12

		2018511911-$ | KR-100914406-$ | KR-20160127887-$ | TW-201208183-$ | TW-201208184-$ | WO-2006018921-$ | WO-2006054604-$ | WO-2009120008-$ | WO-2010040950-$ | WO-2014104466-$ | WO-2014104467-$ | WO-2014128903-$ | WO-2015102091-$ | WO-2016136211-$ | WO-2016175426-$ | WO-2017138309-$).DID.				
S55	160	S54 not (S52 S49 S46 S41 S35 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:13
S56	4	(JP-2017033641-$\|JP-2007258095-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:39
S57	108	(US-20020071991-$ | US-20030087155-$ | US-20080131778-$ | US-20090200509-$ | US-20090239146-$ | US-20100028768-$ | US-20100181540-$ | US-20100301284-$ | US-20110200883-$ | US-20120225357-$ | US-20130029217-$ | US-20130059200-$ | US-20130078504-$ | US-20140065483-$ | US-20140087257-$ | US-20140170492-$ | US-20140212745-$ | US-20140295274-$ | US-20150194672-$ | US-20150214539-$ | US-20150243983-$ | US-20160006032-$ | US-20160079597-$ | US-20170190595-$ | US-5677083-$).DID. (CN-102092798-$ | CN-103715421-$ | CN-104078666-$ | CN-104704654-$ | CN-105185962-$ | CN-106299347-$ | EP-2157639-$ | EP-2784846-$ | GB-2506859-$ | JP-0935715-$ | JP-2000164214-$ | JP-2002231227-$ | JP-2002279991-$ | JP-2003221234-$ | JP-2004014405-$ | JP-2005340056-$ | JP-2006012426-$ | JP-2006036545-$ | JP-2008016232-$ | JP-2008016243-$ | JP-2008041570-$ | JP-2009123465-$ | JP-2010073354-$ | JP-2010073366-$ | JP-2010277929-$ | JP-2011028976-$ | JP-2014523840-$ | JP-2016062683-$ | KR-20110010568-$ | KR-20140118264-$ | WO-2011016553-$ | WO-2011059766-$ | WO-2012170693-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:40
S58	88	S57 not (S54 S52 S49 S46 S41 S35 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:40
S59	6	(WO-2011016553-$\|JP-2011028976-$|JP-2008016243-$).did.	US-PGPUB; USPAT; USOCR; FPRS;	OR	ON	2019/03/07 14:01

			EPO; JPO; DERWENT; IBM_TDB			
S60	4	(US-20120135315-$\|US-20110020708-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:02
S61	42	(US-20180013130-$ or US-20120258358-$ or US-20100233542-$ or US-20110300441-$ or US-20140227606-$ or US-20100173197-$ or US-20030211391-$ or US-20030082448-$ or US-20160104880-$ or US-20040229123-$ or US-20110287321-$ or US-20160149215-$ or US-20030087155-$ or US-20160372739-$ or US-20080131778-$ or US-20160308202-$ or US-20160056457-$ or US-20110042610-$ or US-20070224506-$ or US-20020076613-$ or US-20020071991-$ or US-20160079597-$ or US-20160006032-$ or US-20120225357-$ or US-20100301284-$ or US-20100181540-$).did. or (US-20120135315-$ or US-20110020708-$).did. or (US-8927148-$).did. or (WO-2012005180-$ or WO-2014032405-$ or KR-20060085085-$ or WO-2011016553-$).did. or (JP-2004342554-$ or JP-2004103566-$ or JP-2003331846-$ or JP-2002151078-$ or JP-2012113823-$ or JP-2010118312-$ or JP-2009218198-$ or JP-2017033641-$ or JP-2008016243-$).did.	US-PGPUB; USPAT; FPRS; JPO	OR	ON	2019/03/07 14:02
S62	6	S61 and rock adj salt	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:03
S63	1	S61 and Fm3m	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:06
S64	1	S62 and S63	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:06
S65	0	S61 and (NaCl sodium adj chloride) near2 (crystal structure)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT;	OR	ON	2019/03/07 14:10

			IBM_TDB			
S66	2	S61 and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) and (lithium adj titanate "LiTiO.sub.2") and (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:27
S67	10	S61 and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) and ((lithium adj titanate "LiTiO.sub.2") (magnesium adj oxide MgO))	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:32
S68	3	S61 and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide LCO) and (LTO lithium adj titanate "LiTiO.sub.2") and (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:33
S69	11	S61 and (LCO lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) and ((LTO lithium adj titanate "LiTiO.sub.2") (magnesium adj oxide MgO))	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:34
S70	8	S69 not S68	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:34
S71	5	S61 and (mismatch mis-match crystal$4 near2 align$4)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 15:15
S72	2	S61 and (crystallite crystal) with (nm nanometer nanometre nano-meter nano-metre nano adj (meter metre))	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 15:29
S73	14	S61 and (size diameter) with (nm nanometer nanometre nano-meter nano-metre nano adj (meter metre))	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 15:30
S74	42	(US-20180013130-$ or US-20120258358-$ or US-20100233542-$ or US-20110300441-$ or US-20140227606-$ or US-20100173197-$ or US-20030211391-$	US-PGPUB; USPAT; FPRS; JPO	OR	ON	2019/03/08 12:02

		or US-20030082448-$ or US-20160104880-$ or US-20040229123-$ or US-20110287321-$ or US-20160149215-$ or US-20030087155-$ or US-20160372739-$ or US-20080131778-$ or US-20160308202-$ or US-20160056457-$ or US-20110042610-$ or US-20070224506-$ or US-20020076613-$ or US-20020071991-$ or US-20160079597-$ or US-20160006032-$ or US-20120225357-$ or US-20100301284-$ or US-20100181540-$).did. or (US-20120135315-$ or US-20110020708-$).did. or (US-8927148-$).did. or (WO-2012005180-$ or WO-2014032405-$ or KR-20060085085-$ or WO-2011016553-$).did. or (JP-2004342554-$ or JP-2004103566-$ or JP-2003331846-$ or JP-2002151078-$ or JP-2012113823-$ or JP-2010118312-$ or JP-2009218198-$ or JP-2017033641-$ or JP-2008016243-$).did.				
S75	38	S74 and (F fluoride fluorine)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/08 12:02
S76	17	(US-6984469-$\|US-5604396-$|US-9478796-$\|JP-2002164053-$|JP-08100107-$\|JP-07316551-$|JP-2015527707-$\|EP-0638625-$|WO-2014015139-$\|WO-2012132387-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/06/27 12:18
S77	2	US-20070224506-$.did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/06/27 13:25
S78	16	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) same (lithium adj titanate "LiTiO.sub.2") same (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:09
S79	14	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) with (lithium adj titanate "LiTiO.sub.2") with (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:09
S80	16	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) same (lithium adj titanate "LiTiO.sub.2") same (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT;	OR	ON	2019/07/08 10:09

			IBM_TDB			
S81	14	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) with (lithium adj titanate "LiTiO.sub.2") with (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:10
S82	2	S80 not S81	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:10
S83	21	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) and (lithium adj titanate "LiTiO.sub.2") with (shell coating coat coated layer film) and (magnesium adj oxide MgO) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:11
S84	15	S83 not S80	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:11
S85	16	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.sub.2" LiCo2 lithium adj cobalt adj oxide) same (lithium adj titanate "LiTiO.sub.2") same (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:12
S86	16	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.sub.2" LiCoO2 lithium adj cobalt adj oxide) same (lithium adj titanate "LiTiO.sub.2") same (magnesium adj oxide MgO)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:13
S87	31	H01M.cpcl. and (lithium adj cobaltate "LiCoO.sub.2" LiCoO2 lithium adj cobalt adj oxide) and (lithium adj titanate "LiTiO.sub.2") with (shell coating coat coated layer film) and (magnesium adj oxide MgO) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:13
S88	25	S87 not S80	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/07/08 10:13
S89	8	(JP-2002164053-$\|JP-08100107-$|JP-07316551-$\|JP-2015527707-$).did.	US-PGPUB; USPAT; USOCR; FPRS;	OR	ON	2019/12/03 12:37

			EPO; JPO; DERWENT; IBM_TDB			
S90	2	(WO-2014015139-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/03 12:38
S91	2	US-20070224506-$.did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/03 14:29
S92	44	(US-20180013130-$ or US-20120258358-$ or US-20100233542-$ or US-20110300441-$ or US-20140227606-$ or US-20100173197-$ or US-20030211391-$ or US-20030082448-$ or US-20160104880-$ or US-20040229123-$ or US-20110287321-$ or US-20160149215-$ or US-20030087155-$ or US-20160372739-$ or US-20080131778-$ or US-20160308202-$ or US-20160056457-$ or US-20110042610-$ or US-20070224506-$ or US-20020076613-$ or US-20020071991-$ or US-20160079597-$ or US-20160006032-$ or US-20120225357-$ or US-20100301284-$ or US-20100181540-$).did. or (US-20120135315-$ or US-20110020708-$ or US-20170162917-$).did. or (US-8927148-$).did. or (WO-2012005180-$ or WO-2014032405-$ or KR-20060085085-$ or WO-2011016553-$).did. or (JP-2004342554-$ or JP-2004103566-$ or JP-2003331846-$ or JP-2002151078-$ or JP-2012113823-$ or JP-2010118312-$ or JP-2009218198-$ or JP-2017033641-$ or JP-2008016243-$ or JP-2002164053-$).did.	US-PGPUB; USPAT; FPRS; JPO	OR	ON	2019/12/03 14:30
S93	1	S92 and (rock adj s alt NaCl) adj crystal	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/03 14:30
S94	2	(US-20120258358-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/03 14:32
S95	153	H01M.cpcl. and (LCO lithium adj cobaltate "LiCo.sub.2" LiCo2 lithium adj cobalt adj oxide) same (lithium adj (vanadate titanate manganate iron ferrate chromate niobate zincate zirconate nickelate)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO;	OR	ON	2019/12/03 14:47

		"LiTiO.sub.2" "TiO.sub.2" TiO2 LiTiO2 "V.sub.2O.sub.5" "LiV.sub.2O.sub.5" LiV2O5 "LiMnO.sub.2" "LiMn.sub.2O.sub.4" LiMnO2 LiMn2O4 "LiFeO.sub.4" LiFeO4 "LiCr.sub.2O.sub.3" LiCrO "LiCrO.sub.2" LiCrO2 LiCr2O3 "LiNbO.sub.3" "LiNbO.sub.2" "LiNbO.sub.3" "LiZnO.sub.2" LiZnO2 LiZnO "LiZrO.sub.2" LiZrO2 "LiNi.sub.2O.sub.4" LiNi2O4 NiO LiNiO "LiNiO.sub.2" LiNiO2 "MnO.sub.2" "Mn.sub.2O.sub.4" MnO2 Mn2O4 "FeO.sub.4" FeO4 "Cr.sub.2O.sub.3" CrO "CrO.sub.2" CrO2 Cr2O3 "NbO.sub.3" "NbO.sub.2" "NbO.sub.3" "ZnO.sub.2" ZnO2 ZnO "ZrO.sub.2" ZrO2 "Ni.sub.2O.sub.4" Ni2O4 NiO NiO "NiO.sub.2" NiO2) same ((beryllium magnesium) adj oxide MgO BeO (sodium lithium) adj fluoride NaF LiF)	DERWENT; IBM_TDB			
S96	613	H01M.cpcl. and (lithium adj cobaltate "LiCoO.sub.2" LiCoO2 lithium adj cobalt adj oxide) and (lithium adj (vanadate titanate manganate iron ferrate chromate niobate zincate zirconate nickelate) "LiTiO.sub.2" "TiO.sub.2" TiO2 LiTiO2 "V.sub.2O.sub.5" "LiV.sub.2O.sub.5" LiV2O5 "LiMnO.sub.2" "LiMn.sub.2O.sub.4" LiMnO2 LiMn2O4 "LiFeO.sub.4" LiFeO4 "LiCr.sub.2O.sub.3" LiCrO "LiCrO.sub.2" LiCrO2 LiCr2O3 "LiNbO.sub.3" "LiNbO.sub.2" "LiNbO.sub.3" "LiZnO.sub.2" LiZnO2 LiZnO "LiZrO.sub.2" LiZrO2 "LiNi.sub.2O.sub.4" LiNi2O4 NiO LiNiO "LiNiO.sub.2" LiNiO2 "MnO.sub.2" "Mn.sub.2O.sub.4" MnO2 Mn2O4 "FeO.sub.4" FeO4 "Cr.sub.2O.sub.3" CrO "CrO.sub.2" CrO2 Cr2O3 "NbO.sub.3" "NbO.sub.2" "NbO.sub.3" "ZnO.sub.2" ZnO2 ZnO "ZrO.sub.2" ZrO2 "Ni.sub.2O.sub.4" Ni2O4 NiO NiO "NiO.sub.2" NiO2) with (shell coating coat coated layer film) and ((beryllium magnesium) adj oxide MgO BeO (sodium lithium) adj fluoride NaF LiF) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/03 14:49
S97	579	S96 not S95	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/03 15:18
S98	137	(US-20150340689-$ | US-20170294651-$).DID. (CN-101944599-$ | CN-102324504-$ | CN-102332577-$ | CN-102354750-$ | CN-103746114-$ | CN-104201374-$ | CN-104362295-$ | CN-104766959-$ | CN-105161679-$ | CN-105226271-$ | CN-105244490-$ | CN-105449196-$ | CN-105655566-$ | CN-106395920-$ | CN-107240690-$ | CN-109659518-$ | CN-1416189-$ | CN-1627550-$ | EP-3248233-$ | EP-3316357-	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 09:38

		$ | EP-3367484-$ | JP-2018511911-$ | JP-2018525314-$).DID. (US-20150104708-$).DID. (CN-101678775-$ | CN-101997113-$ | CN-102088084-$ | CN-102479951-$ | CN-102612775-$ | CN-103178249-$ | CN-103296249-$ | CN-103779554-$ | CN-103943822-$ | CN-107978744-$ | CN-108011126-$ | JP-05021066-$ | JP-10144291-$ | JP-2001256979-$ | JP-2005336000-$ | JP-2005524204-$ | JP-2008041570-$ | JP-2008251532-$ | JP-2009081103-$ | JP-2010092848-$ | JP-2010135207-$ | JP-2010177042-$ | JP-2010251194-$ | JP-2013026038-$ | JP-2013218787-$ | KR-20120100022-$ | KR-20130014245-$ | KR-20130084616-$ | KR-20140067508-$ | KR-20150021809-$ | KR-20180003983-$ | WO-2018026650-$ | WO-2018225450-$ | WO-2019159563-$ | WO-2019189679-$).DID. (US-20130183579-$).DID.				
S99	4	(US-20180053940-$\|US-20170207444-$|US-20170187035-$\|US-20150104708-$).DID. and ((beryllium magnesium) adj oxide MgO BeO (sodium lithium) adj fluoride NaF LiF) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 09:44
S100	613	H01M.cpcl. and (lithium adj cobaltate "LiCoO.sub.2" LiCoO2 lithium adj cobalt adj oxide) and (lithium adj (vanadate titanate manganate iron ferrate chromate niobate zincate zirconate nickelate) "LiTiO.sub.2" "TiO.sub.2" TiO2 LiTiO2 "V.sub.2O.sub.5" "LiV.sub.2O.sub.5" LiV2O5 "LiMnO.sub.2" "LiMn.sub.2O.sub.4" LiMnO2 LiMn2O4 "LiFeO.sub.4" LiFeO4 "LiCr.sub.2O.sub.3" LiCrO "LiCrO.sub.2" LiCrO2 LiCr2O3 "LiNbO.sub.3" "LiNbO.sub.2" "LiNbO.sub.3" "LiZnO.sub.2" LiZnO2 LiZnO "LiZrO.sub.2" LiZrO2 "LiNi.sub.2O.sub.4" LiNi2O4 NiO LiNiO "LiNiO.sub.2" LiNiO2 "MnO.sub.2" "Mn.sub.2O.sub.4" MnO2 Mn2O4 "FeO.sub.4" FeO4 "Cr.sub.2O.sub.3" CrO "CrO.sub.2" CrO2 Cr2O3 "NbO.sub.3" "NbO.sub.2" "NbO.sub.3" "ZnO.sub.2" ZnO2 ZnO "ZrO.sub.2" ZrO2 "Ni.sub.2O.sub.4" Ni2O4 NiO NiO "NiO.sub.2" NiO2) with (shell coating coat coated layer film) and ((beryllium magnesium) adj oxide MgO BeO (sodium lithium) adj fluoride NaF LiF) with (shell coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 09:44
S101	1	S98 and S100	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 09:44
S102	2	S99 and (rock NaCl)	US-PGPUB; USPAT;	OR R	ON	2019/12/04 09:47

			USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB			
S103	4	(US-20170207444-$\|US-20170187035-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 12:24
S104	61	(US-20180013130-$ or US-20120258358-$ or US-20100233542-$ or US-20110300441-$ or US-20140227606-$ or US-20100173197-$ or US-20030211391-$ or US-20030082448-$ or US-20160104880-$ or US-20040229123-$ or US-20110287321-$ or US-20160149215-$ or US-20030087155-$ or US-20160372739-$ or US-20080131778-$ or US-20160308202-$ or US-20160056457-$ or US-20110042610-$ or US-20070224506-$ or US-20020076613-$ or US-20020071991-$ or US-20160079597-$ or US-20160006032-$ or US-20120225357-$ or US-20100301284-$ or US-20100181540-$).did. or (US-20120135315-$ or US-20110020708-$ or US-20170162917-$ or US-20190221845-$ or US-20190221837-$ or US-20180079655-$ or US-20180006302-$ or US-20180366723-$ or US-20170207442-$ or US-20180248179-$ or US-20180212237-$ or US-20180053940-$ or US-20140099549-$ or US-20150255789-$ or US-20090202892-$ or US-20090081549-$ or US-20080138710-$ or US-20150340689-$ or US-20150104708-$ or US-20130183579-$).did. or (US-8927148-$).did. or (WO-2012005180-$ or WO-2014032405-$ or KR-20060085085-$ or WO-2011016553-$).did. or (JP-2004342554-$ or JP-2004103566-$ or JP-2003331846-$ or JP-2002151078-$ or JP-2012113823-$ or JP-2010118312-$ or JP-2009218198-$ or JP-2017033641-$ or JP-2008016243-$ or JP-2002164053-$).did.	US-PGPUB; USPAT; FPRS; JPO	OR	ON	2019/12/04 13:49
S105	4	S104 and (mismatch mis-match mis adj match)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 13:49
S106	365	H01M.cpcl. and (mismatch mis-match mis adj match align$4) with (lattice crystal) with (shell coating coat coated layer film region)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 13:51

S107	280	H01M.cpcl. and (mismatch mis-match mis adj match align$4) near4 (lattice crystal) with (shell coating coat coated layer film region)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 13:52
S108	21	H01M.cpcl. and (mismatch mis-match mis adj match align$4) near4 (lattice crystal) with core	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 14:13
S109	0	S108 not S107	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 14:14
S110	92	H01M.cpcl. and distort$4 near4 (lattice crystal) with (shell coating coat coated layer film region)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 14:15
S111	84	S110 not S107	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 14:16
S112	5	H01M.cpcl. and distort$4 near4 (lattice crystal) with core	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 14:24
S113	71	(US-20180013130-$ or US-20120258358-$ or US-20100233542-$ or US-20110300441-$ or US-20140227606-$ or US-20100173197-$ or US-20030211391-$ or US-20030082448-$ or US-20160104880-$ or US-20040229123-$ or US-20110287321-$ or US-20160149215-$ or US-20030087155-$ or US-20160372739-$ or US-20080131778-$ or US-20160308202-$ or US-20160056457-$ or US-20110042610-$ or US-20070224506-$ or US-20020076613-$ or US-20020071991-$ or US-20160079597-$ or US-20160006032-$ or US-20120225357-$ or US-20100301284-$ or US-20100181540-$).did. or (US-20120135315-$ or US-20110020708-$ or US-20170162917-$ or US-20190221845-$ or US-20190221837-$ or US-20180079655-$ or US-20180006302-$ or	US-PGPUB; USPAT; FPRS; JPO	OR	ON	2019/12/04 15:02

		US-20180366723-$ or US-20170207442-$ or US-20180248179-$ or US-20180212237-$ or US-20180053940-$ or US-20140099549-$ or US-20150255789-$ or US-20090202892-$ or US-20090081549-$ or US-20080138710-$ or US-20150340689-$ or US-20150104708-$ or US-20130183579-$ or US-20170040607-$ or US-20180366726-$ or US-20180145368-$ or US-20180102536-$ or US-20170358797-$ or US-20140315090-$ or US-20130029842-$).did. or (US-20110269023-$ or US-20170207444-$ or US-20170187035-$).did. or (US-8927148-$).did. or (WO-2012005180-$ or WO-2014032405-$ or KR-20060085085-$ or WO-2011016553-$).did. or (JP-2004342554-$ or JP-2004103566-$ or JP-2003331846-$ or JP-2002151078-$ or JP-2012113823-$ or JP-2010118312-$ or JP-2009218198-$ or JP-2017033641-$ or JP-2008016243-$ or JP-2002164053-$).did.				
S114	14	S113 and (mismatch mis-match mis adj match align$4 distort$4) near4 (lattice crystal) with (core shell coating coat coated layer film region)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/12/04 15:03

EAST Search History (Interference)

< This search history is empty>

12/ 4/ 2019 3:41:37 PM
C:\ Users\ scullen\ Documents\ EAST\ Workspaces\ 15638449.wsp

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	740756-004563	1074

22204 7590 01/21/2020

NIXON PEABODY, LLP
799 Ninth Street, NW
SUITE 500
WASHINGTON, DC 20001

EXAMINER	
CULLEN, SEAN P	

ART UNIT	PAPER NUMBER
1725	

NOTIFICATION DATE	DELIVERY MODE
01/21/2020	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

ipairlink@nixonpeabody.com
nppatent@nixonpeabody.com

Applicant-Initiated Interview Summary	Application No. 15/638,449	Applicant(s) OCHIAI et al.		
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	AIA (First Inventor to File) Status Yes	Page 1 of 2

All participants (applicant, applicants representative, PTO personnel):

1. Sean P Cullen, Ph.D. (Primary Examiner); WebEx/ Video Conference
2. Francis de Rege (Agent); WebEx/Video Conference
3. Jeffery Costellia (Attorney); WebEx/Video Conference
4. Sumiyo Onda (Agent); WebEx/Video Conference
5. Hirotada Oishi (Applicant); WebEx/Video Conference
6. Takuya Kimishima (Applicant); WebEx/Video Conference
7. Yoshimi Nakashima (Applicant); WebEx/Video Conference
8. Rie Yanagisawa (Applicant); WebEx/Video Conference
9. Ryota Tajima (Applicant); WebEx/Video Conference

Date of Interview: 15 January 2020

Claims Discussed: 35-90

Prior Art Discussed: Yanagihara (WO 2016/017077 A1)

Amendment proposed: See attached proposed amendments

Issues Discussed:

Item(s) under 35 U.S.C. 112:
It was agreed that the 112b rejections will be withdrawn if applicants make a statement that the term " representative element" is used in the plain meaning according to one skilled in the art.

