【０２３８】
粒径の小さい活物質、例えば１μｍ以下の活物質を用いる場合には、活物質の比表面積が大きく、活物質同士を繋ぐ導電パスがより多く必要となる。そのため導電助剤の量が多くなりがちであり、相対的に活物質の担持量が減少してしまう傾向がある。活物質の担持量が減少すると、二次電池の容量が減少してしまう。このような場合には、導電助剤としてグラフェン化合物を用いると、グラフェン化合物は少量でも効率よく導電パスを形成することができるため、活物質の担持量を減らさずに済み、特に好ましい。
【０２３９】
以下では一例として、活物質層２００に、導電助剤としてグラフェン化合物を用いる場合の断面構成例を説明する。
【０２４０】
図７（Ａ）に、活物質層２００の縦断面図を示す。活物質層２００は、粒状の正極活物質１００と、導電助剤としてのグラフェン化合物２０１と、バインダ（図示せず）と、を含む。ここで、グラフェン化合物２０１として例えばグラフェンまたはマルチグラフェンを用いればよい。ここで、グラフェン化合物２０１はシート状の形状を有することが好ましい。また、グラフェン化合物２０１は、複数のマルチグラフェン、または（および）複数のグラフェンが部分的に重なりシート状となっていてもよい。
【０２４１】
活物質層２００の縦断面においては、図７（Ｂ）に示すように、活物質層２００の内部において概略均一にシート状のグラフェン化合物２０１が分散する。図７（Ｂ）においてはグラフェン化合物２０１を模式的に太線で表しているが、実際には炭素分子の単層又は多層の厚みを有する薄膜である。複数のグラフェン化合物２０１は、複数の粒状の正極活物質１００を一部覆うように、あるいは複数の粒状の正極活物質１００の表面上に張り付くように形成されているため、互いに面接触している。
【０２４２】
ここで、複数のグラフェン化合物同士が結合することにより、網目状のグラフェン化合物シート（以下グラフェン化合物ネットまたはグラフェンネットと呼ぶ）を形成することができる。活物質をグラフェンネットが被覆する場合に、グラフェンネットは活物質同士を結合するバインダとしても機能することができる。よって、バインダの量を少なくすることができる、又は使用しないことができるため、電極体積や電極重量に占める活物質の比率を向上させることができる。すなわち、二次電池の容量を増加させることができる。
【０２４３】
ここで、グラフェン化合物２０１として酸化グラフェンを用い、活物質と混合して活物質層２００となる層を形成後、還元することが好ましい。グラフェン化合物２０１の形成に、極性溶媒中での分散性が極めて高い酸化グラフェンを用いることにより、グラフェン化合物２０１を活物質層２００の内部において概略均一に分散させることができる。均一に分散した酸化グラフェンを含有する分散媒から溶媒を揮発除去し、酸化グラフェンを還元するため、活物質層２００に残留するグラフェン化合物２０１は部分的に重なり合い、互いに面接触する程度に分散していることで三次元的な導電パスを形成することができる。なお、酸化グラフェンの還元は、例えば熱処理により行ってもよいし、還元剤を用いて行ってもよい。
【０２４４】
従って、活物質と点接触するアセチレンブラック等の粒状の導電助剤と異なり、グラフェン化合物２０１は接触抵抗の低い面接触を可能とするものであるから、通常の導電助剤よりも少量で粒状の正極活物質１００とグラフェン化合物２０１との電気伝導性を向上させることができる。よって、正極活物質１００の活物質層２００における比率を増加させることができる。これにより、二次電池の放電容量を増加させることができる。
【０２４５】
また、予め、スプレードライ装置を用いてグラフェン化合物で活物質の表面全体を覆ってもよい。その後正極活物質層を作製するときにさらにグラフェン化合物を加えて、活物質

同士間の導電パスをより良好にすることもできる。
【０２４６】
バインダとしては、例えば、スチレン―ブタジエンゴム（ＳＢＲ）、スチレン―イソプレン―スチレンゴム、アクリロニトリル―ブタジエンゴム、ブタジエンゴム、エチレン―プロピレン―ジエン共重合体などのゴム材料を用いることが好ましい。またバインダとして、フッ素ゴムを用いることができる。
【０２４７】
また、バインダとしては、例えば水溶性の高分子を用いることが好ましい。水溶性の高分子としては、例えば多糖類などを用いることができる。多糖類としては、カルボキシメチルセルロース（ＣＭＣ）、メチルセルロース、エチルセルロース、ヒドロキシプロピルセルロース、ジアセチルセルロース、再生セルロースなどのセルロース誘導体や、澱粉などを用いることができる。また、これらの水溶性の高分子を、前述のゴム材料と併用して用いると、さらに好ましい。
【０２４８】
または、バインダとしては、ポリスチレン、ポリアクリル酸メチル、ポリメタクリル酸メチル（ポリメチルメタクリレート（ＰＭＭＡ））、ポリアクリル酸ナトリウム、ポリビニルアルコール（ＰＶＡ）、ポリエチレンオキシド（ＰＥＯ）、ポリプロピレンオキシド、ポリイミド、ポリ塩化ビニル、ポリテトラフルオロエチレン、ポリエチレン、ポリプロピレン、ポリイソブチレン、ポリエチレンテレフタレート、ナイロン、ポリフッ化ビニリデン（ＰＶｄＦ）、ポリアクリロニトリル（ＰＡＮ）、エチレンプロピレンジエンポリマー、ポリ酢酸ビニル、ニトロセルロース等の材料を用いることが好ましい。
【０２４９】
バインダは上記のうち複数を組み合わせて使用してもよい。
【０２５０】
例えば粘度調整効果の特に優れた材料と、他の材料とを組み合わせて使用してもよい。例えばゴム材料等は接着力や弾性力に優れる反面、溶媒に混合した場合に粘度調整が難しい場合がある。このような場合には例えば、粘度調整効果の特に優れた材料と混合することが好ましい。粘度調整効果の特に優れた材料としては、例えば水溶性高分子を用いるとよい。また、粘度調整効果に特に優れた水溶性高分子としては、前述の多糖類、例えばカルボキシメチルセルロース（ＣＭＣ）、メチルセルロース、エチルセルロース、ヒドロキシプロピルセルロースおよびジアセチルセルロース、再生セルロースなどのセルロース誘導体や、澱粉を用いることができる。
【０２５１】
なお、カルボキシメチルセルロースなどのセルロース誘導体は、例えばカルボキシメチルセルロースのナトリウム塩やアンモニウム塩などの塩とすることにより溶解度が上がり、粘度調整剤としての効果を発揮しやすくなる。溶解度が高くなることにより電極のスラリーを作製する際に活物質や他の構成要素との分散性を高めることもできる。本明細書においては、電極のバインダとして使用するセルロースおよびセルロース誘導体としては、それらの塩も含むものとする。
【０２５２】
水溶性高分子は水に溶解することにより粘度を安定化させ、また活物質や、バインダとして組み合わせる他の材料、例えばスチレンブタジエンゴムなどを、水溶液中に安定して分散させることができる。また、官能基を有するために活物質表面に安定に吸着しやすいことが期待される。また、例えばカルボキシメチルセルロースなどのセルロース誘導体は、例えば水酸基やカルボキシル基などの官能基を有する材料が多く、官能基を有するために高分子同士が相互作用し、活物質表面を広く覆って存在することが期待される。
【０２５３】
活物質表面を覆う、または表面に接するバインダが膜を形成する場合には、不動態膜としての役割を果たして電解液の分解を抑える効果も期待される。ここで、不動態膜とは、電子の伝導性のない膜、または電気伝導性の極めて低い膜であり、例えば活物質の表面に不

動態膜が形成された場合には、電池反応電位において、電解液の分解を抑制することができる。また、不動態膜は、電気の伝導性を抑えるとともに、リチウムイオンは伝導できるとさらに望ましい。

【０２５４】

＜正極集電体＞

正極集電体としては、ステンレス、金、白金、アルミニウム、チタン等の金属、及びこれらの合金など、導電性が高い材料をもちいることができる。また正極集電体に用いる材料は、正極の電位で溶出しないことが好ましい。また、シリコン、チタン、ネオジム、スカンジウム、モリブデンなどの耐熱性を向上させる元素が添加されたアルミニウム合金を用いることができる。また、シリコンと反応してシリサイドを形成する金属元素で形成してもよい。シリコンと反応してシリサイドを形成する金属元素としては、ジルコニウム、チタン、ハフニウム、バナジウム、ニオブ、タンタル、クロム、モリブデン、タングステン、コバルト、ニッケル等がある。集電体は、箔状、板状（シート状）、網状、パンチングメタル状、エキスパンドメタル状等の形状を適宜用いることができる。集電体は、厚みが５μｍ以上３０μｍ以下のものを用いるとよい。

【０２５５】

［負極］

負極は、負極活物質層および負極集電体を有する。また、負極活物質層は、導電助剤およびバインダを有していてもよい。

【０２５６】

＜負極活物質＞

負極活物質としては、例えば合金系材料や炭素系材料等を用いることができる。

【０２５７】

負極活物質として、リチウムとの合金化・脱合金化反応により充放電反応を行うことが可能な元素を用いることができる。例えば、シリコン、スズ、ガリウム、アルミニウム、ゲルマニウム、鉛、アンチモン、ビスマス、銀、亜鉛、カドミウム、インジウム等のうち少なくとも一つを含む材料を用いることができる。このような元素は炭素と比べて容量が大きく、特にシリコンは理論容量が４２００ｍＡｈ／ｇと高い。このため、負極活物質にシリコンを用いることが好ましい。また、これらの元素を有する化合物を用いてもよい。例えば、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、ＳｂＳｎ等がある。ここで、リチウムとの合金化・脱合金化反応により充放電反応を行うことが可能な元素、および該元素を有する化合物等を合金系材料と呼ぶ場合がある。

【０２５８】

本明細書等において、ＳｉＯは例えば一酸化シリコンを指す。あるいはＳｉＯは、SiO_xと表すこともできる。ここでｘは１近傍の値を有することが好ましい。例えばｘは、０．２以上１．５以下が好ましく、０．３以上１．２以下がより好ましい。

【０２５９】

炭素系材料としては、黒鉛、易黒鉛化性炭素（ソフトカーボン）、難黒鉛化性炭素（ハードカーボン）、カーボンナノチューブ、グラフェン、カーボンブラック等を用いればよい。

【０２６０】

黒鉛としては、人造黒鉛や、天然黒鉛等が挙げられる。人造黒鉛としては例えば、メソカーボンマイクロビーズ（ＭＣＭＢ）、コークス系人造黒鉛、ピッチ系人造黒鉛等が挙げられる。ここで人造黒鉛として、球状の形状を有する球状黒鉛を用いることができる。例えば、ＭＣＭＢは球状の形状を有する場合があり、好ましい。また、ＭＣＭＢはその表面積を小さくすることが比較的容易であり、好ましい場合がある。天然黒鉛としては例えば、鱗片状黒鉛、球状化天然黒鉛等が挙げられる。

【０２６１】
黒鉛はリチウムイオンが黒鉛に挿入されたとき（リチウム―黒鉛層間化合物の生成時）にリチウム金属と同程度に低い電位を示す（０．０５Ｖ以上０．３Ｖ以下　ｖｓ．Ｌｉ／Ｌｉ$^{+}$）。これにより、リチウムイオン二次電池は高い作動電圧を示すことができる。さらに、黒鉛は、単位体積当たりの容量が比較的高い、体積膨張が比較的小さい、安価である、リチウム金属に比べて安全性が高い等の利点を有するため、好ましい。
【０２６２】
また、負極活物質として、二酸化チタン（Ｔｉ●$_{2}$）、リチウムチタン酸化物（$Li_4Ti_5O_{12}$）、リチウム―黒鉛層間化合物（Li_xC_6）、五酸化ニオブ（Nb_2O_5）、酸化タングステン（WO_2）、酸化モリブデン（MoO_2）等の酸化物を用いることができる。
【０２６３】
また、負極活物質として、リチウムと遷移金属の複窒化物である、Li_3N型構造をもつ$Li_{3-x}M_xN$（Ｍ＝Ｃｏ、Ｎｉ、Ｃｕ）を用いることができる。例えば、$Li_{2.6}Co_{0.4}N_3$は大きな充放電容量（９００ｍＡｈ／ｇ、１８９０ｍＡｈ／ｃｍ^{3}）を示し好ましい。
【０２６４】
リチウムと遷移金属の複窒化物を用いると、負極活物質中にリチウムイオンを含むため、正極活物質としてリチウムイオンを含まないV_2O_5、Cr_3O_8等の材料と組み合わせることができ好ましい。なお、正極活物質にリチウムイオンを含む材料を用いる場合でも、あらかじめ正極活物質に含まれるリチウムイオンを脱離させることで、負極活物質としてリチウムと遷移金属の複窒化物を用いることができる。
【０２６５】
また、コンバージョン反応が生じる材料を負極活物質として用いることもできる。例えば、酸化コバルト（ＣｏＯ）、酸化ニッケル（ＮｉＯ）、酸化鉄（ＦｅＯ）等の、リチウムとの合金を作らない遷移金属酸化物を負極活物質に用いてもよい。コンバージョン反応が生じる材料としては、さらに、Fe_2O_3、ＣｕＯ、Cu_2O、RuO_2、Cr_2O_3等の酸化物、$CoS_{0.89}$、ＮｉＳ、ＣｕＳ等の硫化物、Zn_3N_2、Cu_3N、Ge_3N_4等の窒化物、NiP_2、FeP_2、CoP_3等のリン化物、FeF_3、BiF_3等のフッ化物でも起こる。
【０２６６】
負極活物質層が有することのできる導電助剤およびバインダとしては、正極活物質層が有することのできる導電助剤およびバインダと同様の材料を用いることができる。
【０２６７】
＜負極集電体＞
負極集電体には、正極集電体と同様の材料を用いることができる。なお負極集電体は、リチウム等のキャリアイオンと合金化しない材料を用いることが好ましい。
【０２６８】
［電解液］
電解液は、溶媒と電解質を有する。電解液の溶媒としては、非プロトン性有機溶媒が好ましく、例えば、エチレンカーボネート（ＥＣ）、プロピレンカーボネート（ＰＣ）、ブチレンカーボネート、クロロエチレンカーボネート、ビニレンカーボネート、γ―ブチロラクトン、γ―バレロラクトン、ジメチルカーボネート（ＤＭＣ）、ジエチルカーボネート（ＤＥＣ）、エチルメチルカーボネート（ＥＭＣ）、ギ酸メチル、酢酸メチル、酢酸エチル、プロピオン酸メチル、プロピオン酸エチル、プロピオン酸プロピル、酪酸メチル、１，３―ジオキサン、１，４―ジオキサン、ジメトキシエタン（ＤＭＥ）、ジメチルスルホキシド、ジエチルエーテル、メチルジグライム、アセトニトリル、ベンゾニトリル、テトラヒドロフラン、スルホラン、スルトン等の１種、又はこれらのうちの２種以上を任意の組み合わせおよび比率で用いることができる。
【０２６９】

また、電解液の溶媒として、難燃性および難揮発性であるイオン液体（常温溶融塩）を一つ又は複数用いることで、二次電池の内部短絡や、過充電等によって内部温度が上昇しても、二次電池の破裂や発火などを防ぐことができる。イオン液体は、カチオンとアニオンからなり、有機カチオンとアニオンとを含む。電解液に用いる有機カチオンとして、四級アンモニウムカチオン、三級スルホニウムカチオン、および四級ホスホニウムカチオン等の脂肪族オニウムカチオンや、イミダゾリウムカチオンおよびピリジニウムカチオン等の芳香族カチオンが挙げられる。また、電解液に用いるアニオンとして、１価のアミド系アニオン、１価のメチド系アニオン、フルオロスルホン酸アニオン、パーフルオロアルキルスルホン酸アニオン、テトラフルオロボレートアニオン、パーフルオロアルキルボレートアニオン、ヘキサフルオロホスフェートアニオン、またはパーフルオロアルキルホスフェートアニオン等が挙げられる。