Item(s) under 35 U.S.C. 102:
It was agreed that the proposed amendments would overcome the prior art of Yanagihara. Further consideration and/or search will be required.

Attachment(s): Agenda, Proposed Amendments,

/Sean P Cullen, Ph.D./ Primary Examiner, Art Unit 1725	

Applicant is reminded that a complete written statement as to the substance of the interview must be made of record in the application file. It is the applicants responsibility to provide the written statement, unless the interview was initiated by the Examiner and the Examiner has indicated that a written summary will be provided. See MPEP 713.04
Please further see:
MPEP 713.04
Title 37 Code of Federal Regulations (CFR) § 1.133 Interviews, paragraph (b)
37 CFR § 1.2 Business to be transacted in writing

Applicant recordation instructions: The formal written reply to the last Office action must include the substance of the interview. (See MPEP section 713.04). If a reply to the last Office action has already been filed, applicant is given a non-extendable period of the longer of one month or thirty days from this interview date, or the mailing date of this interview summary form, whichever is later, to file a statement of the substance of the interview.

Examiner recordation instructions: Examiners must summarize the substance of any interview of record. A complete and proper recordation of the substance of an interview should include the items listed in MPEP 713.04 for complete and proper recordation including the identification of the general thrust of each argument or issue discussed, a general indication of any other pertinent matters discussed regarding patentability and the general results or outcome of the interview, to include an indication as to whether or not agreement was reached on the issues raised.

Cullen, Sean

From: de Rege, Francis <fderege@nixonpeabody.com>
Sent: Tuesday, January 14, 2020 3:30 PM
To: Cullen, Sean
Cc: Costellia, Jeffrey; Onda, Sumiyo
Subject: FW: Examiner Interview for US application 15/638,449

Dear Examiner Cullen,

We are looking forward to discussing the application 15/638,449 tomorrow at 10 pm tomorrow.

We will explain how the 112 rejection should be withdrawn since the term "representative element" is clear, for example in view of para. [0083] of the as filed specification.

We will also present claim amendments and explain how the amended claims are not anticipated by Yanagihara, nor rendered obvious by the combination of Yanagihara and Kawakami.
The amended claims show and arrangement and concentrations of atoms in particles that is not rendered obvious by the art of record. Some of the amendments will be at least in part drawn from Figure 28, Para. [0052], para. [0020], Figure 1B, para. [0471] - [0472] and Figure 32: all from the as filed specification.

We are also open to suggestions on how to further advance the prosecution.

Sincerely,
Francis

Francis de Rege
Patent Agent
fderege@nixonpeabody.com
T 617-345-6199 | C 630-802-2152 | F 617-345-1300
Nixon Peabody LLP | Exchange Place | 53 State Street | Boston, MA 02109-2835
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From: Cullen, Sean <sean.cullen@USPTO.GOV>
Sent: December 26, 2019 6:48 PM
To: Onda, Sumiyo <sonda@nixonpeabody.com>
Subject: RE: Test email: Scheduling Video interview in January 2020

[EXTERNAL E-MAIL]

Sumiyo,

Proposed Amendment:

1.-34. (Canceled)

35. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal; and

a second region comprising a second transition metal, a representative element, and fluorine,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein an angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image.

36. (New) The positive electrode active material layer according to claim 35,

wherein the second region covers at least a part of the first region.

37. (New) The positive electrode active material layer according to claim 35,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

38. (New) The positive electrode active material layer according to claim 35,

wherein the second region is provided in a range from a surface of the positive electrode active material particle to a depth of 20 nm.

39. (New) The positive electrode active material layer according to claim 35,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

40. (New) The positive electrode active material layer according to claim 35,

wherein the second region is positioned closer to a surface of the positive electrode active material particle than the first region is.

41. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 35;

a negative electrode; and

an electrolyte solution.

42. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal; and

a second region comprising a second transition metal, a representative element, and fluorine,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

43. (New) The positive electrode active material layer according to claim 42,

wherein the second region covers at least a part of the first region.

44. (New) The positive electrode active material layer according to claim 42,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

45. (New) The positive electrode active material layer according to claim 42,

wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm.

46. (New) The positive electrode active material layer according to claim 42,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

47. (New) The positive electrode active material layer according to claim 42,

wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is.

48. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 42;

a negative electrode; and

an electrolyte solution.

49. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal; and

a second region comprising a second transition metal, a representative element, and fluorine,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein a peak of the second transition metal is positioned closer to a surface of the positive electrode active material particle than the first region is in an EDX line analysis.

50. (New) The positive electrode active material layer according to claim 49,

wherein the second region covers at least a part of the first region.

51. (New) The positive electrode active material layer according to claim 49,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

52. (New) The positive electrode active material layer according to claim 49,

wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm.

53. (New) The positive electrode active material layer according to claim 49,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

54. (New) The positive electrode active material layer according to claim 49,

wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is.

55. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 49;

a negative electrode; and

an electrolyte solution.

56. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising a first transition metal, a second transition metal, a representative element, and fluorine,

wherein the representative element and the fluorine are present in a portion which includes crystal defects in the positive electrode active material particle.

57. (New) The positive electrode active material layer according to claim 56,

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

58. (New) The positive electrode active material layer according to claim 56,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

59. (New) The positive electrode active material layer according to claim 56,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

60. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising a first transition metal, a second transition metal, a representative element, and fluorine,

wherein the representative element and the fluorine are present in a crack portion in the positive electrode active material particle.

61. (New) The positive electrode active material layer according to claim 60,

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

62. (New) The positive electrode active material layer according to claim 60,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

63. (New) The positive electrode active material layer according to claim 60,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

64. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal and aluminum; and

a second region comprising a second transition metal,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein an angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image.

65. (New) The positive electrode active material layer according to claim 64,

wherein the second region covers at least a part of the first region.

66. (New) The positive electrode active material layer according to claim 64,

wherein the second region is provided in a range from a surface of the positive electrode active material particle to a depth of 20 nm.

67. (New) The positive electrode active material layer according to claim 64,

wherein the first transition metal is cobalt, and

wherein the second transition metal is titanium.

68. (New) The positive electrode active material layer according to claim 64,

wherein the second region further comprises fluorine.

69. (New) The positive electrode active material layer according to claim 64,

wherein the second region further comprises fluorine and a representative element.

70. (New) The positive electrode active material layer according to claim 69,

wherein the representative element is magnesium.

71. (New) The positive electrode active material layer according to claim 64,

wherein the second region is positioned closer to a surface of the positive electrode

active material particle than the first region is.

72. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 64;

a negative electrode; and

an electrolyte solution.

73. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal and aluminum; and

a second region comprising a second transition metal,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

74. (New) The positive electrode active material layer according to claim 73,

wherein the second region covers at least a part of the first region.

75. (New) The positive electrode active material layer according to claim 73,

wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm.

76. (New) The positive electrode active material layer according to claim 73,

wherein the first transition metal is cobalt, and

wherein the second transition metal is titanium.

77. (New) The positive electrode active material layer according to claim 73,

wherein the second region further comprises fluorine.

78. (New) The positive electrode active material layer according to claim 73,

wherein the second region further comprises fluorine and a representative element.

79. (New) The positive electrode active material layer according to claim 78,

wherein the representative element is magnesium.

80. (New) The positive electrode active material layer according to claim 73,

wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is.

81. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 73;

a negative electrode; and

an electrolyte solution.

82. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal and aluminum; and

a second region comprising a second transition metal,

wherein the second region is in a superficial portion of the positive electrode active

material particle, and

wherein a peak of the second transition metal is positioned closer to a surface of the positive electrode active material particle than the first region is in an EDX line analysis.

83. (New) The positive electrode active material layer according to claim 82,

wherein the second region covers at least a part of the first region.

84. (New) The positive electrode active material layer according to claim 82,

wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm.

85. (New) The positive electrode active material layer according to claim 82,

wherein the first transition metal is cobalt, and

wherein the second transition metal is titanium.

86. (New) The positive electrode active material layer according to claim 82,

wherein the second region further comprises fluorine.

87. (New) The positive electrode active material layer according to claim 82,

wherein the second region further comprises fluorine and a representative element.

88. (New) The positive electrode active material layer according to claim 87,

wherein the representative element is magnesium.

89. (New) The positive electrode active material layer according to claim 82,

wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is.

90. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 82;

a negative electrode; and

an electrolyte solution.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:	)	
Teruaki **OCHIAI** et al.	)	Confirmation No.: 1074
Application No. 15/638,449	)	Examiner: Sean P. Cullen
Filed: June 30, 2017	)	Group Art Unit: 1725
For POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY	)	

AMENDMENT

VIA EFS Amendment
United States Patent and Trademark Office
Randolph Building
401 Dulany Street
Alexandria, VA 22314

Dear Sir:

In response to the Office Action mailed **December 9, 2019**, please amend the above-identified application and consider Applicant remarks as follows.

Submitted herewith are additional fees associated with excess claims fees.

Amendments to the Claims are reflected in the listing of claims, which begins on page **2** of this paper.

Listing of Claims begins on page **2** of this paper.

Remarks begin on page **11** of this paper.

The listing of claims will replace all prior versions, and listings, of claims in the application:

Listings of Claims:

1.-34. (Canceled)

35. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal; and

a second region comprising a second transition metal, a representative element, and fluorine,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein an angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image.

36. (New) The positive electrode active material layer according to claim 35,

wherein the second region covers at least a part of the first region.

37. (New) The positive electrode active material layer according to claim 35,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

38. (New) The positive electrode active material layer according to claim 35,

wherein the second region is provided in a range from a surface of the positive electrode active material particle to a depth of 20 nm.

39. (New) The positive electrode active material layer according to claim 35,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

40. (New) The positive electrode active material layer according to claim 35,

wherein the second region is positioned closer to a surface of the positive electrode active material particle than the first region is.

41. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 35;

a negative electrode; and

an electrolyte solution.

42. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal; and

a second region comprising a second transition metal, a representative element, and fluorine,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

43. (New) The positive electrode active material layer according to claim 42,

wherein the second region covers at least a part of the first region.

44. (New) The positive electrode active material layer according to claim 42,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

45. (New) The positive electrode active material layer according to claim 42,

wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm.

46. (New) The positive electrode active material layer according to claim 42,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

47. (New) The positive electrode active material layer according to claim 42,

wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is.

48. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 42;

a negative electrode; and

an electrolyte solution.

49. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal; and

a second region comprising a second transition metal, a representative element, and fluorine,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein a peak of the second transition metal is positioned closer to a surface of the positive electrode active material particle than to the first region in an EDX line analysis.

50. (New) The positive electrode active material layer according to claim 49,

wherein the second region covers at least a part of the first region.

51. (New) The positive electrode active material layer according to claim 49,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

52. (New) The positive electrode active material layer according to claim 49,

wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm.

53. (New) The positive electrode active material layer according to claim 49,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

54. (New) The positive electrode active material layer according to claim 49,

wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is.

55. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 49;

a negative electrode; and

an electrolyte solution.

56. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising a first transition metal, a second transition metal, a representative element, and fluorine,

wherein the representative element and the fluorine are present in a portion which includes crystal defects in the positive electrode active material particle.

57. (New) The positive electrode active material layer according to claim 56,

wherein a relative value of a concentration of the second transition metal is greater

than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

58. (New) The positive electrode active material layer according to claim 56,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

59. (New) The positive electrode active material layer according to claim 56,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

60. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising a first transition metal, a second transition metal, a representative element, and fluorine,

wherein the representative element and the fluorine are present in a crack portion in the positive electrode active material particle.

61. (New) The positive electrode active material layer according to claim 60,

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

62. (New) The positive electrode active material layer according to claim 60,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

63. (New) The positive electrode active material layer according to claim 60,
wherein the first transition metal is cobalt,
wherein the second transition metal is titanium, and
wherein the representative element is magnesium.

64. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:
a positive electrode active material particle comprising:
a first region comprising a first transition metal and aluminum; and
a second region comprising a second transition metal,
wherein the second region is in a superficial portion of the positive electrode active material particle, and
wherein an angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image.

65. (New) The positive electrode active material layer according to claim 64,
wherein the second region covers at least a part of the first region.

66. (New) The positive electrode active material layer according to claim 64,
wherein the second region is provided in a range from a surface of the positive electrode active material particle to a depth of 20 nm.

67. (New) The positive electrode active material layer according to claim 64,
wherein the first transition metal is cobalt, and
wherein the second transition metal is titanium.

68. (New) The positive electrode active material layer according to claim 64,
wherein the second region further comprises fluorine.

69. (New) The positive electrode active material layer according to claim 64,
wherein the second region further comprises fluorine and a representative element.

70. (New) The positive electrode active material layer according to claim 69,
wherein the representative element is magnesium.

71. (New) The positive electrode active material layer according to claim 64,

wherein the second region is positioned closer to a surface of the positive electrode active material particle than the first region is.

72. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 64;

a negative electrode; and

an electrolyte solution.

73. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal and aluminum; and

a second region comprising a second transition metal,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

74. (New) The positive electrode active material layer according to claim 73,

wherein the second region covers at least a part of the first region.

75. (New) The positive electrode active material layer according to claim 73,

wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm.

76. (New) The positive electrode active material layer according to claim 73,

wherein the first transition metal is cobalt, and

wherein the second transition metal is titanium.

77. (New) The positive electrode active material layer according to claim 73, wherein the second region further comprises fluorine.

78. (New) The positive electrode active material layer according to claim 73, wherein the second region further comprises fluorine and a representative element.

79. (New) The positive electrode active material layer according to claim 78, wherein the representative element is magnesium.

80. (New) The positive electrode active material layer according to claim 73, wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is.

81. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 73;

a negative electrode; and

an electrolyte solution.

82. (New) A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising:

a positive electrode active material particle comprising:

a first region comprising a first transition metal and aluminum; and

a second region comprising a second transition metal,

wherein the second region is in a superficial portion of the positive electrode active material particle, and

wherein a peak of the second transition metal is positioned closer to a surface of the positive electrode active material particle than to the first region in an EDX line analysis.

83. (New) The positive electrode active material layer according to claim 82, wherein the second region covers at least a part of the first region.

84. (New) The positive electrode active material layer according to claim 82,

wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm.

85. (New) The positive electrode active material layer according to claim 82,

wherein the first transition metal is cobalt, and

wherein the second transition metal is titanium.

86. (New) The positive electrode active material layer according to claim 82,

wherein the second region further comprises fluorine.

87. (New) The positive electrode active material layer according to claim 82,

wherein the second region further comprises fluorine and a representative element.

88. (New) The positive electrode active material layer according to claim 87,

wherein the representative element is magnesium.

89. (New) The positive electrode active material layer according to claim 82,

wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is.

90. (New) An electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising:

a positive electrode comprising the positive electrode active material layer according to claim 82;

a negative electrode; and

an electrolyte solution.

REMARKS

The Office Action mailed **December 9, 2019**, is received and its contents carefully noted. This response is filed within three months of the mailing date of the Office Action and therefore is believed to be timely without extension of time. Accordingly, Applicant respectfully submits that this response is being timely filed.

Interview Summary

Applicant and the undersigned thank the Examiner for the cordial Examiner Interview conducted by Video on January 15, 2020. During the Interview the Examiner indicated that the proposed amendments appear to overcome the art and the new claims would require an additional search. The Examiner also indicated the 112b rejections can be overcome by clarifying that the plain meaning for the term "representative" element is used. Although no agreement regarding the allowability of claims was reached, the Interview has advanced prosecution.

Status of Claims:

By this amendment, pending claims 1, 3-6, 8-11 and 23-34 are cancelled. New claims 35–90 are added by this amendment, of which claims 35, 42, 49, 56, 60, 64, 73, and 82 are independent.

Applicant submits that no new matter is introduced by this amendment.

For the reasons set forth in detail below, all claims are believed to be in condition for allowance. Favorable reconsideration is requested.

Claims Amendments

Independent Claims

Claim 35, 42, 49, 56, 60, 64, 73, 82 recite "a positive electrode active material" as in the originally filed claims and further specifying "for a lithium-ion secondary battery" which is supported in the as filed specification ("the specification"), for example the title. These claims also find support for a first region containing a first transition metal and aluminum, 101, in Fig. 1A and described in the specification para. [0059]–[0062]. The second region can overlap a third region (e.g., 103 FIG. 1A is a third region) as supported in the specification para. [0139], Fig. 28, and Fig. 32. Support for the second region being in a superficial portion is found in the as filed specification, for example in paragraph [0012] filed specification, where region 102 and 103 overlap, and 103 is in a superficial portion. The recitation wherein

the second region comprises a second transition metal, a representative element and fluorine, is supported in the specification in, for example in paragraph [0068], [0077] and [0083].

Claim 35 and 64, recite angles in the first and second region and also finds support in the specification, at least in paragraph [0052] and Fig. 28.

Claim 42 and 73 reciting concentrations of the second transition metal find support in the as filed specification, at least in para. [0142].

Claim 49 and 82 further recite a peak of the second transition metal positioned closer to a surface of the particle as supported in the specification, for example in para. [0140], para. [0472] and Fig. 32.

Claims 56 and 60 recite a portion which includes crystal defects as supported in the specification, for example para. [0057]–[0058] and Fig. 1B.

Dependent Claims

Claims 36, 43, 50, 65, 74 and 83 are dependent claims further specifying partial coverage of the second region with the second region and is supported by the as filed specification para. [0055] and Fig. 1B.

Claims 37, 44, 51, 58 and 62 are dependent claims and recite concentrations of the representative element and are supported in the as files specification, for example in para. [0142].

Claims 38, 45, 52, 66, 75, and 84 are dependent claims and recites a depth for the second region as supported in the as filed specification in para. [0082].

Claims 39, 46, 53, 59, 63, 67– 69, 70, 76–79, and 85–88 are dependent claims that further specify one or more of the following; the first transition metal is cobalt, the second transition metal is titanium, and the representative element is magnesium, the second region further comprises fluorine. This is supported in the as files specification for example in para. [0016]–[0017].

Claims 4, 47, 54, 71, 80 and 89 are dependent claims that further specify the second region is positioned close to the surface of the particle. This is supported in the as files specification, for example as previously described for supporting overlapping regions an in para. [0015] and Fig. 1A.

Claims 41, 48, 55, 72, 81 and 90 are dependent claims and further recite an electronic device comprising an active material, a positive and negative electrode and an electrolyte. Support for this can be found throughout the specification, for example in para. [0001], [0254–0255], and [0267]–[0268].

Claims 57 and 61 are dependent claims and recite concentrations of the second transition metal finding support in the as filed specification, at least in para. [0142] as previously described.

Claim Rejections – 35 USC § 112

Claims 23-34 are rejected under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention. Applicant traverses these rejections.

The Examiner contends the term "representative" is a relative term. Applicant disagrees and the plain meaning as understood by a person of ordinary skill in the art is the meaning intended.

Although this rejection is moot with respect to claims 23–34 which are now cancelled, the rejection is addressed here for clarification of the term representative used in the new claims.

Claim Rejections Under 35 U.S.C. § 102 and §10

Claims 29-34 are rejected under 35 U.S.C. 102(a)(1) as being anticipated by Yanagihara et al. (WO 2016/017077 Al; see English language equivalent, US 2017/0207444 Al; hereinafter Yanagihara).

Claims 1, 3, 4, 6, 8-11, and 23-28 are rejected under 35 U.S.C. 103 as being unpatentable over Yanagihara in view of Kawakami (US 2011/0300441 Al; hereinafter Kawakami). Applicant traverses these rejections.

Applicant traverses these rejections.

Without acquiescing to the Examiner's characterization of the claims and solely to prosecution Applicants have cancelled the claims1, 3-6, 8-11 and 23-34 obviating these rejections. Applicant also respectfully submits that no combination of Yanagihara and Kawakami teaches or renders obvious each and every element of the instant claims.

Applicant notes that it is by the methods described in the instant specification, for example in the specification para. [0021] and para. [0173] which describes two high temperature heating steps to form a crystalline particle and then to add and segregate elements therein. These result in segregation and arrangement of elements as claimed. The Inventors of the instant Application realized that this segregation provides a structure for stabilizing the first region (e.g., para. [0072] of the specification). Furthermore, it is the

instant invention that recognizes benefits with respect to applications such as in secondary battery charging/discharging performance, for example as described in para. [0095] of the specification.

CONCLUSION

In view of the foregoing, Applicant respectfully requests allowance of the instant application. If a conference would be helpful in expediting prosecution of the instant application, the Examiner is invited to telephone the undersigned to arrange such a conference.

Respectfully submitted,

/Jeffrey L. Costellia, Reg. No. 35,483/
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Electronic Patent Application Fee Transmittal

Application Number:	15638449
Filing Date:	30-Jun-2017
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Filer:	Jeffrey Costellia/George Hinton
Attorney Docket Number:	740756-004563

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
CLAIMS IN EXCESS OF 20	1202	34	100	3400
INDEPENDENT CLAIMS IN EXCESS OF 3	1201	3	460	1380
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				
Miscellaneous:				
			Total in USD ($)	**4780**

Electronic Acknowledgement Receipt

EFS ID:	38592168
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	22204
Filer:	Jeffrey Costellia/George Hinton
Filer Authorized By:	Jeffrey Costellia
Attorney Docket Number:	740756-004563
Receipt Date:	14-FEB-2020
Filing Date:	30-JUN-2017
Time Stamp:	17:08:31
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	DA
Payment was successfully received in RAM	$4780
RAM confirmation Number	E20202DH09097093
Deposit Account	192380
Authorized User	George Hinton

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

37 CFR 1.16 (National application filing, search, and examination fees)

37 CFR 1.17 (Patent application and reexamination processing fees)

37 CFR 1.21 (Miscellaneous fees and charges)

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		740756-004563_Response.pdf	189719 cb3b08b18d06c6e0eca0b5c7a3a58ad644b4c45f	yes	14

Multipart Description/PDF files in .zip description		
Document Description	**Start**	**End**
Amendment/Req. Reconsideration-After Non-Final Reject	1	1
Claims	2	10
Applicant Arguments/Remarks Made in an Amendment	11	14

Warnings:

Information:

2	Fee Worksheet (SB06)	fee-info.pdf	32235 37bf7cabd56432db07bb073674b907adbd0c70cc	no	2

Warnings:

Information:

Total Files Size (in bytes):	221954

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

PTO/SB/06 (09-11)
Approved for use through 1/31/2014. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 15/638,449	Filing Date 06/30/2017	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $80 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $420 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	02/14/2020	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 56	Minus	** 22	= 34		x $100 =	3400
	Independent (37 CFR 1.16(h))	* 8	Minus	*** 5	= 3		x $460 =	1380
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	4780

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	LIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/VERONICA EVERETTE/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	740756-004563	1074

22204 7590 03/02/2020

NIXON PEABODY, LLP
799 Ninth Street, NW
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WASHINGTON, DC 20001

EXAMINER	
CULLEN, SEAN P	

ART UNIT	PAPER NUMBER
1725	

NOTIFICATION DATE	DELIVERY MODE
03/02/2020	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

ipairlink@nixonpeabody.com
nppatent@nixonpeabody.com

Office Action Summary	Application No. 15/638,449	Applicant(s) OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTHS FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☑ Responsive to communication(s) filed on 14 February 2020.

☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2a)☑ This action is **FINAL.** 2b) ☐ This action is non-final.

3)☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

4)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims*

5) ☑ Claim(s) 35-90 is/are pending in the application.

5a) Of the above claim(s) _____ is/are withdrawn from consideration.

6) ☑ Claim(s) 56-63 is/are allowed.

7) ☑ Claim(s) 35-55 and 64-90 is/are rejected.

8) ☐ Claim(s) _____ is/are objected to.

9) ☐ Claim(s) _____ are subject to restriction and/or election requirement

* If any claims have been determined allowable, you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see http://www.uspto.gov/patents/init_events/pph/index.jsp or send an inquiry to **PPHfeedback@uspto.gov.**

Application Papers

10)☐ The specification is objected to by the Examiner.

11)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

Certified copies:

a)☐ All b)☐ Some** c)☐ None of the:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. ____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

** See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☑ Notice of References Cited (PTO-892)

2) ☐ Information Disclosure Statement(s) (PTO/SB/08a and/or PTO/SB/08b) Paper No(s)/Mail Date _____.

3) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date _____.

4) ☐ Other: _____.

DETAILED ACTION

Notice of Pre-AIA or AIA Status

1. The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

Status of Claims and Other Notes

2. **Claim(s) 35-90** is/are pending

3. **Claim(s) 1-34** is/are canceled.

4. The text of those sections of Title 35, U.S. Code not included in this action can be found in a prior Office action.

Claim Rejections - 35 USC § 112

5. Applicants' amendment(s) have overcome the rejection(s) of **claim(s) 23-34** under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph.

6. **Claim(s) 35-41, 44, and 64-72** is/are rejected under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

The term "bright" in **claim 35** is a relative term which renders the claim indefinite. The term "bright" is not defined by the claim, the specification does not provide a standard for ascertaining the requisite degree, and one of ordinary skill in the art would not be reasonably apprised of the scope of the invention. Therefore, the limitation "a bright line" is indefinite.

Claims 36-40 are directly dependent from claim 35 and include all the limitations of claim 35. Therefore, claims 36-40 are also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

Claim 41 recites the limitation "the positive electrode active material layer according to claim 35" and includes all the limitations of claim 35. Therefore, claim 41 is also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

Claim 44 recites the limitation "an XPS analysis." Claim 42, which claim 44 is directly dependent, recites the limitation "an XPS analysis." It is unclear if "an XPS analysis" recited in claim 44 is further limiting or referring to "an XPS analysis" recited in claim 42.

Claim 44 recites the limitation "a concentration of the first transition metal." Claim 42, which claim 44 is directly dependent, recites the limitation "a concentration of the first transition metal." It is unclear if "a concentration of the first transition metal" recited in claim 44 is further limiting or referring to "a concentration of the first transition metal" recited in claim 42.

The term "bright" in **claim 64** is a relative term which renders the claim indefinite. The term "bright" is not defined by the claim, the specification does not provide a standard for ascertaining the requisite degree, and one of ordinary skill in the art would not be reasonably apprised of the scope of the invention. Therefore, the limitation "a bright line" is indefinite.

Claims 65-71 are directly or indirectly dependent from claim 64 and include all the limitations of claim 64. Therefore, claims 65-71 are also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

Claim 72 recites the limitation "the positive electrode active material layer according to claim 64" and includes all the limitations of claim 64. Therefore, claim 72 is also indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention.

Claim Rejections - 35 USC § 102

7. **Claim(s) 42-55 and 73-90** is/are rejected under 35 U.S.C. 102(a)(1) as being anticipated by Liu et al. (CN 102610806 A, see machine translation, hereinafter Liu).

Regarding **claim 42**, Liu discloses a positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising a positive electrode active material particle comprising:

- a first region comprising a first transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal, a representative element, and fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1 ([0010]-[0012]).

Regarding **claim 43**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region covers at least a part of the first region ([0010]-[0012]).

Regarding **claim 44**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1 ([0010]-[0012]).

Regarding **claim 45**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm ([0010]-[0012]).

Regarding **claim 46**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the first transition metal is cobalt (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second transition metal is titanium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein the representative element is magnesium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 47**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is ([0010]-[0012]).

Regarding **claim 48**, Liu discloses an electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising a positive electrode comprising a positive electrode active material layer; a negative electrode; and an electrolyte solution, wherein the positive electrode active material layer comprises a positive electrode active material particle comprising:

- a first region comprising a first transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal, a representative element, and fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1 ([0010]-[0012]).

Regarding **claim 49**, Liu discloses a positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising: a positive electrode active material particle comprising:

- a first region comprising a first transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal, a representative element, and fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),

- wherein the second region is in a superficial portion of the positive electrode active material particle (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein a peak of the second transition metal is positioned closer to a surface of the positive electrode active material particle than to the first region in an EDX line analysis ([0010]-[0012]).

Regarding **claim 50**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region covers at least a part of the first region ([0010]-[0012]).

Regarding **claim 51**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1 ([0010]-[0012]).

Regarding **claim 52**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm ([0010]-[0012]).

Regarding **claim 53**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the first transition metal is cobalt (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),

- wherein the second transition metal is titanium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein the representative element is magnesium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 54**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is ([0010]-[0012]).

Regarding **claim 55**, Liu discloses an electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising a positive electrode comprising a positive electrode active material layer; a negative electrode; and an electrolyte solution, wherein the positive electrode active material layer comprises a positive electrode active material particle comprising:

- a first region comprising a first transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal, a representative element, and fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein a peak of the second transition metal is positioned closer to a surface of the positive electrode active material particle than to the first region in an EDX line analysis ([0010]-[0012]).

Regarding **claim 73**, Liu discloses a positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising a positive electrode active material particle comprising:

- a first region comprising a first transition metal and aluminum (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle ([0010]-[0012]), and
- wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1 ([0010]-[0012]).

Regarding **claim 74**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region covers at least a part of the first region ([0010]-[0012]).

Regarding **claim 75**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm ([0010]-[0012]).

Regarding **claim 76**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the first transition metal is cobalt (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein the second transition metal is titanium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 77**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region further comprises fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 78**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region further comprises fluorine and a representative element (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 79**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the representative element is magnesium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 80**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is ([0010]-[0012]).

Regarding **claim 81**, Liu discloses an electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising a positive electrode comprising

a positive electrode active material layer; a negative electrode; and an electrolyte solution, wherein the positive electrode active material layer comprises:

- a first region comprising a first transition metal and aluminum (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle ([0010]-[0012]), and
- wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1 ([0010]-[0012]).

Regarding **claim 82**, Liu discloses a positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising a positive electrode active material particle comprising:

- a first region comprising a first transition metal and aluminum (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle ([0010]-[0012]), and

- wherein a peak of the second transition metal is positioned closer to a surface of the positive electrode active material particle than to the first region in an EDX line analysis ([0010]-[0012]).

Regarding **claim 83**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region covers at least a part of the first region ([0010]-[0012]).

Regarding **claim 84**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is provided in a range from the surface of the positive electrode active material particle to a depth of 20 nm ([0010]-[0012]).

Regarding **claim 85**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the first transition metal is cobalt (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein the second transition metal is titanium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 86**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region further comprises fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 87**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region further comprises fluorine and a representative element (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 88**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the representative element is magnesium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 89**, Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is positioned closer to the surface of the positive electrode active material particle than the first region is ([0010]-[0012]).

Regarding **claim 90**, Liu discloses an electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising a positive electrode comprising a positive electrode active material layer; a negative electrode; and an electrolyte solution, wherein the positive electrode active material layer comprises a positive electrode active material particle comprising:

- a first region comprising a first transition metal and aluminum (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle ([0010]-[0012]), and

- wherein a peak of the second transition metal is positioned closer to a surface of the positive electrode active material particle than to the first region in an EDX line analysis ([0010]-[0012]).

Claim Rejections - 35 USC § 103

8. **Claim(s) 35-41 and 64-72** is/are rejected under 35 U.S.C. 103 as being unpatentable over Liu (CN 102610806 A, see machine translation) in view of Sugie et al. (US 2015/0357641 A1, hereinafter Sugie).

Regarding **claim 35**, Liu discloses a positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising a positive electrode active material particle comprising:

- a first region comprising a first transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal, a representative element, and fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Liu does not explicitly disclose:

- wherein an angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image.

Sugie discloses positive electrode active material particle where an angle between a bright line in a first region and a bright line in a second region is less than or equal to 5° in a

transmission electron microscope image (Fig. 4, [0253]) to improve the structural stability of the positive electrode active material particle (see structural stability, [0043]). Liu and Sugie are analogous art because they are directed to positive electrode active material particles for lithium-ion secondary batteries. Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the positive electrode active material particle of Liu with the angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image as taught by Sugie in order to improve the structural stability of the positive electrode active material particle.

Regarding **claim 36**, modified Liu discloses all claim limitations set forth above and further a positive electrode active material layer:

- wherein the second region covers at least a part of the first region ([0010]-[0012]).

Regarding **claim 37**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1 ([0010]-[0012]).

Regarding **claim 38**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is provided in a range from a surface of the positive electrode active material particle to a depth of 20 nm ([0010]-[0012]).

Regarding **claim 39**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the first transition metal is cobalt (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second transition metal is titanium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and
- wherein the representative element is magnesium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 40**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is positioned closer to a surface of the positive electrode active material particle than the first region is ([0010]-[0012]).

Regarding **claim 41**, Liu discloses an electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising a positive electrode comprising a positive electrode active material layer; a negative electrode; and an electrolyte solution, wherein the positive electrode active material layer comprises a positive electrode active material particle comprising:

- a first region comprising a first transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal, a representative element, and fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Liu does not explicitly disclose:

- wherein an angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image.

Sugie discloses positive electrode active material particle where an angle between a bright line in a first region and a bright line in a second region is less than or equal to 5° in a transmission electron microscope image (Fig. 4, [0253]) to improve the structural stability of the positive electrode active material particle (see structural stability, [0043]). Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the positive electrode active material particle of Liu with the angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image as taught by Sugie in order to improve the structural stability of the positive electrode active material particle.

Regarding **claim 64**, Liu discloses a positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising a positive electrode active material particle comprising:

- a first region comprising a first transition metal and aluminum (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle ([0010]-[0012]).

Liu does not explicitly disclose:

- wherein an angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image.

Sugie discloses positive electrode active material particle where an angle between a bright line in a first region and a bright line in a second region is less than or equal to 5° in a transmission electron microscope image (Fig. 4, [0253]) to improve the structural stability of the positive electrode active material particle (see structural stability, [0043]). Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the positive electrode active material particle of Liu with the angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image as taught by Sugie in order to improve the structural stability of the positive electrode active material particle.

Regarding **claim 65**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region covers at least a part of the first region ([0010]-[0012]).

Regarding **claim 66**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is provided in a range from a surface of the positive electrode active material particle to a depth of 20 nm ([0010]-[0012]).

Regarding **claim 67**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the first transition metal is cobalt (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]), and

- wherein the second transition metal is titanium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 68**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region further comprises fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 69**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region further comprises fluorine and a representative element (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 70**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the representative element is magnesium (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Regarding **claim 71**, modified Liu discloses all claim limitations set forth above and further discloses a positive electrode active material layer:

- wherein the second region is positioned closer to a surface of the positive electrode active material particle than the first region is ([0010]-[0012]).

Regarding **claim 72**, Liu discloses an electronic device comprising a lithium-ion secondary battery, the lithium-ion secondary battery comprising a positive electrode comprising a positive electrode active material layer; a negative electrode; and an electrolyte solution, wherein the positive electrode active material layer comprises:

- a first region comprising a first transition metal and aluminum (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]); and
- a second region comprising a second transition metal (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]),
- wherein the second region is in a superficial portion of the positive electrode active material particle ([0010]-[0012]).

Liu does not explicitly disclose:

- wherein an angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image.

Sugie discloses positive electrode active material particle where an angle between a bright line in a first region and a bright line in a second region is less than or equal to 5° in a transmission electron microscope image (Fig. 4, [0253]) to improve the structural stability of the positive electrode active material particle (see structural stability, [0043]). Therefore, it would have been obvious to one of ordinary skill in the art at the effective filing date of the invention to make the positive electrode active material particle of Liu with the angle between a bright line in the first region and a bright line in the second region is less than or equal to 5° in a transmission electron microscope image as taught by Sugie in order to improve the structural stability of the positive electrode active material particle.

Allowable Subject Matter

9. **Claim(s) 56-63** is/are allowed.

10. The following is a statement of reasons for the indication of allowable subject matter:

The closest prior art of record is Liu (CN 10210806 A, see machine translation).

Liu discloses a positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising a positive electrode active material particle comprising a first transition metal, a second transition metal, a representative element, and fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Liu does not disclose, teach, or suggest the following distinguishing feature(s):

A positive electrode active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising a positive electrode active material particle comprising a representative element and fluorine present in a portion that includes crystal defects or a crack portion in the positive electrode active material particle.

Response to Arguments

11. Applicant's arguments with respect to **claim(s) 35-58 and 64-72** have been considered but are moot because the arguments do not apply to any of the references being used in the current rejection.

Conclusion

12. Applicant's amendment necessitated the new ground(s) of rejection presented in this Office action. Accordingly, **THIS ACTION IS MADE FINAL**. See MPEP § 706.07(a). Applicant is reminded of the extension of time policy as set forth in 37 CFR 1.136(a).

A shortened statutory period for reply to this final action is set to expire THREE MONTHS from the mailing date of this action. In the event a first reply is filed within TWO MONTHS of the mailing date of this final action and the advisory action is not mailed until after

the end of the THREE-MONTH shortened statutory period, then the shortened statutory period will expire on the date the advisory action is mailed, and any extension fee pursuant to 37 CFR 1.136(a) will be calculated from the mailing date of the advisory action. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the date of this final action.

13. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Sean P Cullen, Ph.D. whose telephone number is (571)270-1251. The examiner can normally be reached on Monday-Thursday 6:00 am-4:00 pm CDT, Friday 6:00 am-12:00 pm CDT.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Basia A Ridley can be reached on (571)272-1453. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would

like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

Notice of References Cited	Application/Control No. 15/638,449	Applicant(s)/Patent Under Reexamination OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	CPC Classification	US Classification
*	A	US-20150357641-A1	12-2015	SUGIE; Tsukasa	H01M4/505	429/223
	B					
	C					
	D					
	E					
	F					
	G					
	H					
	I					
	J					
	K					
	L					
	M					

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	CPC Classification
	N	CN-102610806-A	07-2012	CN	NA LIU	
	O					
	P					
	Q					
	R					
	S					
	T					

NON-PATENT DOCUMENTS

*		Include as applicable: Author, Title Date, Publisher, Edition or Volume, Pertinent Pages)
	U	
	V	
	W	
	X	

*A copy of this reference is not being furnished with this Office action. (See MPEP § 707.05(a).)
Dates in MM-YYYY format are publication dates. Classifications may be US or foreign.

Index of Claims	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

✓	Rejected	-	Cancelled	N	Non-Elected	A	Appeal
=	Allowed	÷	Restricted	I	Interference	O	Objected

CLAIMS

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIM		DATE								
Final	Original	12/20/2018	03/07/2019	06/27/2019	12/04/2019	02/25/2020				
	1	÷	✓	✓	✓	-				
	2	÷	✓	✓	-	-				
	3	÷	✓	✓	✓	-				
	4	÷	✓	✓	✓	-				
	5	÷	✓	✓	✓	-				
	6	÷	✓	✓	✓	-				
	7	÷	✓	-	-	-				
	8	÷	✓	✓	✓	-				
	9	÷	✓	✓	✓	-				
	10	÷	✓	✓	✓	-				
	11	÷	✓	✓	✓	-				
	12	÷	✓	✓	-	-				
	13	÷	✓	✓	-	-				
	14	÷	✓	✓	-	-				
	15	÷	✓	✓	-	-				
	16	÷	✓	✓	-	-				
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	20	÷	-	-	-	-				
	21	÷	-	-	-	-				
	22	÷	-	-	-	-				
	23				✓	-				
	24				✓	-				
	25				✓	-				
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	27				✓	-				
	28				✓	-				
	29				✓	-				
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	34				✓	-				
	35					✓				
	36					✓				
	37					✓				
	38					✓				
	39					✓				
	40					✓				
	41					✓				
	42					✓				

Index of Claims	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

CLAIM		DATE								
Final	Original	12/20/2018	03/07/2019	06/27/2019	12/04/2019	02/25/2020				
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	87					✓				
	88					✓				
	89					✓				
	90					✓				

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

CPC - Searched*		
Symbol	Date	Examiner
H01M4/131, 1315, 1391, 13915, 625, 8657	03/07/2019	SPC
Updated CPC search	06/27/2019	SPC
Updated CPC search	12/04/2019	SPC
Updated CPC search	02/25/2020	SPC

CPC Combination Sets - Searched*		
Symbol	Date	Examiner

US Classification - Searched*			
Class	Subclass	Date	Examiner
429	231.3	03/07/2019	SPC
	Updated USPC search	06/27/2019	SPC
	Updated USPC search	12/04/2019	SPC
	Updated USPC search	02/25/2020	SPC

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

Search Notes		
Search Notes	**Date**	**Examiner**
H01M4/131, 1315, 1391, 13915, 625, 8657	03/07/2019	SPC
429/231.3	03/07/2019	SPC
Inventor/applicant search, PALM	03/07/2019	SPC
EAST text search, see search history	03/07/2019	SPC
Updated inventor/applicant search, PALM	06/27/2019	SPC
Updated EAST text search, see search history	06/27/2019	SPC
Updated inventor/applicant search, PALM	12/04/2019	SPC
Updated EAST text search, see search history	12/04/2019	SPC
Updated inventor/applicant search, PALM	02/25/2020	SPC
Updated EAST text search, see search history	02/25/2020	SPC

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
S116	3	(H01M.cpcl. H01M.cicl.) and (bright adj line with (TEM transmission electron))	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/21 10:59
S117	365	(H01M.cpcl. H01M.cicl.) and (crystal adj defect)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/21 11:05
S118	57	(H01M.cpcl. H01M.cicl.) and (crack adj portion)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/21 11:06
S119	78	LiCoTiMgAlOF$ LiCoTiAlMgOF$ LiCoMgTiAlOF$ LiCoMgAlTiOF$ LiCoAlTiMgOF$ LiCoAlMgTiOF$ LiCoTiMgAlO LiCoTiAlMgO LiCoMgTiAlO LiCoMgAlTiO LiCoAlTiMgO LiCoAlMgTiO	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/24 16:02
S120	6	LiCoTiMgAlOF$ LiCoTiAlMgOF$ LiCoMgTiAlOF$ LiCoMgAlTiOF$ LiCoAlTiMgOF$ LiCoAlMgTiOF$	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/24 16:09
S121	11	S119 and xps	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/24 16:10
S123	0	S122 and xds	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 08:54

S122	78	LiCoTiMgAlOF$ LiCoTiAlMgOF$ LiCoMgTiAlOF$ LiCoMgAlTiOF$ LiCoAlTiMgOF$ LiCoAlMgTiOF$ LiCoTiMgAlO LiCoTiAlMgO LiCoMgTiAlO LiCoMgAlTiO LiCoAlTiMgO LiCoAlMgTiO	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 08:54
S125	0	S122 and transmission adj electron	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 09:11
S124	0	S122 and TEM	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 09:11
S127	0	S122 and crack	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 09:12
S126	0	S122 and crystal adj defect	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 09:12
		S122 and energy adj dispersive	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 09:13
S130	1809	S129 and (xds TEM transmission adj electron crystal adj defect crack energy adj dispersive)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 09:14
S129	17923	LiCoOF LiCoAlO	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 09:14
S131	1649	S130 and (H01M.cpcl. H01M.cicl.)	US-PGPUB; USPAT; USOCR;	OR	ON	2020/02/25 09:21