【０２７０】

また、上記の溶媒に溶解させる電解質としては、例えば$LiPF_6$、$LiClO_4$、$LiAsF_6$、$LiBF_4$、$LiAlCl_4$、ＬｉＳＣＮ、ＬｉＢｒ、ＬｉＩ、Li_2SO_4、$Li_2B_{10}Cl_{10}$、$Li_2B_{12}Cl_{12}$、$LiCF_3SO_3$、$LiC_4F_9SO_3$、$LiC(CF_3SO_2)_3$、$LiC(C_2F_5SO_2)_3$、$LiN(CF_3SO_2)_2$、$LiN(C_4F_9SO_2)(CF_3SO_2)$、$LiN(C_2F_5SO_2)_2$等のリチウム塩を一種、又はこれらのうちの二種以上を任意の組み合わせおよび比率で用いることができる。

【０２７１】

二次電池に用いる電解液は、粒状のごみや電解液の構成元素以外の元素（以下、単に「不純物」ともいう。）の含有量が少ない高純度化された電解液を用いることが好ましい。具体的には、電解液に対する不純物の重量比を１％以下、好ましくは０．１％以下、より好ましくは０．０１％以下とすることが好ましい。

【０２７２】

また、電解液にビニレンカーボネート、プロパンスルトン（ＰＳ）、ｔｅｒｔ―ブチルベンゼン（ＴＢＢ）、フルオロエチレンカーボネート（ＦＥＣ）、ＬｉＢＯＢ、またスクシノニトリル、アジポニトリル等のジニトリル化合物などの添加剤を添加してもよい。添加する材料の濃度は、例えば溶媒全体に対して０．１ｗｔ％以上５ｗｔ％以下とすればよい。

【０２７３】

また、ポリマーを電解液で膨潤させたポリマーゲル電解質を用いてもよい。

【０２７４】

ポリマーゲル電解質を用いることで、漏液性等に対する安全性が高まる。また、二次電池の薄型化および軽量化が可能である。

【０２７５】

ゲル化されるポリマーとして、シリコーンゲル、アクリルゲル、アクリロニトリルゲル、ポリエチレンオキサイド系ゲル、ポリプロピレンオキサイド系ゲル、フッ素系ポリマーのゲル等を用いることができる。

【０２７６】

ポリマーとしては、例えばポリエチレンオキシド（ＰＥＯ）などのポリアルキレンオキシド構造を有するポリマーや、ＰＶＤＦ、およびポリアクリロニトリル等、およびそれらを含む共重合体等を用いることができる。例えばＰＶＤＦとヘキサフルオロプロピレン（ＨＦＰ）の共重合体であるＰＶＤＦ―ＨＦＰを用いることができる。また、形成されるポリマーは、多孔質形状を有してもよい。

【０２７７】

また、電解液の代わりに、硫化物系や酸化物系等の無機物材料を有する固体電解質や、ＰＥＯ（ポリエチレンオキシド）系等の高分子材料を有する固体電解質を用いることができる。固体電解質を用いる場合には、セパレータやスペーサの設置が不要となる。また、電池全体を固体化できるため、漏液のおそれがなくなり安全性が飛躍的に向上する。

【０２７８】
［セパレータ］
また二次電池は、セパレータを有することが好ましい。セパレータとしては、例えば、紙、不織布、ガラス繊維、セラミックス、或いはナイロン（ポリアミド）、ビニロン（ポリビニルアルコール系繊維）、ポリエステル、アクリル、ポリオレフィン、ポリウレタンを用いた合成繊維等で形成されたものを用いることができる。セパレータはエンベロープ状に加工し、正極または負極のいずれか一方を包むように配置することが好ましい。
【０２７９】
セパレータは多層構造であってもよい。たとえばポリプロピレン、ポリエチレン等の有機材料フィルムに、セラミック系材料、フッ素系材料、ポリアミド系材料、またはこれらを混合したもの等をコートすることができる。セラミック系材料としては、たとえば酸化アルミニウム粒子、酸化シリコン粒子等を用いることができる。フッ素系材料としては、たとえばＰＶＤＦ、ポリテトラフルオロエチレン等を用いることができる。ポリアミド系材料としては、たとえばナイロン、アラミド（メタ系アラミド、パラ系アラミド）等を用いることができる。
【０２８０】
セラミック系材料をコートすると耐酸化性が向上するため、高電圧充放電の際のセパレータの劣化を抑制し、二次電池の信頼性を向上させることができる。またフッ素系材料をコートするとセパレータと電極が密着しやすくなり、出力特性を向上させることができる。ポリアミド系材料、特にアラミドをコートすると、耐熱性が向上するため、二次電池の安全性を向上させることができる。
【０２８１】
たとえばポリプロピレンのフィルムの両面に酸化アルミニウムとアラミドの混合材料をコートしてもよい。また、ポリプロピレンのフィルムの、正極と接する面に酸化アルミニウムとアラミドの混合材料をコートし、負極と接する面にフッ素系材料をコートしてもよい。
【０２８２】
多層構造のセパレータを用いると、セパレータ全体の厚さが薄くても二次電池の安全性を保つことができるため、二次電池の体積あたりの容量を大きくすることができる。
【０２８３】
［外装体］
二次電池が有する外装体としては、例えばアルミニウムなどの金属材料や樹脂材料を用いることができる。また、フィルム状の外装体を用いることもできる。フィルムとしては、例えばポリエチレン、ポリプロピレン、ポリカーボネート、アイオノマー、ポリアミド等の材料からなる膜上に、アルミニウム、ステンレス、銅、ニッケル等の可撓性に優れた金属薄膜を設け、さらに該金属薄膜上に外装体の外面としてポリアミド系樹脂、ポリエステル系樹脂等の絶縁性合成樹脂膜を設けた三層構造のフィルムを用いることができる。
【０２８４】
［充放電方法］
二次電池の充放電は、たとえば下記のように行うことができる。
【０２８５】
≪ＣＣ充電≫
まず、充電方法の１つとしてＣＣ充電について説明する。ＣＣ充電は、充電期間のすべてで一定の電流を二次電池に流し、所定の電圧になったときに充電を停止する充電方法である。二次電池を、図８（Ａ）に示すように内部抵抗Ｒと二次電池容量Ｃの等価回路と仮定する。この場合、二次電池電圧V_Bは、内部抵抗Ｒにかかる電圧V_Rと二次電池容量Ｃにかかる電圧V_Cの和である。
【０２８６】
ＣＣ充電を行っている間は、図８（Ａ）に示すように、スイッチがオンになり、一定の電流Ｉが二次電池に流れる。この間、電流Ｉが一定であるため、$V_R = R \times I$のオームの法

則により、内部抵抗Ｒにかかる電圧$Ｖ_Ｒ$も一定である。一方、二次電池容量Ｃにかかる電圧$Ｖ_Ｃ$は、時間の経過とともに上昇する。そのため、二次電池電圧$Ｖ_Ｂ$は、時間の経過とともに上昇する。

【０２８７】

そして二次電池電圧$Ｖ_Ｂ$が所定の電圧、例えば４．３Ｖになったときに、充電を停止する。ＣＣ充電を停止すると、図８（Ｂ）に示すように、スイッチがオフになり、電流Ｉ＝０となる。そのため、内部抵抗Ｒにかかる電圧$Ｖ_Ｒ$が０Ｖとなる。そのため、内部抵抗Ｒでの電圧降下がなくなった分、二次電池電圧$Ｖ_Ｂ$が下降する。

【０２８８】

ＣＣ充電を行っている間と、ＣＣ充電を停止してからの、二次電池電圧$Ｖ_Ｂ$と充電電流の例を図８（Ｃ）に示す。ＣＣ充電を行っている間は上昇していた二次電池電圧$Ｖ_Ｂ$が、ＣＣ充電を停止してから若干低下する様子が示されている。

【０２８９】

≪ＣＣＣＶ充電≫

次に、上記と異なる充電方法であるＣＣＣＶ充電について説明する。ＣＣＣＶ充電は、まずＣＣ充電にて所定の電圧まで充電を行い、その後ＣＶ（定電圧）充電にて流れる電流が少なくなるまで、具体的には終止電流値になるまで充電を行う充電方法である。

【０２９０】

ＣＣ充電を行っている間は、図９（Ａ）に示すように、定電流電源のスイッチがオン、定電圧電源のスイッチがオフになり、一定の電流Ｉが二次電池に流れる。この間、電流Ｉが一定であるため、$Ｖ_Ｒ＝Ｒ×Ｉ$のオームの法則により、内部抵抗Ｒにかかる電圧$Ｖ_Ｒ$も一定である。一方、二次電池容量Ｃにかかる電圧$Ｖ_Ｃ$は、時間の経過とともに上昇する。そのため、二次電池電圧$Ｖ_Ｂ$は、時間の経過とともに上昇する。

【０２９１】

そして二次電池電圧$Ｖ_Ｂ$が所定の電圧、例えば４．３Ｖになったときに、ＣＣ充電からＣＶ充電に切り替える。ＣＶ充電を行っている間は、図９（Ｂ）に示すように、定電圧電源のスイッチがオン、定電流電源のスイッチがオフになり、二次電池電圧$Ｖ_Ｂ$が一定となる。一方、二次電池容量Ｃにかかる電圧$Ｖ_Ｃ$は、時間の経過とともに上昇する。$Ｖ_Ｂ＝Ｖ_Ｒ＋Ｖ_Ｃ$であるため、内部抵抗Ｒにかかる電圧$Ｖ_Ｒ$は、時間の経過とともに小さくなる。内部抵抗Ｒにかかる電圧$Ｖ_Ｒ$が小さくなるに従い、$Ｖ_Ｒ＝Ｒ×Ｉ$のオームの法則により、二次電池に流れる電流Ｉも小さくなる。

【０２９２】

そして二次電池に流れる電流Ｉが所定の電流、例えば０．０１Ｃ相当の電流となったとき、充電を停止する。ＣＣＣＶ充電を停止すると、図９（Ｃ）に示すように、全てのスイッチがオフになり、電流Ｉ＝０となる。そのため、内部抵抗Ｒにかかる電圧$Ｖ_Ｒ$が０Ｖとなる。しかし、ＣＶ充電により内部抵抗Ｒにかかる電圧$Ｖ_Ｒ$が十分に小さくなっているため、内部抵抗Ｒでの電圧降下がなくなっても、二次電池電圧$Ｖ_Ｂ$はほとんど降下しない。

【０２９３】

ＣＣＣＶ充電を行っている間と、ＣＣＣＶ充電を停止してからの、二次電池電圧$Ｖ_Ｂ$と充電電流の例を図９（Ｄ）に示す。ＣＣＣＶ充電を停止しても、二次電池電圧$Ｖ_Ｂ$がほとんど降下しない様子が示されている。

【０２９４】

≪ＣＣ放電≫

次に、放電方法の１つであるＣＣ放電について説明する。ＣＣ放電は、放電期間のすべてで一定の電流を二次電池から流し、二次電池電圧$Ｖ_Ｂ$が所定の電圧、例えば２．５Ｖになったときに放電を停止する放電方法である。

【０２９５】

ＣＣ放電を行っている間の二次電池電圧$Ｖ_Ｂ$と放電電流の例を図１０に示す。放電が進むに従い、二次電池電圧$Ｖ_Ｂ$が降下していく様子が示されている。

【０２９６】

次に、放電レート及び充電レートについて説明する。放電レートとは、電池容量に対する放電時の電流の相対的な比率であり、単位Ｃで表される。定格容量Ｘ（Ａｈ）の電池において、１Ｃ相当の電流は、Ｘ（Ａ）である。２Ｘ（Ａ）の電流で放電させた場合は、２Ｃで放電させたといい、Ｘ／５（Ａ）の電流で放電させた場合は、０．２Ｃで放電させたという。また、充電レートも同様であり、２Ｘ（Ａ）の電流で充電させた場合は、２Ｃで充電させたといい、Ｘ／５（Ａ）の電流で充電させた場合は、０．２Ｃで充電させたという。

【０２９７】

（実施の形態３）

本実施の形態では、先の実施の形態で説明した正極活物質１００を有する二次電池の形状の例について説明する。本実施の形態で説明する二次電池に用いる材料は、先の実施の形態の記載を参酌することができる。

【０２９８】

［コイン型二次電池］

まずコイン型の二次電池の一例について説明する。図１１（Ａ）はコイン型（単層偏平型）の二次電池の外観図であり、図１１（Ｂ）は、その断面図である。

【０２９９】

コイン型の二次電池３００は、正極端子を兼ねた正極缶３０１と負極端子を兼ねた負極缶３０２とが、ポリプロピレン等で形成されたガスケット３０３で絶縁シールされている。正極３０４は、正極集電体３０５と、これと接するように設けられた正極活物質層３０６により形成される。また、負極３０７は、負極集電体３０８と、これに接するように設けられた負極活物質層３０９により形成される。

【０３００】

なお、コイン型の二次電池３００に用いる正極３０４および負極３０７は、それぞれ活物質層は片面のみに形成すればよい。

【０３０１】

正極缶３０１、負極缶３０２には、電解液に対して耐食性のあるニッケル、アルミニウム、チタン等の金属、又はこれらの合金やこれらと他の金属との合金（例えばステンレス鋼等）を用いることができる。また、電解液による腐食を防ぐため、ニッケルやアルミニウム等を被覆することが好ましい。正極缶３０１は正極３０４と、負極缶３０２は負極３０７とそれぞれ電気的に接続する。

【０３０２】

これら負極３０７、正極３０４およびセパレータ３１０を電解質に含浸させ、図１１（Ｂ）に示すように、正極缶３０１を下にして正極３０４、セパレータ３１０、負極３０７、負極缶３０２をこの順で積層し、正極缶３０１と負極缶３０２とをガスケット３０３を介して圧着してコイン形の二次電池３００を製造する。

【０３０３】

正極３０４に、先の実施の形態で説明した正極活物質を用いることで、高容量でサイクル特性に優れたコイン型の二次電池３００とすることができる。

【０３０４】

ここで図１１（Ｃ）を用いて二次電池の充電時の電流の流れを説明する。リチウムを用いた二次電池を一つの閉回路とみなした時、リチウムイオンの動きと電流の流れは同じ向きになる。なお、リチウムを用いた二次電池では、充電と放電でアノード（陽極）とカソード（陰極）が入れ替わり、酸化反応と還元反応とが入れ替わることになるため、反応電位が高い電極を正極と呼び、反応電位が低い電極を負極と呼ぶ。したがって、本明細書においては、充電中であっても、放電中であっても、逆パルス電流を流す場合であっても、充電電流を流す場合であっても、正極は「正極」または「＋極（プラス極）」と呼び、負極は「負極」または「―極（マイナス極）」と呼ぶこととする。酸化反応や還元反応に関連したアノード（陽極）やカソード（陰極）という用語を用いると、充電時と放電時とでは、逆になってしまい、混乱を招く可能性がある。したがって、アノード（陽極）やカソー

ド（陰極）という用語は、本明細書においては用いないこととする。仮にアノード（陽極）やカソード（陰極）という用語を用いる場合には、充電時か放電時かを明記し、正極（プラス極）と負極（マイナス極）のどちらに対応するものかも併記することとする。

【０３０５】

図１１（Ｃ）に示す２つの端子には充電器が接続され、二次電池３００が充電される。二次電池３００の充電が進めば、電極間の電位差は大きくなる。

【０３０６】

［円筒型二次電池］

次に円筒型の二次電池の例について図１２を参照して説明する。円筒型の二次電池６００は、図１２（Ａ）に示すように、上面に正極キャップ（電池蓋）６０１を有し、側面および底面に電池缶（外装缶）６０２を有している。これら正極キャップと電池缶（外装缶）６０２とは、ガスケット（絶縁パッキン）６１０によって絶縁されている。