			FPRS; EPO; JPO; DERWENT; IBM_TDB			
S133	65	(LiCoOF LiCoAlO) same (xds TEM transmission adj electron crystal adj defect crack energy adj dispersive)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 09:23
S134	219	(LiCoOF LiCoAlO) same (edx eds TEM transmission adj electron crystal adj defect crack energy adj dispersive)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 10:05
S136	72	(LiCoOF LiCoAlO) same (edx eds! TEM transmission adj electron crystal adj defect crack energy adj dispersive)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 10:06
S135	17	S122 and (eds edx)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 10:06
S137	18	S122 and (photoelectron esca xps)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 10:24
S138	2	(LiCoOF LiCoAlO) same (crystal adj defect)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 11:15
S139	2	(LiCoOF LiCoAlO) same (crystal near4 defect)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 11:16
S140	0	S122 and (crystal near4 defect)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 11:17
L1	0	(LiCoOF) same crystal near2 defect	US-PGPUB;	OR	ON	2020/02/25

			USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB			13:15
L3	24	(LiCoOF) and crystal near2 defect	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 13:16
L2	109	(US-20180013130-$ or US-20120258358-$ or US-20100233542-$ or US-20110300441-$ or US-20140227606-$ or US-20100173197-$ or US-20030211391-$ or US-20030082448-$ or US-20160104880-$ or US-20040229123-$ or US-20110287321-$ or US-20160149215-$ or US-20030087155-$ or US-20160372739-$ or US-20080131778-$ or US-20160308202-$ or US-20160056457-$ or US-20110042610-$ or US-20070224506-$ or US-20020076613-$ or US-20020071991-$ or US-20160079597-$ or US-20160006032-$ or US-20120225357-$ or US-20100301284-$ or US-20100181540-$).did. or (US-20120135315-$ or US-20110020708-$ or US-20170162917-$ or US-20190221845-$ or US-20190221837-$ or US-20180079655-$ or US-20180006302-$ or US-20180366723-$ or US-20170207442-$ or US-20180248179-$ or US-20180212237-$ or US-20180053940-$ or US-20140099549-$ or US-20150255789-$ or US-20090202892-$ or US-20090081549-$ or US-20080138710-$ or US-20150340689-$ or US-20150104708-$ or US-20130183579-$ or US-20170040607-$ or US-20180366726-$ or US-20180145368-$ or US-20180102536-$ or US-20170358797-$ or US-20140315090-$ or US-20130029842-$).did. or (US-20110269023-$ or US-20170207444-$ or US-20170187035-$ or US-20180145317-$ or US-20170005334-$ or US-20160072124-$ or US-20130230770-$ or US-20120321948-$ or US-20120308896-$ or US-20110262796-$ or US-20170301908-$ or US-20160351898-$ or US-20160240842-$ or US-20160006026-$ or US-20150311522-$ or US-20080102369-$ or US-20080193846-$ or US-20110052980-$ or US-20080131780-$ or US-20080118847-$ or US-20080118839-$ or US-20070264573-$ or US-20070254214-$ or US-20070224505-$ or US-20070224504-$ or US-20070117014-$ or US-20170200951-$).did. or (US-20180366728-$ or US-20150357641-$ or US-20170309891-$ or US-20160056469-	US-PGPUB; USPAT; FPRS; JPO; DERWENT	OR	ON	2020/02/25 13:16

		$ or US-20140087262-$ or US-20120104311-$ or US-20080118829-$).did. or (US-8927148-$ or US-8163198-$).did. or (WO-2012005180-$ or WO-2014032405-$ or KR-20060085085-$ or WO-2011016553-$ or CN-103280576-$).did. or (JP-2004342554-$ or JP-2004103566-$ or JP-2003331846-$ or JP-2002151078-$ or JP-2012113823-$ or JP-2010118312-$ or JP-2009218198-$ or JP-2017033641-$ or JP-2008016243-$ or JP-2002164053-$).did. or (WO-2018207049-$ or CN-102610806-$ or JP-2008016267-$ or JP-2007145695-$).did.				
L5	13	l2 and crystal near2 defect	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 13:17
L6	2	JP-2004087487-$.did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 13:19
L7	2	US-20080032199-$.did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 13:20
L8	260	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) with (Mg magnesium fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 13:25
L9	2	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) with (Mg magnesium) with (fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 13:54
L10	49	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) same (Mg magnesium) same (fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 13:55
L11	158	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) with (fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO;	OR	ON	2020/02/25 14:10

			DERWENT; IBM_TDB			
L12	42	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) with (fluorine)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 14:11
L14	25	l2 and (crack crystal near2 defect)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 14:14
L16	2	(H01M.cpcl. H01M.cicl.) and (breakage defect) with (Mg magnesium) with (fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 14:24
L18	41	l17 not l10	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 14:25
L17	48	(H01M.cpcl. H01M.cicl.) and (breakage defect) same (Mg magnesium) same (fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/02/25 14:25

EAST Search History (Interference)

<This search history is empty>

2/25/2020 2:42:22 PM
C:\Users\scullen\Documents\EAST\Workspaces\15638449.wsp

PTO/AIA/80 (07-12)
Approved for use through 11/30/2014. OMB 0651-0035
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POWER OF ATTORNEY TO PROSECUTE APPLICATIONS BEFORE THE USPTO

I hereby revoke all previous powers of attorney given in the application identified in the attached statement under 37 CFR 3.73(c).

I hereby appoint:

☑ Practitioners associated with Customer Number: 26171

OR

☐ Practitioner(s) named below (if more than ten patent practitioners are to be named, then a customer number must be used):

Name	Registration Number	Name	Registration Number

As attorney(s) or agent(s) to represent the undersigned before the United States Patent and Trademark Office (USPTO) in connection with any and all patent applications a ssigned only to the undersigned according to the USPTO assignment records or assignments docu ments attached to this form in accordance with 37 CFR 3.73(c).

Please change the correspondence address for the application identified in the attached statement under 37 CFR 3.73(c) to:

☑ The address associated with Customer Number: 26171

OR

☐	Firm or Individual Name	
	Address	
	City	
	Country	
	Telephone	Email

Assignee Name and Address: SEMICONDUCTOR ENERGY LABORATORY CO., LTD.
398, HASE, ATSUGI-SHI
KANAGAWA-KEN 243-0036
JAPAN

A copy of this form, together with a statement under 37 CFR 3.73(c) (Form PTO/SB/96 or equivalent) is required to be Filed in each application in which this form is used. The statement under 37 CFR 3.73(c) may be completed by one of The practitioners appointed in this form, and must identify the application in which this Power of Attorney is to be filed.

SIGNATURE of Assignee of Record
The individual whose signature and title is supplied below is authorized to act on behalf of the assignee

Signature	Shunpei Yamazaki	Date	09/25/2012
Name	Dr. Shunpei Yamazaki	Telephone	81-46-270-1170
Title	President		

This collection of information is required by 37 CFR 1.31, 1.32 and 1.33. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 3 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

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The information provided by you in this form will be subject to the following routine uses:

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2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (*i.e.*, GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspection or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

PTO/AIA/96 (08-12)
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STATEMENT UNDER 37 CFR 3.73(c)

Applicant/Patent Owner: SEMICONDUCTOR ENERGY LABORATORY CO., LTD.

Application No./Patent No.: 15/638,449 Filed/Issue Date: June 30, 2017

Titled: POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY

Semiconductor Energy Laboratory Co., Ltd., a corporation

(Name of Assignee) (Type of Assignee, e.g., corporation, partnership, university, government agency, etc.)

states that, for the patent application/patent identified above, it is (choose **one** of the option 1, 2, 3 or 4 below):

1. ☒ The assignee of the entire right, title, and interest.

2. ☐ An assignee of less than the entire right, title and interest (check applicable box):

☐ The extent (by percentage) of its ownership interest is _____. Additional Statement(s) by the owners holding the balance of the interest must be submitted to account for 100% of the ownership interest.

☐ There are unspecified percentages of ownership. The other parties, including inventors, who together own the entire right, title and interest are:

Additional Statement(s) by the owner(s) holding the balance of the interest must be submitted to account for the entire right, title, and interest.

3. ☐ The assignee of an undivided interest in the entirety (a complete assignment from one of the joint inventors was made). The other parties, including inventors, who together own the entire right, title, and interest are:

Additional Statement(s) by the owner(s) holding the balance of the interest must be submitted to account for the entire right, title, and interest.

4. ☐ The recipient, via a court proceeding or the like (e.g., bankruptcy, probate), of an undivided interest in the entirety (a complete transfer of ownership interest was made). The certified document(s) showing the transfer is attached.

The interest identified in option 1, 2 or 3 above (not option 4) is evidenced by either (choose **one** of the options A or B below):

A. ☒ An assignment from the inventor(s) of the patent application/patent identified above. The assignment was recorded in the United States Patent and Trademark Office at Reel 044179, Frame 0782, or for which a copy thereof is attached.

B. ☐ A chain of title from the inventor(s), of the patent application/patent identified above, to the current assignee as follows:

1. From: _____ To: _____

The document was recorded in the United States Patent and Trademark Office at

Reel _____, Frame _____, or for which a copy thereof is attached.

2. From: _____ To: _____

The document was recorded in the United States Patent and Trademark Office at

Reel _____, Frame _____, or for which a copy thereof is attached.

This collection of information is required by 37 CFR 3.73(b). The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

Approved for use through 01/31/2013. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
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STATEMENT UNDER 37 CFR 3.73(c)

3. From: _____ To: _____

The document was recorded in the United States Patent and Trademark Office at

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4. From: _____ To: _____

The document was recorded in the United States Patent and Trademark Office at

Reel _____, Frame _____, or for which a copy thereof is attached.

5. From: _____ To: _____

The document was recorded in the United States Patent and Trademark Office at

Reel _____, Frame _____, or for which a copy thereof is attached.

6. From: _____ To: _____

The document was recorded in the United States Patent and Trademark Office at

Reel _____, Frame _____, or for which a copy thereof is attached.

☐ Additional documents in the chain of title are listed on a supplemental sheet(s).

☒ As required by 37 CFR 3.73(c)(1)(i), the documentary evidence of the chain of title from the original owner to the assignee was, or concurrently is being, submitted for recordation pursuant to 37 CFR 3.11.

[NOTE: A separate copy *(i.e.,* a true copy of the original assignment document(s)) must be submitted to Assignment Division in accordance with 37 CFR Part 3, to record the assignment in the records of the USPTO. See MPEP 302.08]

The undersigned (whose title is supplied below) is authorized to act on behalf of the assignee.

/John F. Hayden/	March 12, 2020
Signature	Date
John F. Hayden	37640
Printed or Typed Name	Title or Registration Number

Electronic Acknowledgement Receipt

EFS ID:	38842707
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	22204
Filer:	John F. Hayden/April Saunders-Fuller
Filer Authorized By:	John F. Hayden
Attorney Docket Number:	740756-004563
Receipt Date:	12-MAR-2020
Filing Date:	30-JUN-2017
Time Stamp:	10:01:19
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Power of Attorney	POA.pdf	155302 c3fa4e2f8a13691f69e577b72f8ef9817f89ca32	no	2

Warnings:

Information:					
2	Assignee showing of ownership per 37 CFR 3.73	12732-2003001-373.pdf	95750 a1b430001c4cb7dc4e668b1b4960aae66100 85527	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	251052		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
15/638,449	06/30/2017	Teruaki OCHIAI	740756-004563

CONFIRMATION NO. 1074

26171
FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

POA ACCEPTANCE LETTER

OC000000115466438

Date Mailed: 03/17/2020

NOTICE OF ACCEPTANCE OF POWER OF ATTORNEY

This is in response to the Power of Attorney filed 03/12/2020.

The Power of Attorney in this application is accepted. Correspondence in this application will be mailed to the above address as provided by 37 CFR 1.33.

Questions about the contents of this notice and the requirements it sets forth should be directed to the Office of Data Management, Application Assistance Unit, at **(571) 272-4000** or **(571) 272-4200** or **1-888-786-0101**.

/zretta/

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
15/638,449	06/30/2017	Teruaki OCHIAI	740756-004563

CONFIRMATION NO. 1074

POWER OF ATTORNEY NOTICE

22204
NIXON PEABODY, LLP
799 Ninth Street, NW
SUITE 500
WASHINGTON, DC 20001

OC000000115466399

Date Mailed: 03/17/2020

NOTICE REGARDING CHANGE OF POWER OF ATTORNEY

This is in response to the Power of Attorney filed 03/12/2020.

- The Power of Attorney to you in this application has been revoked by the applicant. Future correspondence will be mailed to the new address of record(37 CFR 1.33).

Questions about the contents of this notice and the requirements it sets forth should be directed to the Office of Data Management, Application Assistance Unit, at **(571) 272-4000** or **(571) 272-4200** or **1-888-786-0101**.

/zretta/

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor	:	Teruaki OCHIAI	Art Unit	:	1725
Application No.	:	15/638,449	Examiner	:	Sean P. Cullen
Filed	:	June 30, 2017	Conf. No.	:	1074
Title	:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY			

MAIL STOP AF
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

AMENDMENT IN REPLY TO ACTION OF MARCH 2, 2020

Please consider the following reply.

Amendments to the Claims are reflected in the listing of claims which begins on page 2 of this paper.

Remarks/Arguments begin on page 7 of this paper.

AMENDMENTS TO THE CLAIMS

This listing of claims replaces all prior versions and listings of claims in the application.

Listing of Claims

1-55. (Canceled)

56. (Currently Amended) A ~~positive electrode active material layer for a~~ lithium-ion secondary battery[[, the]] comprising a positive electrode active material layer comprising:

a positive electrode active material particle comprising a first transition metal, a second transition metal, a representative element, and fluorine,

wherein the representative element and the fluorine are present in a portion which includes crystal defects observed from a TEM image in the positive electrode active material particle.

57. (Currently Amended) The ~~positive electrode active material layer~~ lithium-ion secondary battery according to claim 56,

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

58. (Currently Amended) The ~~positive electrode active material layer~~ lithium-ion secondary battery according to claim 56,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

59. (Currently Amended) The ~~positive electrode active material layer~~ lithium-ion secondary battery according to claim 56,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

60. (Currently Amended) A ~~positive electrode active material layer for a~~ lithium-ion secondary battery[[, the]] comprising a positive electrode active material layer comprising:

a positive electrode active material particle comprising a first transition metal, a second transition metal, a representative element, and fluorine,

wherein the representative element and the fluorine are present in a crack portion observed from a TEM image in the positive electrode active material particle.

61. (Currently Amended) The ~~positive electrode active material layer~~ lithium-ion secondary battery according to claim 60,

wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

62. (Currently Amended) The ~~positive electrode active material layer~~ lithium-ion secondary battery according to claim 60,

wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

63. (Currently Amended) The ~~positive electrode active material layer~~ lithium-ion secondary battery according to claim 60,

wherein the first transition metal is cobalt,

wherein the second transition metal is titanium, and

wherein the representative element is magnesium.

64-90. (Canceled)

REMARKS

Claims 56-63 are pending in the application, with claims 56 and 60 being independent. Claims 56-63 have been amended and claims 35-55 and 64-90 have been cancelled. Support for the amendments to claims 56 and 60 is found throughout the application and drawings as filed, including paragraph [0057] and FIG. 1B. Claims 57-59 and 61-63 have been amended for consistency with claims 56 and 60, respectively. No new matter has been introduced.

Claim Rejections

Claims 35-41, 44, and 64-72 were rejected under 35 U.S.C. § 112(b). Claims 42-55 and 73-90 were rejected under 35 U.S.C. § 102(a)(1) over CN 102610806 (Liu). Claims 35-41 and 64-72 were rejected under 35 U.S.C. § 103 over Liu in view of US 2015/0357641 (Sugie). As noted above, however, claims 35-55 and 64-90 have been cancelled. As such, these rejections are now moot.

Allowable Subject Matter

Applicant thanks Examiner Cullen for acknowledging the allowability of claims 56-63. Claims 56 and 60 have been amended for clarification to recite "A lithium-ion secondary battery comprising a positive electrode active material layer" and to recite "observed from a TEM image."

All claims are in condition for allowance. No fees are believed to be due. Apply any additional necessary charges or credits to Deposit Account 06-1050, referencing attorney docket number 12732-2003001.

Respectfully submitted,

Date: May 11, 2020

/Heather L. Flanagan/
Heather L. Flanagan
Reg. No. 54,101

Customer Number 26171
Fish & Richardson P.C.
Telephone: (202) 783-5070
Facsimile: (877) 769-7945
41447353.doc

Electronic Acknowledgement Receipt

EFS ID:	39405429
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	26171
Filer:	Heather L. Flanagan/Sharon Allen
Filer Authorized By:	Heather L. Flanagan
Attorney Docket Number:	12732-2003001
Receipt Date:	11-MAY-2020
Filing Date:	30-JUN-2017
Time Stamp:	15:14:58
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		12732-2003001-Final-Response.pdf	112354 266ac6731c591b370fc9584e6c2909281fec3029	yes	6

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Response After Final Action	1	1
	Claims	2	4
	Applicant Arguments/Remarks Made in an Amendment	5	6
Warnings:			
Information:			
	Total Files Size (in bytes):	112354	

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

PTO/SB/06 (09-11)
Approved for use through 1/31/2014. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 15/638,449	Filing Date 06/30/2017	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $80 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $420 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	05/11/2020	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 8	Minus	** 20	= 0		x $100 =	0
	Independent (37 CFR 1.16(h))	* 2	Minus	*** 3	= 0		x $460 =	0
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	0

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	LIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/BRUCE D HARRISON/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
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NOTICE OF ALLOWANCE AND FEE(S) DUE

26171 7590 05/20/2020

FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

EXAMINER	
CULLEN, SEAN P	
ART UNIT	PAPER NUMBER
1725	

DATE MAILED: 05/20/2020

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	12732-2003001	1074

TITLE OF INVENTION: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1000	$0.00	$0.00	$1000	08/20/2020

THE APPLICATION IDENTIFIED ABOVE HAS BEEN EXAMINED AND IS ALLOWED FOR ISSUANCE AS A PATENT. PROSECUTION ON THE MERITS IS CLOSED. THIS NOTICE OF ALLOWANCE IS NOT A GRANT OF PATENT RIGHTS. THIS APPLICATION IS SUBJECT TO WITHDRAWAL FROM ISSUE AT THE INITIATIVE OF THE OFFICE OR UPON PETITION BY THE APPLICANT. SEE 37 CFR 1.313 AND MPEP 1308.

THE ISSUE FEE AND PUBLICATION FEE (IF REQUIRED) MUST BE PAID WITHIN THREE MONTHS FROM THE MAILING DATE OF THIS NOTICE OR THIS APPLICATION SHALL BE REGARDED AS ABANDONED. THIS STATUTORY PERIOD CANNOT BE EXTENDED. SEE 35 U.S.C. 151. THE ISSUE FEE DUE INDICATED ABOVE DOES NOT REFLECT A CREDIT FOR ANY PREVIOUSLY PAID ISSUE FEE IN THIS APPLICATION. IF AN ISSUE FEE HAS PREVIOUSLY BEEN PAID IN THIS APPLICATION (AS SHOWN ABOVE), THE RETURN OF PART B OF THIS FORM WILL BE CONSIDERED A REQUEST TO REAPPLY THE PREVIOUSLY PAID ISSUE FEE TOWARD THE ISSUE FEE NOW DUE.

HOW TO REPLY TO THIS NOTICE:

I. Review the ENTITY STATUS shown above. If the ENTITY STATUS is shown as SMALL or MICRO, verify whether entitlement to that entity status still applies.

If the ENTITY STATUS is the same as shown above, pay the TOTAL FEE(S) DUE shown above.

If the ENTITY STATUS is changed from that shown above, on PART B - FEE(S) TRANSMITTAL, complete section number 5 titled "Change in Entity Status (from status indicated above)".

For purposes of this notice, small entity fees are 1/2 the amount of undiscounted fees, and micro entity fees are 1/2 the amount of small entity fees.

II. PART B - FEE(S) TRANSMITTAL, or its equivalent, must be completed and returned to the United States Patent and Trademark Office (USPTO) with your ISSUE FEE and PUBLICATION FEE (if required). If you are charging the fee(s) to your deposit account, section "4b" of Part B - Fee(s) Transmittal should be completed and an extra copy of the form should be submitted. If an equivalent of Part B is filed, a request to reapply a previously paid issue fee must be clearly made, and delays in processing may occur due to the difficulty in recognizing the paper as an equivalent of Part B.

III. All communications regarding this application must give the application number. Please direct all communications prior to issuance to Mail Stop ISSUE FEE unless advised to the contrary.

IMPORTANT REMINDER: Maintenance fees are due in utility patents issuing on applications filed on or after Dec. 12, 1980. It is patentee's responsibility to ensure timely payment of maintenance fees when due. More information is available at www.uspto.gov/PatentMaintenanceFees.

PART B - FEE(S) TRANSMITTAL

Complete and send this form, together with applicable fee(s), by mail or fax, or via EFS-Web.

By mail, send to: Mail Stop ISSUE FEE
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

By fax, send to: (571)-273-2885

INSTRUCTIONS: This form should be used for transmitting the ISSUE FEE and PUBLICATION FEE (if required). Blocks 1 through 5 should be completed where appropriate. All further correspondence including the Patent, advance orders and notification of maintenance fees will be mailed to the current correspondence address as indicated unless corrected below or directed otherwise in Block 1, by (a) specifying a new correspondence address; and/or (b) indicating a separate "FEE ADDRESS" for maintenance fee notifications.

CURRENT CORRESPONDENCE ADDRESS (Note: Use Block 1 for any change of address)

26171 7590 05/20/2020
FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

Note: A certificate of mailing can only be used for domestic mailings of the Fee(s) Transmittal. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawing, must have its own certificate of mailing or transmission.

Certificate of Mailing or Transmission

I hereby certify that this Fee(s) Transmittal is being deposited with the United States Postal Service with sufficient postage for first class mail in an envelope addressed to the Mail Stop ISSUE FEE address above, or being transmitted to the USPTO via EFS-Web or by facsimile to (571) 273-2885, on the date below.

(Typed or printed name)

(Signature)

(Date)

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	12732-2003001	1074

TITLE OF INVENTION: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1000	$0.00	$0.00	$1000	08/20/2020

EXAMINER	ART UNIT	CLASS-SUBCLASS
CULLEN, SEAN P	1725	429-488000

1. Change of correspondence address or indication of "Fee Address" (37 CFR 1.363).

❑ Change of correspondence address (or Change of Correspondence Address form PTO/SB/122) attached.

❑ "Fee Address" indication (or "Fee Address" Indication form PTO/SB/47; Rev 03-09 or more recent) attached. **Use of a Customer Number is required.**

2. For printing on the patent front page, list
(1) The names of up to 3 registered patent attorneys or agents OR, alternatively,
(2) The name of a single firm (having as a member a registered attorney or agent) and the names of up to 2 registered patent attorneys or agents. If no name is listed, no name will be printed.

1_____
2_____
3_____

3. ASSIGNEE NAME AND RESIDENCE DATA TO BE PRINTED ON THE PATENT (print or type)

PLEASE NOTE: Unless an assignee is identified below, no assignee data will appear on the patent. If an assignee is identified below, the document must have been previously recorded, or filed for recordation, as set forth in 37 CFR 3.11 and 37 CFR 3.81(a). Completion of this form is NOT a substitute for filing an assignment.

(A) NAME OF ASSIGNEE

(B) RESIDENCE: (CITY and STATE OR COUNTRY)

Please check the appropriate assignee category or categories (will not be printed on the patent) : ❑ Individual ❑ Corporation or other private group entity ❑ Government

4a. Fees submitted: ❑Issue Fee ❑Publication Fee (if required) ❑Advance Order - # of Copies _____

4b. Method of Payment: *(Please first reapply any previously paid fee shown above)*

❑ Electronic Payment via EFS-Web ❑ Enclosed check ❑ Non-electronic payment by credit card (Attach form PTO-2038)

❑ The Director is hereby authorized to charge the required fee(s), any deficiency, or credit any overpayment to Deposit Account No. _____

5. **Change in Entity Status** (from status indicated above)

❑ Applicant certifying micro entity status. See 37 CFR 1.29

❑ Applicant asserting small entity status. See 37 CFR 1.27

❑ Applicant changing to regular undiscounted fee status.

NOTE: Absent a valid certification of Micro Entity Status (see forms PTO/SB/15A and 15B), issue fee payment in the micro entity amount will not be accepted at the risk of application abandonment.
NOTE: If the application was previously under micro entity status, checking this box will be taken to be a notification of loss of entitlement to micro entity status.
NOTE: Checking this box will be taken to be a notification of loss of entitlement to small or micro entity status, as applicable.

NOTE: This form must be signed in accordance with 37 CFR 1.31 and 1.33. See 37 CFR 1.4 for signature requirements and certifications.

Authorized Signature _____ Date _____

Typed or printed name _____ Registration No. _____

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	12732-2003001	1074

26171 7590 05/20/2020
FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

EXAMINER	
CULLEN, SEAN P	
ART UNIT	PAPER NUMBER
1725	

DATE MAILED: 05/20/2020

Determination of Patent Term Adjustment under 35 U.S.C. 154 (b)
(Applications filed on or after May 29, 2000)

The Office has discontinued providing a Patent Term Adjustment (PTA) calculation with the Notice of Allowance.

Section 1(h)(2) of the AIA Technical Corrections Act amended 35 U.S.C. 154(b)(3)(B)(i) to eliminate the requirement that the Office provide a patent term adjustment determination with the notice of allowance. See Revisions to Patent Term Adjustment, 78 Fed. Reg. 19416, 19417 (Apr. 1, 2013). Therefore, the Office is no longer providing an initial patent term adjustment determination with the notice of allowance. The Office will continue to provide a patent term adjustment determination with the Issue Notification Letter that is mailed to applicant approximately three weeks prior to the issue date of the patent, and will include the patent term adjustment on the patent. Any request for reconsideration of the patent term adjustment determination (or reinstatement of patent term adjustment) should follow the process outlined in 37 CFR 1.705.

Any questions regarding the Patent Term Extension or Adjustment determination should be directed to the Office of Patent Legal Administration at (571)-272-7702. Questions relating to issue and publication fee payments should be directed to the Customer Service Center of the Office of Patent Publication at 1-(888)-786-0101 or (571)-272-4200.

OMB Clearance and PRA Burden Statement for PTOL-85 Part B

The Paperwork Reduction Act (PRA) of 1995 requires Federal agencies to obtain Office of Management and Budget approval before requesting most types of information from the public. When OMB approves an agency request to collect information from the public, OMB (i) provides a valid OMB Control Number and expiration date for the agency to display on the instrument that will be used to collect the information and (ii) requires the agency to inform the public about the OMB Control Number's legal significance in accordance with 5 CFR 1320.5(b).

The information collected by PTOL-85 Part B is required by 37 CFR 1.311. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 30 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, Virginia 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, Virginia 22313-1450. Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether disclosure of these records is required by the Freedom of Information Act.
2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspection or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

Notice of Allowability	**Application No.** 15/638,449	**Applicant(s)** OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	**Art Unit** 1725	**AIA (FITF) Status** Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address--

All claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice of Allowance (PTOL-85) or other appropriate communication will be mailed in due course. **THIS NOTICE OF ALLOWABILITY IS NOT A GRANT OF PATENT RIGHTS.** This application is subject to withdrawal from issue at the initiative of the Office or upon petition by the applicant. See 37 CFR 1.313 and MPEP 1308.

1. ☑ This communication is responsive to the communication(s) filed on 11 May 2020.
   ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2. ☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

3. ☑ The allowed claim(s) is/are 56-63 . As a result of the allowed claim(s), you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see **http://www.uspto.gov/patents/init_events/pph/index.jsp** or send an inquiry to **PPHfeedback@uspto.gov.**

4. ☑ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

   Certified copies:

   a) ☑ All b) ☐ Some *c) ☐ None of the:

   1. ☑ Certified copies of the priority documents have been received.
   2. ☐ Certified copies of the priority documents have been received in Application No. _____ .
   3. ☐ Copies of the certified copies of the priority documents have been received in this national stage application from the International Bureau (PCT Rule 17.2(a)).

   * Certified copies not received: _____ .

Applicant has THREE MONTHS FROM THE "MAILING DATE" of this communication to file a reply complying with the requirements noted below. Failure to timely comply will result in ABANDONMENT of this application.
THIS THREE-MONTH PERIOD IS NOT EXTENDABLE.

5. ☐ CORRECTED DRAWINGS (as "replacement sheets") must be submitted.
   ☐ including changes required by the attached Examiner's Amendment / Comment or in the Office action of Paper No./Mail Date _____ .

   Identifying indicia such as the application number (see 37 CFR 1.84(c)) should be written on the drawings in the front (not the back) of each sheet. Replacement sheet(s) should be labeled as such in the header according to 37 CFR 1.121(d).

6. ☐ DEPOSIT OF and/or INFORMATION about the deposit of BIOLOGICAL MATERIAL must be submitted. Note the attached Examiner's comment regarding REQUIREMENT FOR THE DEPOSIT OF BIOLOGICAL MATERIAL.

Attachment(s)

1. ☐ Notice of References Cited (PTO-892)
2. ☐ Information Disclosure Statements (PTO/SB/08), Paper No./Mail Date _____.
3. ☐ Examiner's Comment Regarding Requirement for Deposit of Biological Material _____.
4. ☐ Interview Summary (PTO-413), Paper No./Mail Date. _____.
5. ☐ Examiner's Amendment/Comment
6. ☑ Examiner's Statement of Reasons for Allowance
7. ☐ Other _____.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

DETAILED ACTION

Notice of Pre-AIA or AIA Status

1. The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

Status of Claims

2. **Claim(s) 56-63** is/are pending.

3. **Claim(s) 1-55 and 64-90** is/are canceled.

Claim Rejections - 35 USC § 112

4. Applicants' amendment(s) have overcome the rejection(s) of **claim(s) 35-41, 44, and 64-72** under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph.

Allowable Subject Matter

5. **Claim(s) 56-63** is/are allowed.

6. The following is an examiner's statement of reasons for allowance:

The closest prior art of record is Liu (CN 10210806 A, see machine translation).

Liu discloses a lithium-ion secondary battery, comprising a positive electrode active material layer comprising a positive electrode active material particle comprising a first transition metal, a second transition metal, a representative element, and fluorine (see $Li_{1.01}Co_{0.99}Mg_{0.005}Al_{0.005}Ti_{0.01}O_{1.995}F_{0.005}$, [0029]).

Li does not disclose, teach, or suggest the following distinguishing feature(s):

A lithium-ion secondary battery, comprising a positive electrode active material layer comprising a positive electrode active material particle comprising a representative element and fluorine present in a portion that include crystal defects or a crack portion in the positive electrode active material particle.

Any comments considered necessary by applicant must be submitted no later than the payment of the issue fee and, to avoid processing delays, should preferably accompany the issue fee. Such submissions should be clearly labeled "Comments on Statement of Reasons for Allowance."

Conclusion

7. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Sean P Cullen, Ph.D. whose telephone number is (571)270-1251. The examiner can normally be reached on Monday-Thursday 6:00 am-4:00 pm Central, Friday 6:00 am-12:00 pm Central.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Basia A Ridley can be reached on (571)272-1453. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications

may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

CPC - Searched*		
Symbol	**Date**	**Examiner**
H01M4/131, 1315, 1391, 13915, 625, 8657	03/07/2019	SPC
Updated CPC search	06/27/2019	SPC
Updated CPC search	12/04/2019	SPC
Updated CPC search	02/25/2020	SPC
Updated CPC search	05/13/2020	SPC

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**
429	231.3	03/07/2019	SPC
	Updated USPC search	06/27/2019	SPC
	Updated USPC search	12/04/2019	SPC
	Updated USPC search	02/25/2020	SPC
	Updated USPC search	05/13/2020	SPC

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

Search Notes		
Search Notes	**Date**	**Examiner**
H01M4/131, 1315, 1391, 13915, 625, 8657	03/07/2019	SPC
429/231.3	03/07/2019	SPC
Inventor/applicant search, PALM	03/07/2019	SPC
EAST text search, see search history	03/07/2019	SPC
Updated inventor/applicant search, PALM	06/27/2019	SPC
Updated EAST text search, see search history	06/27/2019	SPC
Updated inventor/applicant search, PALM	12/04/2019	SPC
Updated EAST text search, see search history	12/04/2019	SPC
Updated inventor/applicant search, PALM	02/25/2020	SPC
Updated EAST text search, see search history	02/25/2020	SPC
Updated inventor/applicant search, PALM	05/13/2020	SPC
Updated EAST text search, see search history	05/13/2020	SPC

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**
H01M4	131, 1315, 1391, 13915, 625, 8657	05/13/2020	SPC
429	231.3, 231.5, 231.6, 231.95	05/13/2020	SPC

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	**Art Unit**
	Sean P Cullen, Ph.D.	1725

CPC								
Symbol							**Type**	**Version**
H01M	/	4	/	131			F	2013-01-01
H01M	/	4	/	1315			I	2013-01-01
H01M	/	4	/	1391			I	2013-01-01
H01M	/	4	/	625			I	2013-01-01
H01M	/	4	/	13915			I	2013-01-01
H01M	/	4	/	8657			I	2013-01-01
H01M	/	2004	/	021			A	2013-01-01
H01M	/	2004	/	028			A	2013-01-01
H01M	/	10	/	0525			A	2013-01-01

CPC Combination Sets								
Symbol					**Type**	**Set**	**Ranking**	**Version**
	/		/					

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	8	
/Sean P Cullen, Ph.D./ Primary Examiner, Art Unit 1725	13 May 2020	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	1C

U.S. Patent and Trademark Office — Part of Paper No.: 20200513

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	**Art Unit**
	Sean P Cullen, Ph.D.	1725

INTERNATIONAL CLASSIFICATION				
CLAIMED				
H01M4/131	/	4	/	131
H01M4/1315	/	4	/	1315
H01M4/1391	/	4	/	1391
H01M4/62	/	4	/	62
H01M4/13915	/	4	/	13915
H01M4/86	/	4	/	86
H01M4/02	/	4	/	02
H01M10/0525	/	10	/	0525

NON-CLAIMED				
	/		/	

US ORIGINAL CLASSIFICATION	
CLASS	**SUBCLASS**
429	231.3

CROSS REFERENCES(S)						
CLASS	**SUBCLASS (ONE SUBCLASS PER BLOCK)**					
429	231.5	231.6	231.95			

NONE		**Total Claims Allowed:**	
(Assistant Examiner)	(Date)	8	
/Sean P Cullen, Ph.D./ Primary Examiner, Art Unit 1725	13 May 2020	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	1C

U.S. Patent and Trademark Office — Part of Paper No.: 20200513

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	**Art Unit**
	Sean P Cullen, Ph.D.	1725

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIMS

Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original
1	60														
2	61														
3	62														
4	63														
5	56														
6	57														
7	58														
8	59														

NONE		**Total Claims Allowed:**	
(Assistant Examiner)	(Date)	8	
/Sean P Cullen, Ph.D./ Primary Examiner, Art Unit 1725	13 May 2020	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	1C

U.S. Patent and Trademark Office

Part of Paper No.: 20200513

OK TO ENTER: /SPC/

05/13/2020

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor	:	Teruaki OCHIAI	Art Unit	:	1725
Application No.	:	15/638,449	Examiner	:	Sean P. Cullen
Filed	:	June 30, 2017	Conf. No.	:	1074
Title	:	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY			

MAIL STOP AF
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

AMENDMENT IN REPLY TO ACTION OF MARCH 2, 2020

Please consider the following reply.

Amendments to the Claims are reflected in the listing of claims which begins on page 2 of this paper.

Remarks/Arguments begin on page 7 of this paper.

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L2	103	((H01M4/131,1315,1391,13915,625,8657).cpc. (429/231.3,231.5,231.6,231.95).ccls.) and (crack breakage defect) with (Mg magnesium fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/05/13 09:16
L4	53	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) same (Mg magnesium) same (fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/05/13 09:16
L5	51	(H01M.cpcl. H01M.cicl.) and (breakage defect) same (Mg magnesium) same (fluorine F)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/05/13 09:16
L6	95	L5 L4	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/05/13 09:16
L7	44	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) with (fluorine)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/05/13 09:17

L8	44	L7	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2020/05/13 09:17
L12	88	l2 not (l4 l5 l7)	~~US-PGPUB;~~ USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB		ON	2020/05/13 09:17

EAST Search History (Interference)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L3	35	((H01M4/131,1315,1391,13915,625,8657).cpc. (429/231.3,231.5,231.6,231.95).ccls.) and (crack breakage defect) with (Mg magnesium fluorine F)	USPAT	OR	ON	2020/05/13 09:16
L9	14	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) with (fluorine)	USPAT	OR	ON	2020/05/13 09:17
L10	6	(H01M.cpcl. H01M.cicl.) and (crack crystal near2 defect) same (Mg magnesium) same (fluorine F)	USPAT	OR	ON	2020/05/13 09:17
L11	7	(H01M.cpcl. H01M.cicl.) and (breakage defect) same (Mg magnesium) same (fluorine F)	USPAT	OR	ON	2020/05/13 09:17
L13	27	l3 not (l9 l10 l11)	USPAT	OR	ON	2020/05/13 09:18

5/13/2020 9:28:39 AM
C:\Users\scullen\Documents\EAST\Workspaces\15638449.wsp

Bibliographic Data

Application No: 15/638,449

Foreign Priority claimed: ◉ Yes ○ No

35 USC 119 (a-d) conditions met: ☑ Yes ☐ No ☐ Met After Allowance

Verified and Acknowledged: /Sean P Cullen, Ph.D./ [ ]

Examiner's Signature Initials

Title: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE

FILING or 371(c) DATE	CLASS	GROUP ART UNIT	ATTORNEY DOCKET NO.
06/30/2017	429	1725	12732-2003001
RULE			

APPLICANTS

SEMICONDUCTOR ENERGY LABORATORY CO., LTD., Atsugi-shi, JAPAN

INVENTORS

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Yohei MOMMA Isehara, JAPAN

Masahiro TAKAHASHI Atsugi, JAPAN

Ayae TSURUTA Atsugi, JAPAN

CONTINUING DATA

FOREIGN APPLICATIONS

JAPAN 2016-133143 07/05/2016

JAPAN 2016-133997 07/06/2016

JAPAN 2017-002831 01/11/2017

JAPAN 2017-030693 02/22/2017

JAPAN 2017-084321 04/21/2017

JAPAN 2017-119272 06/19/2017

IF REQUIRED, FOREIGN LICENSE GRANTED**

07/18/2017

STATE OR COUNTRY

JAPAN

ADDRESS

FISH & RICHARDSON P.C. (DC)

P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022
UNITED STATES

FILING FEE RECEIVED

$2,720

PTO/SB/08A (07-06)
Approved for use through 03/31/2007. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 1

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-8,945,770	02-03-2015	Koo.C et al.	

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2016-054151A	04-14-2016			Abst.

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant. 1 Applicant's unique citation designation number (optional). 2 See Kinds Codes of USPTO Patent Documents a www.uspto.gov or MPEP 901.04. 3 Enter Office that issued the document, by the two-letter code (WIPO Standard ST.3). 4 For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 5 Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. 6 Applicant is to place a check mark here if English language Translation is attached.

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 2 hours to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor	:	Teruaki OCHIAI	Art Unit	:	1725
Application No.	:	15/638,449	Examiner	:	Sean P. Cullen
Filed	:	June 30, 2017	Conf. No.	:	1074
Title	:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE (AS AMENDED)			

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

INFORMATION DISCLOSURE STATEMENT

Please consider the references listed on the enclosed disclosure form. A copy of the document required under 37 C.F.R. § 1.98(a)(2) is being submitted with this statement.

Each item of information in this information disclosure statement, with the exception of US8945770, was first cited in a communication from a foreign patent office in a counterpart foreign application dated March 17, 2020, which is not more than three months prior to the filing of this information disclosure statement. See 37 CFR 1.97(e)(1). The communication was an office action from the Japan Patent Office in counterpart Japanese Application No. 2019-174936.

US8945770 is an English-language document corresponding to JP2016-054151, and is cited in accordance with MPEP 609.04(b)(V).

EP2352190, which was previously submitted, is an English-language document corresponding to JP2016-054151.

This statement is being filed after a final action or a notice of allowance, but before payment of the issue fee. A late submission fee in the amount of $240 specified by 37 CFR §1.17(p) is being paid with this statement. Please apply any charges or credits to Deposit Account No. 06-1050, referencing the above attorney docket number.

Respectfully submitted,

Date: June 11, 2020

/John F. Hayden/
John F. Hayden
Reg. No. 37640

Customer Number 26171
Fish & Richardson P.C.
Telephone: (202) 783-5070
Facsimile: (877) 769-7945

100192186.doc

Electronic Patent Application Fee Transmittal

Application Number:	15638449
Filing Date:	30-Jun-2017
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Filer:	John F. Hayden/Alexis Bailey
Attorney Docket Number:	12732-2003001

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Miscellaneous:				
SUBMISSION- INFORMATION DISCLOSURE STMT	1806	1	240	240
		Total in USD ($)		**240**

Electronic Acknowledgement Receipt

EFS ID:	39688681
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	26171
Filer:	John F. Hayden/Alexis Bailey
Filer Authorized By:	John F. Hayden
Attorney Docket Number:	12732-2003001
Receipt Date:	11-JUN-2020
Filing Date:	30-JUN-2017
Time Stamp:	10:15:46
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	DA
Payment was successfully received in RAM	$240
RAM confirmation Number	E20206AA16016142
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Information Disclosure Statement (IDS) Form (SB08)	IDS.pdf	162280 06ac140b8ebc651d58929f3f5131294e46e6 9071	no	3
Warnings:					
Information:					
This is not an USPTO supplied IDS fillable form					
2	Foreign Reference	JP2016-054151.pdf	20409980 5573f8bbe32cbad4be3826d05ba4085dcd 941803	no	24
Warnings:					
Information:					
3	Fee Worksheet (SB06)	fee-info.pdf	30777 f8f89786567763d041903203e5a7065ba9b 64dde	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	20603037		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	12732-2003001	1074

26171 7590 06/19/2020
FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

EXAMINER
CULLEN, SEAN P

ART UNIT	PAPER NUMBER
1725	

NOTIFICATION DATE	DELIVERY MODE
06/19/2020	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

PATDOCTC@fr.com

Supplemental ***Notice of Allowability***	**Application No.** 15/638,449	**Applicant(s)** OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	**Art Unit** 1725	**AIA (FITF) Status** Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address--

All claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice of Allowance (PTOL-85) or other appropriate communication will be mailed in due course. **THIS NOTICE OF ALLOWABILITY IS NOT A GRANT OF PATENT RIGHTS.** This application is subject to withdrawal from issue at the initiative of the Office or upon petition by the applicant. See 37 CFR 1.313 and MPEP 1308.

1. ☑ This communication is responsive to the IDS filed on 11 June 2020.
   ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2. ☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

3. ☑ The allowed claim(s) is/are 56-63 . As a result of the allowed claim(s), you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see **http://www.uspto.gov/patents/init_events/pph/index.jsp** or send an inquiry to **PPHfeedback@uspto.gov.**

4. ☑ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

   Certified copies:

   a) ☑ All b) ☐ Some *c) ☐ None of the:

   1. ☑ Certified copies of the priority documents have been received.
   2. ☐ Certified copies of the priority documents have been received in Application No. _____ .
   3. ☐ Copies of the certified copies of the priority documents have been received in this national stage application from the International Bureau (PCT Rule 17.2(a)).

   * Certified copies not received: _____ .

Applicant has THREE MONTHS FROM THE "MAILING DATE" of this communication to file a reply complying with the requirements noted below. Failure to timely comply will result in ABANDONMENT of this application.
THIS THREE-MONTH PERIOD IS NOT EXTENDABLE.

5. ☐ CORRECTED DRAWINGS (as "replacement sheets") must be submitted.
   ☐ including changes required by the attached Examiner's Amendment / Comment or in the Office action of Paper No./Mail Date _____ .

   Identifying indicia such as the application number (see 37 CFR 1.84(c)) should be written on the drawings in the front (not the back) of each sheet. Replacement sheet(s) should be labeled as such in the header according to 37 CFR 1.121(d).