【０３０７】

図１２（Ｂ）は、円筒型の二次電池の断面を模式的に示した図である。中空円柱状の電池缶６０２の内側には、帯状の正極６０４と負極６０６とがセパレータ６０５を間に挟んで捲回された電池素子が設けられている。図示しないが、電池素子はセンターピンを中心に捲回されている。電池缶６０２は、一端が閉じられ、他端が開いている。電池缶６０２には、電解液に対して耐腐食性のあるニッケル、アルミニウム、チタン等の金属、又はこれらの合金やこれらと他の金属との合金（例えば、ステンレス鋼等）を用いることができる。また、電解液による腐食を防ぐため、ニッケルやアルミニウム等を被覆することが好ましい。電池缶６０２の内側において、正極、負極およびセパレータが捲回された電池素子は、対向する一対の絶縁板６０８、６０９により挟まれている。また、電池素子が設けられた電池缶６０２の内部は、非水電解液（図示せず）が注入されている。非水電解液は、コイン型の二次電池と同様のものを用いることができる。

【０３０８】

円筒型の蓄電池に用いる正極および負極は捲回するため、集電体の両面に活物質を形成することが好ましい。正極６０４には正極端子（正極集電リード）６０３が接続され、負極６０６には負極端子（負極集電リード）６０７が接続される。正極端子６０３および負極端子６０７は、ともにアルミニウムなどの金属材料を用いることができる。正極端子６０３は安全弁機構６１２に、負極端子６０７は電池缶６０２の底にそれぞれ抵抗溶接される。安全弁機構６１２は、ＰＴＣ素子（Ｐｏｓｉｔｉｖｅ　Ｔｅｍｐｅｒａｔｕｒｅ　Ｃｏｅｆｆｉｃｉｅｎｔ）６１１を介して正極キャップ６０１と電気的に接続されている。安全弁機構６１２は電池の内圧の上昇が所定の閾値を超えた場合に、正極キャップ６０１と正極６０４との電気的な接続を切断するものである。また、ＰＴＣ素子６１１は温度が上昇した場合に抵抗が増大する熱感抵抗素子であり、抵抗の増大により電流量を制限して異常発熱を防止するものである。ＰＴＣ素子には、チタン酸バリウム（ＢａＴｉＯ$_3$）系半導体セラミックス等を用いることができる。

【０３０９】

また、図１２（Ｃ）のように複数の二次電池６００を、導電板６１３および導電板６１４の間に挟んでモジュール６１５を構成してもよい。複数の二次電池６００は、並列接続されていてもよいし、直列接続されていてもよいし、並列に接続された後さらに直列に接続されていてもよい。複数の二次電池６００を有するモジュール６１５を構成することで、大きな電力を取り出すことができる。

【０３１０】

図１２（Ｄ）はモジュール６１５の上面図である。図を明瞭にするために導電板６１３を点線で示した。図１２（Ｄ）に示すようにモジュール６１５は、複数の二次電池６００を電気的に接続する導線６１６を有していてもよい。導線６１６上に導電板６１３を重畳して設けることができる。また複数の二次電池６００の間に温度制御装置６１７を有していてもよい。二次電池６００が過熱されたときは、温度制御装置６１７により冷却し、二次電池６００が冷えすぎているときは温度制御装置６１７により加熱することができる。そ

のためモジュール６１５の性能が外気温に影響されにくくなる。
【０３１１】
正極６０４に、先の実施の形態で説明した正極活物質を用いることで、高容量でサイクル特性に優れた円筒型の二次電池６００とすることができる。
【０３１２】
［二次電池の構造例］
二次電池の別の構造例について、図１３乃至図１７を用いて説明する。
【０３１３】
図１３（Ａ）及び図１３（Ｂ）は、二次電池の外観図を示す図である。二次電池は、回路基板９００と、二次電池９１３と、を有する。二次電池９１３には、ラベル９１０が貼られている。さらに、図１３（Ｂ）に示すように、二次電池は、端子９５１と、端子９５２と、アンテナ９１４と、アンテナ９１５と、を有する。
【０３１４】
回路基板９００は、端子９１１と、回路９１２と、を有する。端子９１１は、端子９５１、端子９５２、アンテナ９１４、アンテナ９１５、及び回路９１２に接続される。なお、端子９１１を複数設けて、複数の端子９１１のそれぞれを、制御信号入力端子、電源端子などとしてもよい。
【０３１５】
回路９１２は、回路基板９００の裏面に設けられていてもよい。なお、アンテナ９１４及びアンテナ９１５は、コイル状に限定されず、例えば線状、板状であってもよい。また、平面アンテナ、開口面アンテナ、進行波アンテナ、ＥＨアンテナ、磁界アンテナ、誘電体アンテナ等のアンテナを用いてもよい。又は、アンテナ９１４若しくはアンテナ９１５は、平板状の導体でもよい。この平板状の導体は、電界結合用の導体の一つとして機能することができる。つまり、コンデンサの有する２つの導体のうちの一つの導体として、アンテナ９１４若しくはアンテナ９１５を機能させてもよい。これにより、電磁界、磁界だけでなく、電界で電力のやり取りを行うこともできる。
【０３１６】
アンテナ９１４の線幅は、アンテナ９１５の線幅よりも大きいことが好ましい。これにより、アンテナ９１４により受電する電力量を大きくできる。
【０３１７】
二次電池は、アンテナ９１４及びアンテナ９１５と、二次電池９１３との間に層９１６を有する。層９１６は、例えば二次電池９１３による電磁界を遮蔽することができる機能を有する。層９１６としては、例えば磁性体を用いることができる。
【０３１８】
なお、二次電池の構造は、図１３に限定されない。
【０３１９】
例えば、図１４（Ａ―１）及び図１４（Ａ―２）に示すように、図１３（Ａ）及び図１３（Ｂ）に示す二次電池９１３のうち、対向する一対の面のそれぞれにアンテナを設けてもよい。図１４（Ａ―１）は、上記一対の面の一方側方向から見た外観図であり、図１４（Ａ―２）は、上記一対の面の他方側方向から見た外観図である。なお、図１３（Ａ）及び図１３（Ｂ）に示す二次電池と同じ部分については、図１３（Ａ）及び図１３（Ｂ）に示す二次電池の説明を適宜援用できる。
【０３２０】
図１４（Ａ―１）に示すように、二次電池９１３の一対の面の一方に層９１６を挟んでアンテナ９１４が設けられ、図１４（Ａ―２）に示すように、二次電池９１３の一対の面の他方に層９１７を挟んでアンテナ９１５が設けられる。層９１７は、例えば二次電池９１３による電磁界を遮蔽することができる機能を有する。層９１７としては、例えば磁性体を用いることができる。
【０３２１】
上記構造にすることにより、アンテナ９１４及びアンテナ９１８の両方のサイズを大きく

することができる。アンテナ９１８は、例えば、外部機器とのデータ通信を行うことができる機能を有する。アンテナ９１８には、例えばアンテナ９１４に適用可能な形状のアンテナを適用することができる。アンテナ９１８を介した二次電池と他の機器との通信方式としては、ＮＦＣなど、二次電池と他の機器との間で用いることができる応答方式などを適用することができる。

【０３２２】

又は、図１４（Ｂ―１）に示すように、図１３（Ａ）及び図１３（Ｂ）に示す二次電池９１３に表示装置９２０を設けてもよい。表示装置９２０は、端子９１１に電気的に接続される。なお、表示装置９２０が設けられる部分にラベル９１０を設けなくてもよい。なお、図１３（Ａ）及び図１３（Ｂ）に示す二次電池と同じ部分については、図１３（Ａ）及び図１３（Ｂ）に示す二次電池の説明を適宜援用できる。

【０３２３】

表示装置９２０には、例えば充電中であるか否かを示す画像、蓄電量を示す画像などを表示してもよい。表示装置９２０としては、例えば電子ペーパー、液晶表示装置、エレクトロルミネセンス（ＥＬともいう）表示装置などを用いることができる。例えば、電子ペーパーを用いることにより表示装置９２０の消費電力を低減することができる。

【０３２４】

又は、図１４（Ｂ―２）に示すように、図１３（Ａ）及び図１３（Ｂ）に示す二次電池９１３にセンサ９２１を設けてもよい。センサ９２１は、端子９２２を介して端子９１１に電気的に接続される。なお、図１３（Ａ）及び図１３（Ｂ）に示す二次電池と同じ部分については、図１３（Ａ）及び図１３（Ｂ）に示す二次電池の説明を適宜援用できる。