6. ☐ DEPOSIT OF and/or INFORMATION about the deposit of BIOLOGICAL MATERIAL must be submitted. Note the attached Examiner's comment regarding REQUIREMENT FOR THE DEPOSIT OF BIOLOGICAL MATERIAL.

Attachment(s)

1. ☐ Notice of References Cited (PTO-892)
2. ☑ Information Disclosure Statements (PTO/SB/08), Paper No./Mail Date _____.
3. ☐ Examiner's Comment Regarding Requirement for Deposit of Biological Material _____.
4. ☐ Interview Summary (PTO-413), Paper No./Mail Date. _____.
5. ☑ Examiner's Amendment/Comment
6. ☐ Examiner's Statement of Reasons for Allowance
7. ☐ Other _____.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

DETAILED ACTION

Information Disclosure Statement

1. The information disclosure statement (IDS) submitted on 11 June 2020 was filed after the mailing date of the Notice of Allowance on 20 May 2020. The submission complies with the provisions of 37 CFR 1.97. Accordingly, the information disclosure statement is being considered by the examiner.

Conclusion

2. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Sean P Cullen, Ph.D. whose telephone number is (571)270-1251. The examiner can normally be reached on Monday-Thursday 6:00 am-4:00 pm Central, Friday 6:00 am-12:00 pm Central.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Basia A Ridley can be reached on (571)272-1453. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR

system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

PTO/SB/08A (07-06)
Approved for use through 03/31/2007. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 1

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-8,945,770	02-03-2015	Koo.C et al.	

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2016-054151A	04-14-2016			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/16/2020

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant. 1 Applicant's unique citation designation number (optional). 2 See Kinds Codes of USPTO Patent Documents a www.uspto.gov or MPEP 901.04. 3 Enter Office that issued the document, by the two-letter code (WIPO Standard ST.3). 4 For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 5 Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. 6 Applicant is to place a check mark here if English language Translation is attached.

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If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /SPC/

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor	:	Teruaki OCHIAI	Art Unit	:	1725
Application No.	:	15/638,449	Examiner	:	Sean P. Cullen
Filed	:	June 30, 2017	Conf. No.	:	1074
Title	:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE (AS AMENDED)			

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

INFORMATION DISCLOSURE STATEMENT

Please consider the references listed on the enclosed disclosure form. A copy of the document required under 37 C.F.R. § 1.98(a)(2) is being submitted with this statement.

Each item of information in this information disclosure statement, with the exception of US8945770, was first cited in a communication from a foreign patent office in a counterpart foreign application dated March 17, 2020, which is not more than three months prior to the filing of this information disclosure statement. See 37 CFR 1.97(e)(1). The communication was an office action from the Japan Patent Office in counterpart Japanese Application No. 2019-174936.

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ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /SPC/

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Respectfully submitted,

Date: June 11, 2020

/John F. Hayden/
John F. Hayden
Reg. No. 37640

Customer Number 26171
Fish & Richardson P.C.
Telephone: (202) 783-5070
Facsimile: (877) 769-7945

100192186.doc

AMPEREX Ex. 1002
Page 1372

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /SPC/

PTO/SB/30 (11-17)
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U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
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Request for Continued Examination (RCE) Transmittal

Address to:
Mail Stop RCE
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Sean P. Cullen
Attorney Docket Number	12732-2003001

This is a Request for Continued Examination (RCE) under 37 CFR 1 .114 of the above-identified application.
Request for Continued Examination (RCE) practice under 37 CFR 1.114 does not apply to any utility or plant application filed prior to June 8, 1995, or to any design application. See Instruction Sheet for RCEs (not to be submitted to the USPTO) on page 2.

1. Submission required under 37 CFR 1.114 Note: If the RCE is proper, any previously filed unentered amendments and amendments enclosed with the RCE will be entered in the order in which they were filed unless applicant instructs otherwise. If applicant does not wish to have any previously filed unentered amendment(s) entered, applicant must request non-entry of such amendment(s).

   a. ☐ Previously submitted. If a final Office action is outstanding, any amendments filed after the final Office action may be considered as a submission even if this box is not checked.

      i. ☐ Consider the arguments in the Appeal Brief or Reply Brief previously filed on _____

      ii. ☐ Other _____

   b. ☒ Enclosed

      i. ☐ Amendment/Reply

      ii. ☐ Affidavit(s)/ Declaration(s)

      iii. ☒ Information Disclosure Statement (IDS)

      iv. ☐ Other _____

2. Miscellaneous

   a. ☐ Suspension of action on the above-identified application is requested under 37 CFR 1.103(c) for a period of _____ months. (Period of suspension shall not exceed 3 months; Fee under 37 CFR 1.17(i) required)

   b. ☐ Other _____

3. Fees The RCE fee under 37 CFR 1.17(e) is required by 37 CFR 1.114 when the RCE is filed.
   The Director is hereby authorized to charge the following fees any underpayment of fees, or credit any overpayments to

   a. ☒ Deposit Account No. 06-1050.

      i. ☒ RCE fee required under 37 CFR 1.17(e)

      ii ☐ Extension of time fee (37 CFR 1.136 and 1.17)

      iii ☒ Other any deficiencies

   b. ☐ Check in the amount of $ _____ enclosed

   c. ☐ Payment by credit card (Form PTO-2038 enclosed)

   d. ☒ Payment by EFS-Web

WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT REQUIRED

Signature	/Heather L. Flanagan/	Date	June 19, 2020
Name (Print/Type)	Heather L. Flanagan	Registration No.	54,101

CERTIFICATE OF MAILING OR TRANSMISSION

I hereby certify that this correspondence is being EFS-Web transmitted to the United States Patent and Trademark Office (USPTO), deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to: Mail Stop RCE, Commissioner for Patents, P. O. Box 1450, Alexandria, VA 22313-1450 or facsimile transmitted to the USPTO on the date shown below.

Signature			
Name (Print/Type)		Date	

This collection of information is required by 37 CFR 1.114. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Mail Stop RCE, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**
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PTO/SB/08A (07-06)
Approved for use through 03/31/2007. OMB 0651-0031
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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-8,236,449	08-07-2012	Nakura.K	
		US-10,128,495	11-13-2018	Satow.T et al.	
		US-10,243,215	03-26-2019	Shitaba.J et al.	
		US-10,361,432	07-23-2019	Takaichi.Y et al.	
		US-2002/0110736	08-15-2002	Kweon.H et al.	
		US-2003/0104279	06-05-2003	Miyazaki.S et al.	
		US-2004/0229124	11-18-2004	Miyamoto.Y et al.	
		US-2005/0019662	01-27-2005	Suhara.M et al.	
		US-2007/0117014	05-24-2007	Saito.N et al.	
		US-2008/0241693	10-02-2008	Fukuchi.M et al.	
		US-2009/0104532	04-23-2009	Hosoya.Y	
		US-2010/0019194	01-28-2010	Fujiwara.T et al.	
		US-2010/0247986	09-30-2010	Toyama.T et al.	
		US-2011/0059367	03-10-2011	Morita.K et al.	
		US-2011/0256437	10-20-2011	Katsuki.T et al.	
		US-2011/0297876	12-08-2011	Masukuni.H et al.	
		US-2012/0045692	02-23-2012	Takemura.Y et al.	
		US-2012/0064406	03-15-2012	Sato.H et al.	
		US-2012/0244430	09-27-2012	Yamazaki.S et al.	

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2004-342554A	12-02-2004			Full
		CN-102610806A	07-25-2012			Full
		JP-2015-069958A	04-13-2015			Full
		JP-2003-221235A	08-05-2003			Full
		JP-2012-209077A	10-25-2012			Full
		JP-2007-213866A	08-23-2007			Full

Examiner Signature		Date Considered	

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 2 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-2012/0261622	10-18-2012	Honma.M	
		US-2012/0295163	11-22-2012	Yanagita.H et al.	
		US-2012/0330044	12-27-2012	Hou.S	
		US-2013/0040193	02-14-2013	Tsuchida.Y et al.	
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		US-2014/0127567	05-08-2014	Kuriki.K et al.	
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		US-2016/0028080	01-28-2016	Sugiura.R	
		US-2016/0276658	09-22-2016	Choi.S et al.	
		US-2016/0285102	09-29-2016	Shitaba.J et al.	
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		US-2016/0349905	12-01-2016	Momma.Y et al.	

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		JP-05-314965A	11-26-1993			Abst.
		JP-05-314995A	11-26-1993			Abst.
		JP-08-236114A	09-13-1996			Abst.
		JP-11-096993A	04-09-1999			Full
		JP-2000-203842A	07-25-2000			Abst.
		JP-2001-319692A	11-16-2001			Abst.

Examiner Signature		Date Considered	

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Sheet 3 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

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		US-2016/0380271	12-29-2016	Ochiai.T et al.	
		US-2017/0005364	01-05-2017	Yamazaki.S et al.	
		US-2017/0040594	02-09-2017	Yamaki.T et al.	
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		JP-2002-216760A	08-02-2002			Abst.
		JP-2002-352802A	12-06-2002			Abst.
		JP-2004-014381A	01-15-2004			Full
		JP-2005-158612A	06-16-2005			Abst.
		JP-2005-302510A	10-27-2005			Full
		JP-2006-164758A	06-22-2006			Abst.

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 4 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

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		US-6,737,195	05-18-2004	Kweon.H et al.	
		US-6,919,144	07-19-2005	Miyazaki.S et al.	
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Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2006-261132A	09-28-2006			Abst.
		JP-2007-128714A	05-24-2007			Abst.
		JP-2010-080407A	04-08-2010			Abst.
		JP-2010-102895A	05-06-2010			Full
		JP-2010-244847A	10-28-2010			Full
		JP-2010-272239A	12-02-2010			Abst.

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 5 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

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		US-RE.43,276	03-27-2012	Kweon.H et al.	

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		JP-2011-138718A	07-14-2011			Full
		JP-2014-049239A	03-17-2014			Abst.
		JP-2014-063707A	04-10-2014			Abst.
		JP-2014-063708A	04-10-2014			Abst.
		JP-2015-099722A	05-28-2015			Abst.
		JP-2015-156363A	08-27-2015			Abst.

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Sheet 6 of 13

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		JP-2017-021942A	01-26-2017			Abst.
		JP-2017-091777A	05-25-2017			Full
		JP-2018-092934A	06-14-2018			Abst.
		JP-3031546	04-10-2000			Abst.
		JP-4739780	08-03-2011			Abst.

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Sheet 7 of 13

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Art Unit	1725
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		WO-2012/029729	03-08-2012			Abst.
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		CN-102569775A	07-11-2012			Full

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Attorney Docket Number	12732-2003001

NON PATENT LITERATURE DOCUMENTS

Examiner Initials*	Cite No.[1]	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published.	T[2]
		Sun.Y et al., "High-energy cathode material for long-life and safe lithium batteries", NATURE MATERIALS, March 22, 2009, Vol. 8, pp. 320-324.	Eng.
		JOSHI.T et al., "Effects of Dissolved Transition Metals on the Electrochemical Performance and SEI Growth in Lithium-Ion Batteries", J. Electrochem. SOC. (Journal of the Electrochemical Society), 2014, Vol. 161, No. 12, pp. A1915-A1921.	Eng.
		Chung.K et al., "Structural Studies on the Effects of ZrO2 Coating on LiCoO2 during Cycling Using In Situ X-Ray Diffraction Technique", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), 2006, Vol. 153, No. 11, pp. A2152-A2157.	Eng.
		Liu.L et al., "Electrochemical and In Situ Synchrotron XRD Studies on Al2O3-Coated LiCoO2 Cathode Material", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), 2004, Vol. 151, No. 9, pp. A1344-A1351.	Eng.
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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 9 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

NON PATENT LITERATURE DOCUMENTS

Examiner Initials*	Cite No.[1]	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published.	T[2]
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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 10 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

NON PATENT LITERATURE DOCUMENTS

Examiner Initials*	Cite No.[1]	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published.	T[2]
		Mladenov.M et al., "Effect of Mg doping and MgO-surface modification on the cycling stability of LiCoO2 electrodes.", Electrochemistry Communications, August 1, 2001, Vol. 3, No. 8, pp. 410-416.	Eng.
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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 11 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

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		Wang.Z et al., "Electrochemical Evaluation and Structural Characterization of Commercial LiCoO2 Surfaces Modified with MgO for Lithium-Ion Batteries", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), March 4, 2002, Vol. 149, No. 4, pp. A466-A471.	Eng.
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Sheet 12 of 13

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Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

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		Quinlan.R et al., "XPS Investigation of the Electrolyte Induced Stabilization of LiCoO2 and "AlPO4"-Coated LiCoO2 Composite Electrodes", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), December 3, 2015, Vol. 163, No. 2, pp. A300-A308.	Eng.
		Cho.J et al., "Comparison of Al2O3- and AlPO4-coated LiCoO2 cathode materials for a Li-ion cell", JOURNAL OF POWER SOURCES, 2005, Vol. 146, pp. 58-64, ELSEVIER.	Eng.
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		Hong.W et al., "Modification of LiCoO2 by Surface Coating with MgO/TiO2/SiO2 for High-Performance Lithium-Ion Battery", ELECTROCHEMICAL AND SOLID-STATE LETTERS, December 20, 2005, Vol. 9, No. 2, pp. A82-A85.	Eng.
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First Named Inventor	Teruaki OCHIAI
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Attorney Docket Number	12732-2003001

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		Bensalah.N et al., "Review on Synthesis, Characterizations, and Electrochemical Properties of Cathode Materials for Lithium Ion Batteries", Journal of Material Science & Engineering, 2016, Vol. 5, No. 4, pp. 1000258-1-1000258-21.	Eng.
		Lu.Y et al., "Recent progress on lithium-ion batteries with high electrochemical performance", SCIENCE CHINA CHEMISTRY, February 25, 2019, Vol. 62, No. 5, pp. 533-548.	Eng.
		Xiao.B, "Surface Modifification of Electrode Materials for Lithium-Ion Batteries", Graduate Program in Mechanical and Materials Engineering, 2006, pp. 1-235.	Eng.
		Ohnishi.M et al., "Investigation of the surface degradation of LiCoO2 particles in the cathode materials of Li-ion batteries using FIB-TOF-SIMS", Journal of Surface Analysis, 2013, Vol. 20, No. 2, pp. 99-110.	Eng.
		Iwaya.K et al., "Impact of Lithium-Ion Ordering on Surface Electronic States of LixCoO2", PHYS. REV. LETT. (PHYSICAL REVIEW LETTERS), September 20, 2013, Vol. 111, No. 12, pp. 126104-1-126104-5.	Eng.
		Zou.M et al., "Synthesis of High-Voltage (4.5 V) Cycling Doped LiCoO2 for Use in Lithium Rechargeable Cells", CHEM. MATER. (CHEMISTRY OF MATERIALS), December 16, 2003, Vol. 15, No. 25, pp. 4699-4702.	Eng.
		Schipper.F et al., "Study of Cathode Materials for Lithium-Ion Batteries: Recent Progress and New Challenges", Inorganics, April 28, 2017, Vol. 5, No. 2, pp. 1-29.	Eng.

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor	:	Teruaki OCHIAI	Art Unit	:	1725
Application No.	:	15/638,449	Examiner	:	Sean P. Cullen
Filed	:	June 30, 2017	Conf. No.	:	1074
Title	:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE (AS AMENDED)			

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

INFORMATION DISCLOSURE STATEMENT

Please consider the references listed on the enclosed disclosure form. Copies of the documents required under 37 C.F.R. § 1.98(a)(2) are being submitted with this statement.

This statement is being filed with a Request for Continued Examination. No fee is required. Please apply any charges or credits to Deposit Account No. 06-1050, referencing the above attorney docket number.

Respectfully submitted,

Date: June 19, 2020

/Heather L. Flanagan/
Heather L. Flanagan
Reg. No. 54,101

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Electronic Patent Application Fee Transmittal

Application Number:	15638449
Filing Date:	30-Jun-2017
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Filer:	Heather L. Flanagan/Alexis Bailey
Attorney Docket Number:	12732-2003001

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Miscellaneous:				
RCE- 2ND AND SUBSEQUENT REQUEST	1820	1	1900	1900
		Total in USD ($)		**1900**

Electronic Acknowledgement Receipt

EFS ID:	39777254
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	26171
Filer:	Heather L. Flanagan/Alexis Bailey
Filer Authorized By:	Heather L. Flanagan
Attorney Docket Number:	12732-2003001
Receipt Date:	19-JUN-2020
Filing Date:	30-JUN-2017
Time Stamp:	16:54:09
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	DA
Payment was successfully received in RAM	$1900
RAM confirmation Number	E20206IG54485471
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

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New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

Electronic Acknowledgement Receipt

EFS ID:	39777677
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	26171
Filer:	Heather L. Flanagan/Alexis Bailey
Filer Authorized By:	Heather L. Flanagan
Attorney Docket Number:	12732-2003001
Receipt Date:	19-JUN-2020
Filing Date:	30-JUN-2017
Time Stamp:	17:17:24
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

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<u>New Applications Under 35 U.S.C. 111</u>
If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.
<u>National Stage of an International Application under 35 U.S.C. 371</u>
If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.
<u>New International Application Filed with the USPTO as a Receiving Office</u>
If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

PTO/SB/08A (07-06)
Approved for use through 03/31/2007. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 1

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2012-018914A	01-26-2012			Abst.

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant. 1 Applicant's unique citation designation number (optional). 2 See Kinds Codes of USPTO Patent Documents at www.uspto.gov or MPEP 901.04. 3 Enter Office that issued the document, by the two-letter code (WIPO Standard ST.3). 4 For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 5 Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. 6 Applicant is to place a check mark here if English language Translation is attached.

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 2 hours to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor	:	Teruaki OCHIAI	Art Unit	:	1725
Application No.	:	15/638,449	Examiner	:	Sean P. Cullen
Filed	:	June 30, 2017	Conf. No.	:	1074
Title	:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE (AS AMENDED)			

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

INFORMATION DISCLOSURE STATEMENT

Please consider the references listed on the enclosed disclosure form. Copies of the documents required under 37 C.F.R. § 1.98(a)(2) are being submitted with this statement.

This statement is being filed after a request for continued examination and before the mailing of a first action. Please apply any charges or credits to Deposit Account No. 06-1050, referencing the above attorney docket number.

Respectfully submitted,

Date: June 22, 2020

/Heather L. Flanagan/
Heather L. Flanagan
Reg. No. 54,101

Customer Number 26171
Fish & Richardson P.C.
Telephone: +1 512-226-8156
Facsimile: (877) 769-7945

100199854.doc

Electronic Acknowledgement Receipt

EFS ID:	39792230
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	26171
Filer:	Heather L. Flanagan/Alexis Bailey
Filer Authorized By:	Heather L. Flanagan
Attorney Docket Number:	12732-2003001
Receipt Date:	22-JUN-2020
Filing Date:	30-JUN-2017
Time Stamp:	17:31:50
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Information Disclosure Statement (IDS) Form (SB08)	IDS.pdf	160249 4954ffa2317a0e7db8c49d17bd1fac5b0959 65ab	no	2

Warnings:

Information:					
This is not an USPTO supplied IDS fillable form					
2	Foreign Reference	JP2012-018914.pdf	10030151 bfc0cb8179f94a4b0c9666afc72621bf0e9b6139	no	34
Warnings:					
Information:					
		Total Files Size (in bytes):	10190400		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

<u>New Applications Under 35 U.S.C. 111</u>
If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.
<u>National Stage of an International Application under 35 U.S.C. 371</u>
If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.
<u>New International Application Filed with the USPTO as a Receiving Office</u>
If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

NOTICE OF ALLOWANCE AND FEE(S) DUE

26171 7590 06/25/2020
FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

EXAMINER	
CULLEN, SEAN P	
ART UNIT	PAPER NUMBER
1725	

DATE MAILED: 06/25/2020

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	12732-2003001	1074

TITLE OF INVENTION: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1000	$0.00	$0.00	$1000	09/25/2020

THE APPLICATION IDENTIFIED ABOVE HAS BEEN EXAMINED AND IS ALLOWED FOR ISSUANCE AS A PATENT. PROSECUTION ON THE MERITS IS CLOSED. THIS NOTICE OF ALLOWANCE IS NOT A GRANT OF PATENT RIGHTS. THIS APPLICATION IS SUBJECT TO WITHDRAWAL FROM ISSUE AT THE INITIATIVE OF THE OFFICE OR UPON PETITION BY THE APPLICANT. SEE 37 CFR 1.313 AND MPEP 1308.

THE ISSUE FEE AND PUBLICATION FEE (IF REQUIRED) MUST BE PAID WITHIN THREE MONTHS FROM THE MAILING DATE OF THIS NOTICE OR THIS APPLICATION SHALL BE REGARDED AS ABANDONED. THIS STATUTORY PERIOD CANNOT BE EXTENDED. SEE 35 U.S.C. 151. THE ISSUE FEE DUE INDICATED ABOVE DOES NOT REFLECT A CREDIT FOR ANY PREVIOUSLY PAID ISSUE FEE IN THIS APPLICATION. IF AN ISSUE FEE HAS PREVIOUSLY BEEN PAID IN THIS APPLICATION (AS SHOWN ABOVE), THE RETURN OF PART B OF THIS FORM WILL BE CONSIDERED A REQUEST TO REAPPLY THE PREVIOUSLY PAID ISSUE FEE TOWARD THE ISSUE FEE NOW DUE.

HOW TO REPLY TO THIS NOTICE:

I. Review the ENTITY STATUS shown above. If the ENTITY STATUS is shown as SMALL or MICRO, verify whether entitlement to that entity status still applies.

If the ENTITY STATUS is the same as shown above, pay the TOTAL FEE(S) DUE shown above.

If the ENTITY STATUS is changed from that shown above, on PART B - FEE(S) TRANSMITTAL, complete section number 5 titled "Change in Entity Status (from status indicated above)".

For purposes of this notice, small entity fees are 1/2 the amount of undiscounted fees, and micro entity fees are 1/2 the amount of small entity fees.