【０３２５】

センサ９２１としては、例えば、変位、位置、速度、加速度、角速度、回転数、距離、光、液、磁気、温度、化学物質、音声、時間、硬度、電場、電流、電圧、電力、放射線、流量、湿度、傾度、振動、におい、又は赤外線を測定することができる機能を有すればよい。センサ９２１を設けることにより、例えば、二次電池が置かれている環境を示すデータ（温度など）を検出し、回路９１２内のメモリに記憶しておくこともできる。

【０３２６】

さらに、二次電池９１３の構造例について図１５及び図１６を用いて説明する。

【０３２７】

図１５（Ａ）に示す二次電池９１３は、筐体９３０の内部に端子９５１と端子９５２が設けられた捲回体９５０を有する。捲回体９５０は、筐体９３０の内部で電解液に含浸される。端子９５２は、筐体９３０に接し、端子９５１は、絶縁材などを用いることにより筐体９３０に接していない。なお、図１５（Ａ）では、便宜のため、筐体９３０を分離して図示しているが、実際は、捲回体９５０が筐体９３０に覆われ、端子９５１及び端子９５２が筐体９３０の外に延在している。筐体９３０としては、金属材料（例えばアルミニウムなど）又は樹脂材料を用いることができる。

【０３２８】

なお、図１５（Ｂ）に示すように、図１５（Ａ）に示す筐体９３０を複数の材料によって形成してもよい。例えば、図１５（Ｂ）に示す二次電池９１３は、筐体９３０ａと筐体９３０ｂが貼り合わされており、筐体９３０ａ及び筐体９３０ｂで囲まれた領域に捲回体９５０が設けられている。

【０３２９】

筐体９３０ａとしては、有機樹脂など、絶縁材料を用いることができる。特に、アンテナが形成される面に有機樹脂などの材料を用いることにより、二次電池９１３による電界の遮蔽を抑制できる。なお、筐体９３０ａによる電界の遮蔽が小さければ、筐体９３０ａの内部にアンテナ９１４やアンテナ９１５などのアンテナを設けてもよい。筐体９３０ｂとしては、例えば金属材料を用いることができる。

【０３３０】

さらに、捲回体９５０の構造について図１６に示す。捲回体９５０は、負極９３１と、正

極９３２と、セパレータ９３３と、を有する。捲回体９５０は、セパレータ９３３を挟んで負極９３１と、正極９３２が重なり合って積層され、該積層シートを捲回させた捲回体である。なお、負極９３１と、正極９３２と、セパレータ９３３と、の積層を、さらに複数重ねてもよい。

【０３３１】

負極９３１は、端子９５１及び端子９５２の一方を介して図１３に示す端子９１１に接続される。正極９３２は、端子９５１及び端子９５２の他方を介して図１３に示す端子９１１に接続される。

【０３３２】

正極９３２に、先の実施の形態で説明した正極活物質を用いることで、高容量でサイクル特性に優れた二次電池９１３とすることができる。

【０３３３】

［ラミネート型二次電池］

次に、ラミネート型の二次電池の例について、図１７乃至図２３を参照して説明する。ラミネート型の二次電池は、可撓性を有する構成とすれば、可撓性を有する部位を少なくとも一部有する電子機器に実装すれば、電子機器の変形に合わせて二次電池も曲げることもできる。

【０３３４】

図１７を用いて、ラミネート型の二次電池９８０について説明する。ラミネート型の二次電池９８０は、図１７（Ａ）に示す捲回体９９３を有する。捲回体９９３は、負極９９４と、正極９９５と、セパレータ９９６と、を有する。捲回体９９３は、図１６で説明した捲回体９５０と同様に、セパレータ９９６を挟んで負極９９４と、正極９９５とが重なり合って積層され、該積層シートを捲回したものである。

【０３３５】

なお、負極９９４、正極９９５およびセパレータ９９６からなる積層の積層数は、必要な容量と素子体積に応じて適宜設計すればよい。負極９９４はリード電極９９７およびリード電極９９８の一方を介して負極集電体（図示せず）に接続され、正極９９５はリード電極９９７およびリード電極９９８の他方を介して正極集電体（図示せず）に接続される。

【０３３６】

図１７（Ｂ）に示すように、外装体となるフィルム９８１と、凹部を有するフィルム９８２とを熱圧着などにより貼り合わせて形成される空間に上述した捲回体９９３を収納することで、図１７（Ｃ）に示すように二次電池９８０を作製することができる。捲回体９９３は、リード電極９９７およびリード電極９９８を有し、フィルム９８１と、凹部を有するフィルム９８２との内部で電解液に含浸される。

【０３３７】

フィルム９８１と、凹部を有するフィルム９８２は、例えばアルミニウムなどの金属材料や樹脂材料を用いることができる。フィルム９８１および凹部を有するフィルム９８２の材料として樹脂材料を用いれば、外部から力が加わったときにフィルム９８１と、凹部を有するフィルム９８２を変形させることができ、可撓性を有する蓄電池を作製することができる。

【０３３８】

また、図１７（Ｂ）および図１７（Ｃ）では２枚のフィルムを用いる例を示しているが、１枚のフィルムを折り曲げることによって空間を形成し、その空間に上述した捲回体９９３を収納してもよい。

【０３３９】

正極９９５に、先の実施の形態で説明した正極活物質を用いることで、高容量でサイクル特性に優れた二次電池９８０とすることができる。

【０３４０】

また図１７では外装体となるフィルムにより形成された空間に捲回体を有する二次電池９８０の例について説明したが、たとえば図１８のように、外装体となるフィルムにより形

成された空間に、短冊状の複数の正極、セパレータおよび負極を有する二次電池としてもよい。

【０３４１】

図１８（Ａ）に示すラミネート型の二次電池５００は、正極集電体５０１および正極活物質層５０２を有する正極５０３と、負極集電体５０４および負極活物質層５０５を有する負極５０６と、セパレータ５０７と、電解液５０８と、外装体５０９と、を有する。外装体５０９内に設けられた正極５０３と負極５０６との間にセパレータ５０７が設置されている。また、外装体５０９内は、電解液５０８で満たされている。電解液５０８には、実施の形態２で示した電解液を用いることができる。

【０３４２】

図１８（Ａ）に示すラミネート型の二次電池５００において、正極集電体５０１および負極集電体５０４は、外部との電気的接触を得る端子の役割も兼ねている。そのため、正極集電体５０１および負極集電体５０４の一部は、外装体５０９から外側に露出するように配置してもよい。また、正極集電体５０１および負極集電体５０４を、外装体５０９から外側に露出させず、リード電極を用いてそのリード電極と正極集電体５０１、或いは負極集電体５０４と超音波接合させてリード電極を外側に露出するようにしてもよい。

【０３４３】

ラミネート型の二次電池５００において、外装体５０９には、例えばポリエチレン、ポリプロピレン、ポリカーボネート、アイオノマー、ポリアミド等の材料からなる膜上に、アルミニウム、ステンレス、銅、ニッケル等の可撓性に優れた金属薄膜を設け、さらに該金属薄膜上に外装体の外面としてポリアミド系樹脂、ポリエステル系樹脂等の絶縁性合成樹脂膜を設けた三層構造のラミネートフィルムを用いることができる。

【０３４４】

また、ラミネート型の二次電池５００の断面構造の一例を図１８（Ｂ）に示す。図１８（Ａ）では簡略のため、２つの集電体で構成する例を示しているが、実際は、複数の電極層で構成する。

【０３４５】

図１９（Ｂ）では、一例として、電極層数を１６としている。なお、電極層数を１６としても二次電池５００は、可撓性を有する。図１８（Ｂ）では負極集電体５０４が８層と、正極集電体５０１が８層の合計１６層の構造を示している。なお、図１８（Ｂ）は負極の取り出し部の断面を示しており、８層の負極集電体５０４を超音波接合させている。勿論、電極層数は１６に限定されず、多くてもよいし、少なくてもよい。電極層数が多い場合には、より多くの容量を有する二次電池とすることができる。また、電極層数が少ない場合には、薄型化でき、可撓性に優れた二次電池とすることができる。

【０３４６】

ここで、ラミネート型の二次電池５００の外観図の一例を図１９及び図２０に示す。図１９及び図２０は、正極５０３、負極５０６、セパレータ５０７、外装体５０９、正極リード電極５１０及び負極リード電極５１１を有する。

【０３４７】

図２１（Ａ）は正極５０３及び負極５０６の外観図を示す。正極５０３は正極集電体５０１を有し、正極活物質層５０２は正極集電体５０１の表面に形成されている。また、正極５０３は正極集電体５０１が一部露出する領域（以下、タブ領域という）を有する。負極５０６は負極集電体５０４を有し、負極活物質層５０５は負極集電体５０４の表面に形成されている。また、負極５０６は負極集電体５０４が一部露出する領域、すなわちタブ領域を有する。正極及び負極が有するタブ領域の面積や形状は、図２１（Ａ）に示す例に限られない。

【０３４８】

［ラミネート型二次電池の作製方法］

ここで、図１９に外観図を示すラミネート型二次電池の作製方法の一例について、図２１（Ｂ）、（Ｃ）を用いて説明する。

【０３４９】
まず、負極５０６、セパレータ５０７及び正極５０３を積層する。図２１（Ｂ）に積層された負極５０６、セパレータ５０７及び正極５０３を示す。ここでは負極を５組、正極を４組使用する例を示す。次に、正極５０３のタブ領域同士の接合と、最表面の正極のタブ領域への正極リード電極５１０の接合を行う。接合には、例えば超音波溶接等を用いればよい。同様に、負極５０６のタブ領域同士の接合と、最表面の負極のタブ領域への負極リード電極５１１の接合を行う。
【０３５０】
次に外装体５０９上に、負極５０６、セパレータ５０７及び正極５０３を配置する。
【０３５１】
次に、図２１（Ｃ）に示すように、外装体５０９を破線で示した部分で折り曲げる。その後、外装体５０９の外周部を接合する。接合には例えば熱圧着等を用いればよい。この時、後に電解液５０８を入れることができるように、外装体５０９の一部（または一辺）に接合されない領域（以下、導入口という）を設ける。
【０３５２】
次に、外装体５０９に設けられた導入口から、電解液５０８を外装体５０９の内側へ導入する。電解液５０８の導入は、減圧雰囲気下、或いは不活性ガス雰囲気下で行うことが好ましい。そして最後に、導入口を接合する。このようにして、ラミネート型の二次電池である二次電池５００を作製することができる。
【０３５３】
正極５０３に、先の実施の形態で説明した正極活物質を用いることで、高容量でサイクル特性に優れた二次電池５００とすることができる。
【０３５４】
［曲げることのできる二次電池］
次に、曲げることのできる二次電池の例について図２２および図２３を参照して説明する。
【０３５５】
図２２（Ａ）に、曲げることのできる二次電池５０の上面概略図を示す。図２２（Ｂ１）、（Ｂ２）、（Ｃ）にはそれぞれ、図２２（Ａ）中の切断線Ｃ１―Ｃ２、切断線Ｃ３―Ｃ４、切断線Ａ１―Ａ２における断面概略図である。電池５０は、外装体５１と、外装体５１の内部に収容された正極１１ａおよび負極１１ｂを有する。正極１１ａと電気的に接続されたリード１２ａ、および負極１１ｂと電気的に接続されたリード１２ｂは、外装体５１の外側に延在している。また外装体５１で囲まれた領域には、正極１１ａおよび負極１１ｂに加えて電解液（図示しない）が封入されている。
【０３５６】
電池５０が有する正極１１ａおよび負極１１ｂについて、図２３を用いて説明する。図２３（Ａ）は、正極１１ａ、負極１１ｂおよびセパレータ１４の積層順を説明する斜視図である。図２３（Ｂ）は正極１１ａおよび負極１１ｂに加えて、リード１２ａおよびリード１２ｂを示す斜視図である。
【０３５７】
図２３（Ａ）に示すように、電池５０は、複数の短冊状の正極１１ａ、複数の短冊状の負極１１ｂおよび複数のセパレータ１４を有する。正極１１ａおよび負極１１ｂはそれぞれ突出したタブ部分と、タブ以外の部分を有する。正極１１ａの一方の面のタブ以外の部分に正極活物質層が形成され、負極１１ｂの一方の面のタブ以外の部分に負極活物質層が形成される。
【０３５８】
正極１１ａの正極活物質層の形成されていない面同士、および負極１１ｂの負極活物質の形成されていない面同士が接するように、正極１１ａおよび負極１１ｂは積層される。
【０３５９】
また、正極１１ａの正極活物質が形成された面と、負極１１ｂの負極活物質が形成された

面の間にはセパレータ１４が設けられる。図２３（Ａ）では見やすくするためセパレータ１４を点線で示す。

【０３６０】

また図２３（Ｂ）に示すように、複数の正極１１ａとリード１２ａは、接合部１５ａにおいて電気的に接続される。また複数の負極１１ｂとリード１２ｂは、接合部１５ｂにおいて電気的に接続される。

【０３６１】

次に、外装体５１について図２２（Ｂ１）、（Ｂ２）、（Ｃ）、（Ｄ）を用いて説明する。

【０３６２】

外装体５１は、フィルム状の形状を有し、正極１１ａおよび負極１１ｂを挟むように２つに折り曲げられている。外装体５１は、折り曲げ部６１と、一対のシール部６２と、シール部６３と、を有する。一対のシール部６２は、正極１１ａおよび負極１１ｂを挟んで設けられ、サイドシールとも呼ぶことができる。また、シール部６３は、リード１２ａ及びリード１２ｂと重なる部分を有し、トップシールとも呼ぶことができる。

【０３６３】

外装体５１は、正極１１ａおよび負極１１ｂと重なる部分に、稜線７１と谷線７２が交互に並んだ波形状を有することが好ましい。また、外装体５１のシール部６２及びシール部６３は、平坦であることが好ましい。

【０３６４】

図２２（Ｂ１）は、稜線７１と重なる部分で切断した断面であり、図２２（Ｂ２）は、谷線７２と重なる部分で切断した断面である。図２２（Ｂ１）、（Ｂ２）は共に、電池５０及び正極１１ａおよび負極１１ｂの幅方向の断面に対応する。

【０３６５】

ここで、負極１１ｂの幅方向の端部と、シール部６２との間の距離を距離Ｌａとする。電池５０に曲げるなどの変形を加えたとき、後述するように正極１１ａおよび負極１１ｂが長さ方向に互いにずれるように変形する。その際、距離Ｌａが短すぎると、外装体５１と正極１１ａおよび負極１１ｂとが強く擦れ、外装体５１が破損してしまう場合がある。特に外装体５１の金属フィルムが露出すると、当該金属フィルムが電解液により腐食されてしまう恐れがある。したがって、距離Ｌａを出来るだけ長く設定することが好ましい。一方で、距離Ｌａを大きくしすぎると、電池５０の体積が増大してしまう。

【０３６６】

また、積層された正極１１ａおよび負極１１ｂの合計の厚さが厚いほど、負極１１ｂとシール部６２との間の距離Ｌａを大きくすることが好ましい。

【０３６７】

より具体的には、積層された正極１１ａおよび負極１１ｂおよび図示しないがセパレータ２１４の合計の厚さを厚さｔとしたとき、距離Ｌａは、厚さｔの０．８倍以上３．０倍以下、好ましくは０．９倍以上２．５倍以下、より好ましくは１．０倍以上２．０倍以下であることが好ましい。距離Ｌａをこの範囲とすることで、コンパクトで、且つ曲げに対する信頼性の高い電池を実現できる。

【０３６８】

また、一対のシール部６２の間の距離を距離Ｌｂとしたとき、距離Ｌｂを負極１１ｂの幅Ｗｂよりも十分大きくすることが好ましい。これにより、電池５０に繰り返し曲げるなどの変形を加えたときに、正極１１ａおよび負極１１ｂと外装体５１とが接触しても、正極１１ａおよび負極１１ｂの一部が幅方向にずれることができるため、正極１１ａおよび負極１１ｂと外装体５１とが擦れてしまうことを効果的に防ぐことができる。