II. PART B - FEE(S) TRANSMITTAL, or its equivalent, must be completed and returned to the United States Patent and Trademark Office (USPTO) with your ISSUE FEE and PUBLICATION FEE (if required). If you are charging the fee(s) to your deposit account, section "4b" of Part B - Fee(s) Transmittal should be completed and an extra copy of the form should be submitted. If an equivalent of Part B is filed, a request to reapply a previously paid issue fee must be clearly made, and delays in processing may occur due to the difficulty in recognizing the paper as an equivalent of Part B.

III. All communications regarding this application must give the application number. Please direct all communications prior to issuance to Mail Stop ISSUE FEE unless advised to the contrary.

IMPORTANT REMINDER: Maintenance fees are due in utility patents issuing on applications filed on or after Dec. 12, 1980. It is patentee's responsibility to ensure timely payment of maintenance fees when due. More information is available at www.uspto.gov/PatentMaintenanceFees.

PART B - FEE(S) TRANSMITTAL

Complete and send this form, together with applicable fee(s), by mail or fax, or via EFS-Web.

By mail, send to: Mail Stop ISSUE FEE
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

By fax, send to: (571)-273-2885

INSTRUCTIONS: This form should be used for transmitting the ISSUE FEE and PUBLICATION FEE (if required). Blocks 1 through 5 should be completed where appropriate. All further correspondence including the Patent, advance orders and notification of maintenance fees will be mailed to the current correspondence address as indicated unless corrected below or directed otherwise in Block 1, by (a) specifying a new correspondence address; and/or (b) indicating a separate "FEE ADDRESS" for maintenance fee notifications.

CURRENT CORRESPONDENCE ADDRESS (Note: Use Block 1 for any change of address)

26171 7590 06/25/2020
FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

Note: A certificate of mailing can only be used for domestic mailings of the Fee(s) Transmittal. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawing, must have its own certificate of mailing or transmission.

Certificate of Mailing or Transmission

I hereby certify that this Fee(s) Transmittal is being deposited with the United States Postal Service with sufficient postage for first class mail in an envelope addressed to the Mail Stop ISSUE FEE address above, or being transmitted to the USPTO via EFS-Web or by facsimile to (571) 273-2885, on the date below.

(Typed or printed name)

(Signature)

(Date)

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	12732-2003001	1074

TITLE OF INVENTION: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1000	$0.00	$0.00	$1000	09/25/2020

EXAMINER	ART UNIT	CLASS-SUBCLASS
CULLEN, SEAN P	1725	429-231300

1. Change of correspondence address or indication of "Fee Address" (37 CFR 1.363).

❑ Change of correspondence address (or Change of Correspondence Address form PTO/SB/122) attached.

❑ "Fee Address" indication (or "Fee Address" Indication form PTO/SB/47; Rev 03-09 or more recent) attached. **Use of a Customer Number is required.**

2. For printing on the patent front page, list
(1) The names of up to 3 registered patent attorneys or agents OR, alternatively,
(2) The name of a single firm (having as a member a registered attorney or agent) and the names of up to 2 registered patent attorneys or agents. If no name is listed, no name will be printed.

1_____
2_____
3_____

3. ASSIGNEE NAME AND RESIDENCE DATA TO BE PRINTED ON THE PATENT (print or type)

PLEASE NOTE: Unless an assignee is identified below, no assignee data will appear on the patent. If an assignee is identified below, the document must have been previously recorded, or filed for recordation, as set forth in 37 CFR 3.11 and 37 CFR 3.81(a). Completion of this form is NOT a substitute for filing an assignment.

(A) NAME OF ASSIGNEE

(B) RESIDENCE: (CITY and STATE OR COUNTRY)

Please check the appropriate assignee category or categories (will not be printed on the patent) : ❑ Individual ❑ Corporation or other private group entity ❑ Government

4a. Fees submitted: ❑Issue Fee ❑Publication Fee (if required) ❑Advance Order - # of Copies _____

4b. Method of Payment: *(Please first reapply any previously paid fee shown above)*

❑ Electronic Payment via EFS-Web ❑ Enclosed check ❑ Non-electronic payment by credit card (Attach form PTO-2038)

❑ The Director is hereby authorized to charge the required fee(s), any deficiency, or credit any overpayment to Deposit Account No. _____

5. **Change in Entity Status** (from status indicated above)

❑ Applicant certifying micro entity status. See 37 CFR 1.29

❑ Applicant asserting small entity status. See 37 CFR 1.27

❑ Applicant changing to regular undiscounted fee status.

NOTE: Absent a valid certification of Micro Entity Status (see forms PTO/SB/15A and 15B), issue fee payment in the micro entity amount will not be accepted at the risk of application abandonment.
NOTE: If the application was previously under micro entity status, checking this box will be taken to be a notification of loss of entitlement to micro entity status.
NOTE: Checking this box will be taken to be a notification of loss of entitlement to small or micro entity status, as applicable.

NOTE: This form must be signed in accordance with 37 CFR 1.31 and 1.33. See 37 CFR 1.4 for signature requirements and certifications.

Authorized Signature _____ Date _____

Typed or printed name _____ Registration No. _____

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	12732-2003001	1074

26171 7590 06/25/2020
FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

EXAMINER	
CULLEN, SEAN P	
ART UNIT	PAPER NUMBER
1725	

DATE MAILED: 06/25/2020

Determination of Patent Term Adjustment under 35 U.S.C. 154 (b)
(Applications filed on or after May 29, 2000)

The Office has discontinued providing a Patent Term Adjustment (PTA) calculation with the Notice of Allowance.

Section 1(h)(2) of the AIA Technical Corrections Act amended 35 U.S.C. 154(b)(3)(B)(i) to eliminate the requirement that the Office provide a patent term adjustment determination with the notice of allowance. See Revisions to Patent Term Adjustment, 78 Fed. Reg. 19416, 19417 (Apr. 1, 2013). Therefore, the Office is no longer providing an initial patent term adjustment determination with the notice of allowance. The Office will continue to provide a patent term adjustment determination with the Issue Notification Letter that is mailed to applicant approximately three weeks prior to the issue date of the patent, and will include the patent term adjustment on the patent. Any request for reconsideration of the patent term adjustment determination (or reinstatement of patent term adjustment) should follow the process outlined in 37 CFR 1.705.

Any questions regarding the Patent Term Extension or Adjustment determination should be directed to the Office of Patent Legal Administration at (571)-272-7702. Questions relating to issue and publication fee payments should be directed to the Customer Service Center of the Office of Patent Publication at 1-(888)-786-0101 or (571)-272-4200.

OMB Clearance and PRA Burden Statement for PTOL-85 Part B

The Paperwork Reduction Act (PRA) of 1995 requires Federal agencies to obtain Office of Management and Budget approval before requesting most types of information from the public. When OMB approves an agency request to collect information from the public, OMB (i) provides a valid OMB Control Number and expiration date for the agency to display on the instrument that will be used to collect the information and (ii) requires the agency to inform the public about the OMB Control Number's legal significance in accordance with 5 CFR 1320.5(b).

The information collected by PTOL-85 Part B is required by 37 CFR 1.311. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 30 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, Virginia 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, Virginia 22313-1450. Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether disclosure of these records is required by the Freedom of Information Act.
2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspection or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

Notice of Allowability	Application No. 15/638,449	Applicant(s) OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address--

All claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice of Allowance (PTOL-85) or other appropriate communication will be mailed in due course. **THIS NOTICE OF ALLOWABILITY IS NOT A GRANT OF PATENT RIGHTS.** This application is subject to withdrawal from issue at the initiative of the Office or upon petition by the applicant. See 37 CFR 1.313 and MPEP 1308.

1. ☑ This communication is responsive to the IDS filed on 19 June 2020.
   ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2. ☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

3. ☑ The allowed claim(s) is/are 56-63 . As a result of the allowed claim(s), you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see **http://www.uspto.gov/patents/init_events/pph/index.jsp** or send an inquiry to **PPHfeedback@uspto.gov.**

4. ☑ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

   Certified copies:

   a) ☑ All b) ☐ Some *c) ☐ None of the:

   1. ☑ Certified copies of the priority documents have been received.
   2. ☐ Certified copies of the priority documents have been received in Application No. _____ .
   3. ☐ Copies of the certified copies of the priority documents have been received in this national stage application from the International Bureau (PCT Rule 17.2(a)).

   * Certified copies not received: _____ .

Applicant has THREE MONTHS FROM THE "MAILING DATE" of this communication to file a reply complying with the requirements noted below. Failure to timely comply will result in ABANDONMENT of this application.
THIS THREE-MONTH PERIOD IS NOT EXTENDABLE.

5. ☐ CORRECTED DRAWINGS (as "replacement sheets") must be submitted.
   ☐ including changes required by the attached Examiner's Amendment / Comment or in the Office action of Paper No./Mail Date _____ .

   Identifying indicia such as the application number (see 37 CFR 1.84(c)) should be written on the drawings in the front (not the back) of each sheet. Replacement sheet(s) should be labeled as such in the header according to 37 CFR 1.121(d).

6. ☐ DEPOSIT OF and/or INFORMATION about the deposit of BIOLOGICAL MATERIAL must be submitted. Note the attached Examiner's comment regarding REQUIREMENT FOR THE DEPOSIT OF BIOLOGICAL MATERIAL.

Attachment(s)

1. ☑ Notice of References Cited (PTO-892)
2. ☑ Information Disclosure Statements (PTO/SB/08), Paper No./Mail Date _____.
3. ☐ Examiner's Comment Regarding Requirement for Deposit of Biological Material _____.
4. ☐ Interview Summary (PTO-413), Paper No./Mail Date. _____.
5. ☑ Examiner's Amendment/Comment
6. ☐ Examiner's Statement of Reasons for Allowance
7. ☐ Other _____.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

DETAILED ACTION

Notice of Pre-AIA or AIA Status

1. The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

Status of Claims

2. **Claim(s) 56–63** is/are pending.

3. **Claim(s) 1–55 and 64–90** is/are canceled.

Information Disclosure Statement

4. The information disclosure statement (IDS) submitted on 22 June 2020 was filed before the mailing of a first Office Action on the merits following a request for continued examination. The submission complies with the provisions of 37 CFR 1.97. Accordingly, the information disclosure statement is being considered by the examiner.

5. The information disclosure statement filed on 19 June 2020 does not fully comply with the requirements of 37 CFR 1.98(b) because:

A copy of CN 102569775 **A** cited on page 7 of the information disclosure statement has not been provided. A copy of CN 102569775 **B** has been provided. CN 102569775 A has been cited on the accompanying PTO-892 and a copy has been provided by the Office.

A copy of " Electronic phase diagram of the layered cobalt oxide system Li_xCoO_2 ($0.0 \leq x \leq 1.0$)" cited on page 8 has not been provided. "Electronic phase diagram of the layered cobalt

oxide system Li_xCoO_2 ($0.0 \leq x \leq 1.0$)" has been cited on the accompanying PTO-892 and a copy has been provided by the Office.

A copy of "Stabilization of the Electronic Structure at the Cathode/Electrolyte Interface via MgO Ultra-Thin Layer during Lithium-Ions Insertion/Extraction" has not been provided. "Stabilization of the Electronic Structure at the Cathode/Electrolyte Interface via MgO Ultra-Thin Layer during Lithium-Ions Insertion/Extraction" has been cited on the accompanying PTO-892 and a copy has been provided by the Office.

A copy of "Investigation of the surface degradation of $LiCoO_2$ particles in the cathode materials of Li-ion batteries using FIB-TOF-SIMS" has not been provided. "Investigation of the surface degradation of $LiCoO_2$ particles in the cathode materials of Li-ion batteries using FIB-TOF-SIMS" has been cited on the accompanying PTO-892 and a copy has been provided by the Office.

Since the submission appears to be *bona fide*, applicant is given **ONE (1) MONTH** from the date of this notice to supply the above mentioned omissions or corrections in the information disclosure statement. NO EXTENSION OF THIS TIME LIMIT MAY BE GRANTED UNDER EITHER 37 CFR 1.136(a) OR (b). Failure to timely comply with this notice will result in the above mentioned information disclosure statement being placed in the application file with the noncomplying information **not** being considered. See 37 CFR 1.97(i).

Conclusion

6. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Sean P Cullen, Ph.D. whose telephone number is (571)270-1251.

The examiner can normally be reached on Monday-Thursday 6:00 am-4:00 pm Central, Friday 6:00 am-12:00 pm Central.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Basia A Ridley can be reached on (571)272-1453. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Sean P Cullen, Ph.D./
Primary Examiner, Art Unit 1725

Notice of References Cited	Application/Control No. 15/638,449	Applicant(s)/Patent Under Reexamination OCHIAI et al.	
	Examiner Sean P Cullen, Ph.D.	Art Unit 1725	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	CPC Classification	US Classification
	A					
	B					
	C					
	D					
	E					
	F					
	G					
	H					
	I					
	J					
	K					
	L					
	M					

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	CPC Classification
	N	CN-102569775-A	07-2012	CN	FENGGANG ZHAO	
	O					
	P					
	Q					
	R					
	S					
	T					

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*		Include as applicable: Author, Title Date, Publisher, Edition or Volume, Pertinent Pages)
	U	Motohashi, T. et al., Electronic phase diagram of the layered cobalt oxide system $LiCoO_2$ ($0.0 \leq x \leq 1.0$), 10-2008, Physical Review B, 80, 165114-1–165114-9 (Year: 2008)
	V	Yamamoto, K. et al., Stabilization of the Electronic Structure at the Cathode/Electrolyte Interface via MgO Ultra-Thin Layer during Lithium-Ions Insertion/Extraction, 10-2014, Electrochemistry, 82, 891–896. (Year: 2014)
	W	Ohnishi, M. et al., Investigation of the surface degradation of $LiCoO_2$ particles in the cathode materials of Li-ion batteries using FIB-TOF-SIMS, 2013, Journal of Surface Analysis, 20, 99-110 (Year: 2013)
	X	

*A copy of this reference is not being furnished with this Office action. (See MPEP § 707.05(a).)
Dates in MM-YYYY format are publication dates. Classifications may be US or foreign.

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

CPC - Searched*		
Symbol	**Date**	**Examiner**
H01M4/131, 1315, 1391, 13915, 625, 8657	03/07/2019	SPC
Updated CPC search	06/27/2019	SPC
Updated CPC search	12/04/2019	SPC
Updated CPC search	02/25/2020	SPC
Updated CPC search	05/13/2020	SPC
Updated CPC search	06/22/2020	SPC

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**
429	231.3	03/07/2019	SPC
	Updated USPC search	06/27/2019	SPC
	Updated USPC search	12/04/2019	SPC
	Updated USPC search	02/25/2020	SPC
	Updated USPC search	05/13/2020	SPC
	Updated USPC search	06/22/2020	SPC

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
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	Examiner	Art Unit
	Sean P Cullen, Ph.D.	1725

Search Notes		
Search Notes	**Date**	**Examiner**
H01M4/131, 1315, 1391, 13915, 625, 8657	03/07/2019	SPC
429/231.3	03/07/2019	SPC
Inventor/applicant search, PALM	03/07/2019	SPC
EAST text search, see search history	03/07/2019	SPC
Updated inventor/applicant search, PALM	06/27/2019	SPC
Updated EAST text search, see search history	06/27/2019	SPC
Updated inventor/applicant search, PALM	12/04/2019	SPC
Updated EAST text search, see search history	12/04/2019	SPC
Updated inventor/applicant search, PALM	02/25/2020	SPC
Updated EAST text search, see search history	02/25/2020	SPC
Updated inventor/applicant search, PALM	05/13/2020	SPC
Updated EAST text search, see search history	05/13/2020	SPC
Updated inventor/applicant search, PALM	06/22/2020	SPC
Updated EAST text search, see search history	06/22/2020	SPC

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**
H01M4	131, 1315, 1391, 13915, 625, 8657	05/13/2020	SPC
429	231.3, 231.5, 231.6, 231.95	05/13/2020	SPC
	Updated interference search	06/22/2020	SPC

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	15/638,449	OCHIAI et al.
	Examiner	**Art Unit**
	Sean P Cullen, Ph.D.	1725

CPC								
Symbol							**Type**	**Version**
H01M	/	4	/	131			F	2013-01-01
H01M	/	4	/	1315			I	2013-01-01
H01M	/	4	/	1391			I	2013-01-01
H01M	/	4	/	625			I	2013-01-01
H01M	/	4	/	13915			I	2013-01-01
H01M	/	4	/	8657			I	2013-01-01
H01M	/	2004	/	021			A	2013-01-01
H01M	/	2004	/	028			A	2013-01-01
H01M	/	10	/	0525			A	2013-01-01

CPC Combination Sets								
Symbol					**Type**	**Set**	**Ranking**	**Version**
	/		/					

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	8	
/Sean P Cullen, Ph.D./ Primary Examiner, Art Unit 1725	22 June 2020	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	1C

U.S. Patent and Trademark Office — Part of Paper No.: 20200622

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	15/638,449	OCHIAI et al.
	Examiner	**Art Unit**
	Sean P Cullen, Ph.D.	1725

INTERNATIONAL CLASSIFICATION				
CLAIMED				
H01M4/131	/	4	/	131
H01M4/1315	/	4	/	1315
H01M4/1391	/	4	/	1391
H01M4/62	/	4	/	62
H01M4/13915	/	4	/	13915
H01M4/86	/	4	/	86
H01M4/02	/	4	/	02
H01M10/0525	/	10	/	0525

NON-CLAIMED				
	/		/	

US ORIGINAL CLASSIFICATION	
CLASS	**SUBCLASS**
429	231.3

CROSS REFERENCES(S)						
CLASS	**SUBCLASS (ONE SUBCLASS PER BLOCK)**					
429	231.5	231.6	231.95			

NONE		**Total Claims Allowed:**	
(Assistant Examiner)	(Date)	8	
/Sean P Cullen, Ph.D./ Primary Examiner, Art Unit 1725	22 June 2020	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	1C

U.S. Patent and Trademark Office — Part of Paper No.: 20200622

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	15/638,449	OCHIAI et al.
	Examiner	**Art Unit**
	Sean P Cullen, Ph.D.	1725

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIMS

Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original
1	60														
2	61														
3	62														
4	63														
5	56														
6	57														
7	58														
8	59														

NONE		**Total Claims Allowed:**	
(Assistant Examiner)	(Date)	8	
/Sean P Cullen, Ph.D./ Primary Examiner, Art Unit 1725	22 June 2020	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	1C

U.S. Patent and Trademark Office

Part of Paper No.: 20200622

Bibliographic Data

Application No: 15/638,449

Foreign Priority claimed: (•) Yes ( ) No

35 USC 119 (a-d) conditions met: [✓] Yes [ ] No [ ] Met After Allowance

Verified and Acknowledged: /Sean P Cullen, Ph.D./ Examiner's Signature | Initials

Title: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE

FILING or 371(c) DATE	CLASS	GROUP ART UNIT	ATTORNEY DOCKET NO.
06/30/2017	429	1725	12732-2003001
RULE			

APPLICANTS

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
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Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2004-342554A	12-02-2004			Full
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Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 2 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
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		US-2012/0295163	11-22-2012	Yanagita.H et al.	
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Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-05-314965A	11-26-1993			Abst.
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		JP-08-236114A	09-13-1996			Abst.
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		JP-2001-319692A	11-16-2001			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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Sheet 3 of 13

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Filing Date	June 30, 2017
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Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

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Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 4 of 13

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Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

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Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3] -Number[4] -Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		JP-2006-261132A	09-28-2006			Abst.
		JP-2007-128714A	05-24-2007			Abst.
		JP-2010-080407A	04-08-2010			Abst.
		JP-2010-102895A	05-06-2010			Full
		JP-2010-244847A	10-28-2010			Full
		JP-2010-272239A	12-02-2010			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 5 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-RE.43,276	03-27-2012	Kweon.H et al.	

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		JP-2011-138718A	07-14-2011			Full
		JP-2014-049239A	03-17-2014			Abst.
		JP-2014-063707A	04-10-2014			Abst.
		JP-2014-063708A	04-10-2014			Abst.
		JP-2015-099722A	05-28-2015			Abst.
		JP-2015-156363A	08-27-2015			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 6 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

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		JP-2016-076454A	05-12-2016			Abst.
		JP-2017-021942A	01-26-2017			Abst.
		JP-2017-091777A	05-25-2017			Full
		JP-2018-092934A	06-14-2018			Abst.
		JP-3031546	04-10-2000			Abst.
		JP-4739780	08-03-2011			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 7 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

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		WO-2012/029729	03-08-2012			Abst.
		WO-2014/098238	06-26-2014			Abst.
		WO-2018/211375	11-22-2018			Eng.
		CN-102694201A	09-26-2012			Full
		CN-102569775A	07-11-2012			Full

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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Sheet 8 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

NON PATENT LITERATURE DOCUMENTS

Examiner Initials*	Cite No.[1]	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published.	T[2]
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Sheet 9 of 13

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Filing Date	June 30, 2017
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Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 11 of 13

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

NON PATENT LITERATURE DOCUMENTS

Examiner Initials*	Cite No.[1]	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published.	T[2]
		Wang.Z et al., "Electrochemical Evaluation and Structural Characterization of Commercial LiCoO2 Surfaces Modified with MgO for Lithium-Ion Batteries", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), March 4, 2002, Vol. 149, No. 4, pp. A466-A471.	Eng.
		Lee.Y et al., "Phase Transitions of Bare and Coated LixCoO2 (x = 0.4 and 0.24) at 300°C", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), July 28, 2005, Vol. 152, No. 9, pp. A1824-A1827.	Eng.
		Geder.J et al., "Impact of active material surface area on thermal stability of LiCoO2 cathode", JOURNAL OF POWER SOURCES, July 1, 2014, Vol. 257, pp. 286-292, ELSEVIER.	Eng.
		Benecke.M et al., "Effect of LiF on Hot-Pressing of MgO", J. Am. Ceram. Soc. (JOURNAL OF THE AMERICAN CERAMIC SOCIETY), July 1, 1967, Vol. 50, No. 7, pp. 365-368.	Eng.
		Hart.P et al., "Densification Mechanisms in Hot-Pressing of Magnesia with a Fugitive Liquid", J. Am. Ceram. Soc. (JOURNAL OF THE AMERICAN CERAMIC SOCIETY), February 1, 1970, Vol. 53, No. 2, pp. 83-86.	Eng.
		Belsky.A et al., "New developments in the Inorganic Crystal Structure Database (ICSD): Accessibility in support of materials research and design.", ACTA. CRYST.(ACTA CRYSTALLOGRAPHICA SECTION B), June 1, 2002, Vol. B58, No. 3, pp. 364-369.	Eng.
		Alcantara.R et al., "SPES, 6Li MAS NMR, and Ni3+ EPR evidence for the formation of Co2+-containing spinel phases in LiCoO2 cycled electrode materials", Journal of Electroanalytical Chemistry, August 28, 1998, Vol. 454, No. 1-2, pp. 173-181.	Eng.
		Shi.S et al., "Enhanced cycling stability of Li[Li 0.2Mn0.54Ni0.13Co0.13]O2 by surface modification of MgO with melting impregnation method", Electrochimica Acta, November 1, 2012, Vol. 88, pp. 671-679.	Eng.
		Antaya.M et al., "Preparation and Characterization of LiCoO2 Thin Films by Laser Ablation Deposition", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), March 1, 1993, Vol. 140, No. 3, pp. 575-578.	Eng.
		Koyama.Y et al., "Co K-edge XANES of LiCoO2 and CoO2 with a variety of structures by supercell density functional calculations with a core hole", PHYS. REV. B (PHYSICAL REVIEW. B), February 27, 2012, Vol. 85, No. 7, pp. 075129-1-075129-7.	Eng.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.
1 Applicant's unique citation designation number (optional). 2 Applicant is to place a check mark here if English language Translation is attached.
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ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /SPC/

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Substitute for form 1449/PTO				Complete if Known	
INFORMATION DISCLOSURE STATEMENT BY APPLICANT				Application Number	15/638,449
				Filing Date	June 30, 2017
				First Named Inventor	Teruaki OCHIAI
(Use as many sheets as necessary)				Art Unit	1725
				Examiner Name	S. CULLEN
Sheet	12	of	13	Attorney Docket Number	12732-2003001

NON PATENT LITERATURE DOCUMENTS			
Examiner Initials*	Cite No.[1]	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published.	T[2]
		Quinlan.R et al., "XPS Investigation of the Electrolyte Induced Stabilization of LiCoO2 and "AlPO4"-Coated LiCoO2 Composite Electrodes", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), December 3, 2015, Vol. 163, No. 2, pp. A300-A308.	Eng.
		Cho.J et al., "Comparison of Al2O3- and AlPO4-coated LiCoO2 cathode materials for a Li-ion cell", JOURNAL OF POWER SOURCES, 2005, Vol. 146, pp. 58-64, ELSEVIER.	Eng.
		Jin.Y et al., "Electrochemical Characterizations of Commercial LiCoO2 Powders with Surface Modified by Li3PO4 Nanoparticles", ELECTROCHEMICAL AND SOLID-STATE LETTERS, April 3, 2006, Vol. 9, No. 6, pp. A273-A276.	Eng.
		Hong.W et al., "Modification of LiCoO2 by Surface Coating with MgO/TiO2/SiO2 for High-Performance Lithium-Ion Battery", ELECTROCHEMICAL AND SOLID-STATE LETTERS, December 20, 2005, Vol. 9, No. 2, pp. A82-A85.	Eng.
		Eom.J et al., "M3(PO4)2-Nanoparticle-Coated LiCoO2 vs LiCo0.96M0.04O2(M = Mg and Zn) on Electrochemical and Storage Characteristics", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), January 8, 2008, Vol. 155, No. 3, pp. A201-A205.	Eng.
		Kim.Y et al., "Suppression of Cobalt Dissolution from the LiCoO2 Cathodes with Various Metal-Oxide Coatings", J. ELECTROCHEM. SOC. (JOURNAL OF THE ELECTROCHEMICAL SOCIETY), 2003, Vol. 150, No. 12, pp. A1723-A1725.	Eng.
		Fang.T et al., "Effect of calcination temperature on the electrochemical behavior of ZnO-coated LiCoO2 cathode", Surface & Coatings Technology , April 19, 2006, Vol. 201, No. 3-4, pp. 1886-1893, ELSEVIER.	Eng.
		Park.S et al., "Novel Surface Modification Technique to Improve Electrochemical Performance of LiCoO2 at High Voltage", ELECTROCHEMICAL AND SOLID-STATE LETTERS, April 21, 2005, Vol. 8, No. 6, pp. A299-A302.	Eng.
		Jo.C et al., "An effective method to reduce residual lithium compounds on Ni-rich Li[Ni0.6Co0.2Mn0.2]O2 active material using a phosphoric acid derived Li3PO4 nanolayer", Nano Research, December 4, 2014, Vol. 8, No. 5, pp. 1464-1479, Springer.	Eng.
		Tebbe.J et al., "Mechanisms of LiCoO2 Cathode Degradation by Reaction with HF and Protection by Thin Oxide Coatings", ACS APPLIED MATERIALS & INTERFACES, October 12, 2015, Vol. 7, No. 43, pp. 24265-24278.	Eng.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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1 Applicant's unique citation designation number (optional). 2 Applicant is to place a check mark here if English language Translation is attached.
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Sheet 13 of 13

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First Named Inventor	Teruaki OCHIAI
Art Unit	1725
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Attorney Docket Number	12732-2003001

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		Bensalah.N et al., "Review on Synthesis, Characterizations, and Electrochemical Properties of Cathode Materials for Lithium Ion Batteries", Journal of Material Science & Engineering, 2016, Vol. 5, No. 4, pp. 1000258-1-1000258-21.	Eng.
		Lu.Y et al., "Recent progress on lithium-ion batteries with high electrochemical performance", SCIENCE CHINA CHEMISTRY, February 25, 2019, Vol. 62, No. 5, pp. 533-548.	Eng.
		Xiao.B, "Surface Modifification of Electrode Materials for Lithium-Ion Batteries", Graduate Program in Mechanical and Materials Engineering, 2006, pp. 1-235.	Eng.
		Ohnishi.M et al., "Investigation of the surface degradation of LiCoO2 particles in the cathode materials of Li-ion batteries using FIB-TOF-SIMS", Journal of Surface Analysis, 2013, Vol. 20, No. 2, pp. 99-110.	Eng.
		Iwaya.K et al., "Impact of Lithium-Ion Ordering on Surface Electronic States of LixCoO2", PHYS. REV. LETT. (PHYSICAL REVIEW LETTERS), September 20, 2013, Vol. 111, No. 12, pp. 126104-1-126104-5.	Eng.
		Zou.M et al., "Synthesis of High-Voltage (4.5 V) Cycling Doped LiCoO2 for Use in Lithium Rechargeable Cells", CHEM. MATER. (CHEMISTRY OF MATERIALS), December 16, 2003, Vol. 15, No. 25, pp. 4699-4702.	Eng.
		Schipper.F et al., "Study of Cathode Materials for Lithium-Ion Batteries: Recent Progress and New Challenges", Inorganics, April 28, 2017, Vol. 5, No. 2, pp. 1-29.	Eng.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/22/2020

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Attorney Docket: 12732-2003001 / US24652/.../26813

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor	:	Teruaki OCHIAI	Art Unit	:	1725
Application No.	:	15/638,449	Examiner	:	Sean P. Cullen
Filed	:	June 30, 2017	Conf. No.	:	1074
Title	:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE (AS AMENDED)			

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

INFORMATION DISCLOSURE STATEMENT

Please consider the references listed on the enclosed disclosure form. Copies of the documents required under 37 C.F.R. § 1.98(a)(2) are being submitted with this statement.

This statement is being filed with a Request for Continued Examination. No fee is required. Please apply any charges or credits to Deposit Account No. 06-1050, referencing the above attorney docket number.

Respectfully submitted,

Date: June 19, 2020

/Heather L. Flanagan/
Heather L. Flanagan
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ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /SPC/

PE2E SEARCH - Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	British Equivalents	Time Stamp
L169	345	(IN-726813-$ | CN-102569775-$ | CN-102610806-$ | CN-102694201-$ | JP-05314965-$ | JP-05314995-$ | JP-08236114-$ | JP-11096993-$ | JP-2000203842-$ | JP-2001319692-$ | JP-2002216760-$ | JP-2002352802-$ | JP-2003221235-$ | JP-2004014381-$ | JP-2004342554-$ | JP-2005158612-$ | JP-2005302510-$ | JP-2006164758-$ | JP-2006261132-$ | JP-2007128714-$ | JP-2007213866-$ | JP-2010080407-$ | JP-2010102895-$ | JP-2010244847-$ | JP-2010272239-$ | JP-2011138718-$ | JP-2012209077-$ | JP-2014049239-$ | JP-2014063707-$ | JP-2014063708-$ | JP-2015069958-$ | JP-2015099722-$ | JP-2015156363-$ | JP-2016076454-$ | JP-2017021942-$ | JP-2017091777-$ | JP-2018092934-$ | JP-3031546-$ | JP-4739780-$ | VA-223131450-$ | WO-2012029729-$ | WO-2014098238-$ | WO-2018211375-$).DID. (US-0005364-$ | US-0019194-$ | US-0019462-$ | US-0019662-$ | US-0028080-$ | US-0040193-$ | US-0040594-$ | US-0045692-$ | US-0059367-$ | US-0064406-$ | US-0069907-$ | US-	US-PGPUB; USPAT; USOCR; FIT (CN, FR, WO); FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	ON	2020/06/22 11:02 AM

		0076489-$ | US- 0078516-$ | US- 0079995-$ | US- 0099554-$ | US- 0102536-$ | US- 0104279-$ | US- 0104532-$ | US- 0108944-$ | US- 0110736-$ | US- 0117014-$ | US- 0117589-$ | US- 0123050-$ | US- 0127567-$ | US- 0144601-$ | US- 0145317-$ | US- 0145368-$ | US- 0152961-$ | US- 0176770-$ | US- 0229124-$ | US- 0241693-$ | US- 0244430-$ | US- 0247986-$ | US- 0256437-$ | US- 0256817-$ | US- 0261622-$ | US- 0262762-$ | US- 0266858-$ | US- 0276658-$ | US- 0285102-$ | US- 0295163-$ | US- 0297876-$ | US- 0309910-$ | US- 0313471-$ | US- 0329533-$ | US- 0330044-$ | US- 0333324-$ | US- 0349905-$ | US- 0366729-$ | US- 0380271-$ | US- 0380737-$ | US- 10128495-$ | US- 10243215-$ | US- 10361432-$ | US- 1000258-$ | US- 1400195-$ | US- 2003001-$ | US- 3031546-$ | US- 4739780-$ | US- 5705291-$ | US- 6218050-$ | US- 6737195-$ | US- 6919144-$ | US- 6974601-$ | US- 7018741-$ | US- 7294435-$ | US- 7309546-$ | US- 8007941-$ | US- 8034486-$ | US- 8236449-$ | US- 8808918-$ | US- 8877377-$ | US-					

		8906547-$ | US- 9293236-$ | US- 9362557-$ | US- 9505631-$ | US- 9515313-$ | US- 9666326-$ | US- 9786903-$ | US- 9899664-$ | US- 9923244-$ | US- 20020110736-$ | US- 20030104279-$ | US- 20040229124-$ | US- 20050019662-$ | US- 20070117014-$ | US- 20080241693-$ | US- 20090104532-$ | US- 20100019194-$ | US- 20100247986-$ | US- 20110059367-$ | US- 20110256437-$ | US- 20110297876-$ | US- 20120045692-$ | US- 20120064406-$ | US- 20120244430-$ | US- 20120261622-$ | US- 20120295163-$ | US- 20120330044-$ | US- 20130040193-$ | US- 20130078516-$ | US- 20130266858-$ | US- 20130313471-$ | US- 20140079995-$ | US- 20140099554-$ | US- 20140127567-$ | US- 20150123050-$ | US- 20150262762-$ | US- 20150333324-$ | US- 20150380737-$ | US- 20160028080-$ | US- 20160276658-$ | US- 20160285102-$ | US- 20160329533-$ | US- 20160349905-$ | US- 20160380271-$ | US- 20170005364-$ | US- 20170040594-$ | US- 20170069907-$ | US- 20170117589-$ | US- 20170256817-$ | US- 20170309910-$ | US- 20180019462-$ | US- 20180076489-$ | US- 20180102536-$ | US- 20180108944-$ | US- 20180145317-$ | US- 20180145368-$ | US- 20180366729-$ | US- 20200144601-$ | US- 20200152961-$ | US- 20200176770-$).DID.					

L170	2	L169 AND (crack crystal near2 defect) WITH (fluorine fluoride F!)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	ON	2020/06/22 11:03 AM
L171	23	L169 AND (crack crystal near2 defect)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	ON	2020/06/22 11:05 AM
L172	3	L169 AND (crack breakag defect) WITH (fluorine fluoride F!)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	ON	2020/06/22 11:52 AM
L173	128	((H01M4/131,1315,1391, 13915,625,8657).cpc. (429/231.3,231.5,231.6,231.95).ccls.) AND (crack breakage defect) WITH (Mg magnesium fluorine F MgF.sub.2 MgF2)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	ON	2020/06/22 11:53 AM
L174	0	L173 NOT L156	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	ON	2020/06/22 11:54 AM

PE2E SEARCH - Search History (Interference)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	British Equivalents	Time Stamp
N168	107	((H01M4/131,1315,1391, 13915,625,8657).cpc. (429/231.3,231.5,231.6,231.95).ccls.) AND (crack breakage defect) WITH (Mg magnesium fluorine F MgF.sub.2 MgF2)	US-PGPUB; USPAT	OR	ON	ON	2020/06/22 11:54 AM
N169	0	N168 NOT N163	US-PGPUB; USPAT	OR	ON	ON	2020/06/22 11:54 AM

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Sheet 1 of 1

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Art Unit	1725
Examiner Name	S. CULLEN
Attorney Docket Number	12732-2003001

U. S. PATENT DOCUMENTS

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		JP-2012-018914A	01-26-2012			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	06/23/2020

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Respectfully submitted,

Date: June 22, 2020

/Heather L. Flanagan/
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MAIL STOP ISSUE FEE
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

REPLY TO NOTICE OF ALLOWANCE

In response to the Notice of Allowance mailed June 25, 2020, enclosed is a completed issue fee transmittal form PTOL 85b. A fee in the amount of $1000 for the issue fee is being paid concurrently herewith on the Electronic Filing System (EFS) by way of Deposit Account authorization.

Please apply any other charges or credits to Deposit Account No. 06-1050, referencing attorney docket number 12732-2003001.

Respectfully submitted,

Date: July 1, 2020

/John F. Hayden/
John F. Hayden
Reg. No. 37640

Customer Number 26171
Fish & Richardson P.C.
Telephone: (202) 783-5070
Facsimile: (877) 769-7945

41467178.doc

PART B - FEE(S) TRANSMITTAL

Complete and send this form, together with applicable fee(s), by mail or fax, or via EFS-Web

By mail, send to: Mail Stop ISSUE FEE
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22314-1450

By fax, send to: (571) 273-2885

INSTRUCTIONS: This form should be used for transmitting the ISSUE FEE and PUBLICATION FEE (if required). Blocks 1 through 5 should be completed where appropriate. All further correspondence including the Patent, advance orders and notification of maintenance fees will be mailed to the current correspondence address as indicated unless corrected below or directed otherwise in Block 1, by (a) specifying a new correspondence address; and/or (b) indicating a separate "FEE ADDRESS" for maintenance fee notifications.

CURRENT CORRESPONDENCE ADDRESS (Note: Use Block 1 for any change of address)

26171 7590 06/25/2020

FISH & RICHARDSON P.C.
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

Note: A certificate of mailing can only be used for domestic mailings of the Fee(s) Transmittal. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawing, must have its own certificate of mailing or transmission.

Certificate of Mailing or Transmission

I hereby certify that this Fee(s) Transmittal is being deposited with the United States Postal Service with sufficient postage for first class mail in an envelope addressed to the Mail Stop ISSUE FEE address above, or being transmitted to the USPTO via EFS-Web or by facsimile to (571) 273-2885, on the date below.

(Typed or printed name)
(Signature)
(Date)

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	06/30/2017	Teruaki OCHIAI	12732-2003001	1074

TITLE OF INVENTION: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE (AS AMENDED)

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	LARGE	$1000	$0		$1000	09/25/2020

EXAMINER	ART UNIT	CLASS-SUBCLASS
CULLEN, SEAN P.	1725	429-231300

1. Change of correspondence address or indication of "Fee Address" (37 CFR 1.363).

[ ] Change of correspondence address (or Change of Correspondence Address form PTO/SB/122) attached.

[ ] "Fee Address" indication (or "Fee Address" Indication form PTO/SB/47; Rev 03-09 or more recent) attached. **Use of a Customer Number is required.**

2. For printing on the patent front page, list

(1) The names of up to 3 registered patent attorneys or agents OR, alternatively,

(2) The name of a single firm (having as a member a registered attorney or agent) and the names of up to 2 registered patent attorneys or agents. If no name is listed, no name will be printed.

1 Fish & Richardson P.C.

2

3

3. ASSIGNEE NAME AND RESIDENCE DATA TO BE PRINTED ON THE PATENT (print or type)

PLEASE NOTE: Unless an assignee is identified below, no assignee data will appear on the patent. If an assignee is identified below, the document must have been previously recorded, or filed for recordation, as set forth in 37 CFR 3.11 and 37 CFR 3.81(a). Completion of this form is NOT a substitute for filing an assignment.

(A) NAME OF ASSIGNEE: Semiconductor Energy Laboratory Co., Ltd.

(B) RESIDENCE: (CITY and STATE OR COUNTRY): Atsugi-shi, Kanagawa-ken, JAPAN

Please check the appropriate assignee category or categories (will not be printed on the patent): [ ] Individual [X] Corporation or other private group entity [ ] Government

4a. Fees Submitted: [X] Issue Fee [ ] Publication Fee (if required) [ ] Advance Order - # of Copies

4b. Method of Payment (*Please first reapply any previously paid fee shown above*):

[X] Electronic Payment via EFS-Web [ ] Enclosed check [ ] Non-electronic payment by credit card (Attach form PTO-2038)

[X] The Director is hereby authorized to charge the required fee(s), any deficiency, or credit any overpayment, to Deposit Account No. 06-1050

5. **Change of Entity Status** (from status indicated above)

[ ] Applicant certifying micro entity status. See 37 CFR 1.29 — **NOTE**: Absent a valid Certification of Micro Entity Status (see forms PTO/SB/15A and 15B), issue fee payment in the micro entity amount will not be accepted at the risk of application abandonment.

[ ] Applicant asserting small entity status. See 37 CFR 1.27. — **NOTE**: If the application was previously under micro entity status, checking this box will be taken as a notification of loss of entitlement to micro entity status.

[ ] Applicant changing to regular undiscounted fee status. — **NOTE**: Checking this box will be taken as a notification of loss of entitlement to small or micro entity status, as applicable.

NOTE: This form must be signed in accordance with 37 CFR 1.31 and 1.33. See 37 CFR 1.4 for signature requirements and certifications.

Authorized Signature /John F. Hayden/ Date July 1, 2020

Typed or printed name John F. Hayden Registration No. 37640

41467174.doc

Electronic Patent Application Fee Transmittal

Application Number:	15638449
Filing Date:	30-Jun-2017
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Filer:	John F. Hayden/April Saunders-Fuller
Attorney Docket Number:	12732-2003001

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
UTILITY APPL ISSUE FEE	1501	1	1000	1000

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Extension-of-Time:				
Miscellaneous:				
			Total in USD ($)	**1000**

Electronic Acknowledgement Receipt

EFS ID:	39887932
Application Number:	15638449
International Application Number:	
Confirmation Number:	1074
Title of Invention:	POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE
First Named Inventor/Applicant Name:	Teruaki OCHIAI
Customer Number:	26171
Filer:	John F. Hayden/April Saunders-Fuller
Filer Authorized By:	John F. Hayden
Attorney Docket Number:	12732-2003001
Receipt Date:	01-JUL-2020
Filing Date:	30-JUN-2017
Time Stamp:	15:13:21
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	DA
Payment was successfully received in RAM	$1000
RAM confirmation Number	E202071F14058256
Deposit Account	061050
Authorized User	April Saunders-Fuller

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

37 CFR 1.17 (Patent application and reexamination processing fees)

37 CFR 1.19 (Document supply fees)

37 CFR 1.21 (Miscellaneous fees and charges)

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Transmittal Letter	12732-2003001-REPLY.pdf	56842 5c43823958dada0ed2533831214e5a0bd043cfd1	no	1
Warnings:					
Information:					
2	Issue Fee Payment (PTO-85B)	12732-2003001-IF.pdf	123134 5aef2b8d6ded5b8de4e140e2661fafafbd43badf	no	1
Warnings:					
Information:					
3	Fee Worksheet (SB06)	fee-info.pdf	30713 861c28679a4b4f32b50de6154f8186c10fea9344	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	210689		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

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APPLICATION NO.	ISSUE DATE	PATENT NO.	ATTORNEY DOCKET NO.	CONFIRMATION NO.
15/638,449	08/11/2020	10741828	12732-2003001	1074

26171 7590 07/22/2020

FISH & RICHARDSON P.C. (DC)
P.O. BOX 1022
MINNEAPOLIS, MN 55440-1022

ISSUE NOTIFICATION

The projected patent number and issue date are specified above.

Determination of Patent Term Adjustment under 35 U.S.C. 154 (b)
(application filed on or after May 29, 2000)

The Patent Term Adjustment is 62 day(s). Any patent to issue from the above-identified application will include an indication of the adjustment on the front page.

If a Continued Prosecution Application (CPA) was filed in the above-identified application, the filing date that determines Patent Term Adjustment is the filing date of the most recent CPA.

Applicant will be able to obtain more detailed information by accessing the Patent Application Information Retrieval (PAIR) WEB site (http://pair.uspto.gov).

Any questions regarding the Patent Term Extension or Adjustment determination should be directed to the Office of Patent Legal Administration at (571)-272-7702. Questions relating to issue and publication fee payments should be directed to the Application Assistance Unit (AAU) of the Office of Data Management (ODM) at (571)-272-4200.

APPLICANT(s) (Please see PAIR WEB site http://pair.uspto.gov for additional applicants):

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Ayae TSURUTA, Atsugi, JAPAN;

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