【０３６９】

例えば、一対のシール部６２の間の距離Ｌｂと、負極１１ｂの幅Ｗｂとの差が、正極１１ａおよび負極１１ｂの厚さｔの１．６倍以上６．０倍以下、好ましくは１．８倍以上５．０倍以下、より好ましくは、２．０倍以上４．０倍以下を満たすことが好ましい。

【０３７０】
言い換えると、距離Ｌｂ、幅Ｗｂ、及び厚さｔが、下記数式２の関係を満たすことが好ましい。
【０３７１】
【数２】

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

【０３７２】
ここで、ａは、０．８以上３．０以下、好ましくは０．９以上２．５以下、より好ましくは１．０以上２．０以下を満たす。
【０３７３】
また、図２２（Ｃ）はリード１２ａを含む断面であり、電池５０、正極１１ａおよび負極１１ｂの長さ方向の断面に対応する。図２２（Ｃ）に示すように、折り曲げ部６１において、正極１１ａおよび負極１１ｂの長さ方向の端部と、外装体５１との間に空間７３を有することが好ましい。
【０３７４】
図２２（Ｄ）に、電池５０を曲げたときの断面概略図を示している。図２２（Ｄ）は、図２２（Ａ）中の切断線Ｂ１―Ｂ２における断面に相当する。
【０３７５】
電池５０を曲げると、曲げの外側に位置する外装体５１の一部は伸び、内側に位置する他の一部は縮むように変形する。より具体的には、外装体５１の外側に位置する部分は、波の振幅が小さく、且つ波の周期が大きくなるように変形する。一方、外装体５１の内側に位置する部分は、波の振幅が大きく、且つ波の周期が小さくなるように変形する。このように、外装体５１が変形することにより、曲げに伴って外装体５１にかかる応力が緩和されるため、外装体５１を構成する材料自体が伸縮する必要がない。その結果、外装体５１は破損することなく、小さな力で電池５０を曲げることができる。
【０３７６】
また、図２２（Ｄ）に示すように、電池５０を曲げると、正極１１ａおよび負極１１ｂとがそれぞれ相対的にずれる。このとき、複数の積層された正極１１ａおよび負極１１ｂは、シール部６３側の一端が固定部材１７で固定されているため、折り曲げ部６１に近いほどずれ量が大きくなるように、それぞれずれる。これにより、正極１１ａおよび負極１１ｂにかかる応力が緩和され、正極１１ａおよび負極１１ｂ自体が伸縮する必要がない。その結果、正極１１ａおよび負極１１ｂが破損することなく電池５０を曲げることができる。
【０３７７】
また、正極１１ａおよび負極１１ｂの端部と、外装体５１との間に空間７３を有していることにより、曲げた時内側に位置する正極１１ａおよび負極１１ｂの端部が、外装体５１に接触することなく、相対的にずれることができる。
【０３７８】
図２２および図２３で例示した電池５０は、繰り返し曲げ伸ばしを行っても、外装体の破損、正極１１ａおよび負極１１ｂの破損などが生じにくく、電池特性も劣化しにくい電池である。電池５０が有する正極１１ａに、先の実施の形態で説明した正極活物質を用いることで、さらにサイクル特性に優れた電池とすることができる。
【０３７９】
（実施の形態４）
本実施の形態では、本発明の一態様である二次電池を電子機器に実装する例について説明する。
【０３８０】

まず実施の形態３の一部で説明した、曲げることのできる二次電池を電子機器に実装する例を図２４（Ａ）乃至図２４（Ｇ）に示す。曲げることのできる二次電池を適用した電子機器として、例えば、テレビジョン装置（テレビ、又はテレビジョン受信機ともいう）、コンピュータ用などのモニタ、デジタルカメラ、デジタルビデオカメラ、デジタルフォトフレーム、携帯電話機（携帯電話、携帯電話装置ともいう）、携帯型ゲーム機、携帯情報端末、音響再生装置、パチンコ機などの大型ゲーム機などが挙げられる。

【０３８１】

また、フレキシブルな形状を備える二次電池を、家屋やビルの内壁または外壁や、自動車の内装または外装の曲面に沿って組み込むことも可能である。

【０３８２】

図２４（Ａ）は、携帯電話機の一例を示している。携帯電話機７４００は、筐体７４０１に組み込まれた表示部７４０２の他、操作ボタン７４０３、外部接続ポート７４０４、スピーカ７４０５、マイク７４０６などを備えている。なお、携帯電話機７４００は、二次電池７４０７を有している。上記の二次電池７４０７に本発明の一態様の二次電池を用いることで、軽量で長寿命な携帯電話機を提供できる。

【０３８３】

図２４（Ｂ）は、携帯電話機７４００を湾曲させた状態を示している。携帯電話機７４００を外部の力により変形させて全体を湾曲させると、その内部に設けられている二次電池７４０７も湾曲される。また、その時、曲げられた二次電池７４０７の状態を図２４（Ｃ）に示す。二次電池７４０７は薄型の蓄電池である。二次電池７４０７は曲げられた状態で固定されている。なお、二次電池７４０７は集電体７４０９と電気的に接続されたリード電極７４０８を有している。例えば、集電体７４０９は銅箔であり、一部ガリウムと合金化させて、集電体７４０９と接する活物質層との密着性を向上し、二次電池７４０７が曲げられた状態での信頼性が高い構成となっている。

【０３８４】

図２４（Ｄ）は、バングル型の表示装置の一例を示している。携帯表示装置７１００は、筐体７１０１、表示部７１０２、操作ボタン７１０３、及び二次電池７１０４を備える。また、図２４（Ｅ）に曲げられた二次電池７１０４の状態を示す。二次電池７１０４は曲げられた状態で使用者の腕への装着時に、筐体が変形して二次電池７１０４の一部または全部の曲率が変化する。なお、曲線の任意の点における曲がり具合を相当する円の半径の値で表したものを曲率半径であり、曲率半径の逆数を曲率と呼ぶ。具体的には、曲率半径が４０ｍｍ以上１５０ｍｍ以下の範囲内で筐体または二次電池７１０４の主表面の一部または全部が変化する。二次電池７１０４の主表面における曲率半径が４０ｍｍ以上１５０ｍｍ以下の範囲であれば、高い信頼性を維持できる。上記の二次電池７１０４に本発明の一態様の二次電池を用いることで、軽量で長寿命な携帯表示装置を提供できる。

【０３８５】

図２４（Ｆ）は、腕時計型の携帯情報端末の一例を示している。携帯情報端末７２００は、筐体７２０１、表示部７２０２、バンド７２０３、バックル７２０４、操作ボタン７２０５、入出力端子７２０６などを備える。

【０３８６】

携帯情報端末７２００は、移動電話、電子メール、文章閲覧及び作成、音楽再生、インターネット通信、コンピュータゲームなどの種々のアプリケーションを実行することができる。

【０３８７】

表示部７２０２はその表示面が湾曲して設けられ、湾曲した表示面に沿って表示を行うことができる。また、表示部７２０２はタッチセンサを備え、指やスタイラスなどで画面に触れることで操作することができる。例えば、表示部７２０２に表示されたアイコン７２０７に触れることで、アプリケーションを起動することができる。

【０３８８】

操作ボタン７２０５は、時刻設定のほか、電源のオン、オフ動作、無線通信のオン、オフ

動作、マナーモードの実行及び解除、省電力モードの実行及び解除など、様々な機能を持たせることができる。例えば、携帯情報端末７２００に組み込まれたオペレーティングシステムにより、操作ボタン７２０５の機能を自由に設定することもできる。

【０３８９】

また、携帯情報端末７２００は、通信規格された近距離無線通信を実行することが可能である。例えば無線通信可能なヘッドセットと相互通信することによって、ハンズフリーで通話することもできる。

【０３９０】

また、携帯情報端末７２００は入出力端子７２０６を備え、他の情報端末とコネクターを介して直接データのやりとりを行うことができる。また入出力端子７２０６を介して充電を行うこともできる。なお、充電動作は入出力端子７２０６を介さずに無線給電により行ってもよい。

【０３９１】

携帯情報端末７２００の表示部７２０２には、本発明の一態様の二次電池を有している。本発明の一態様の二次電池を用いることで、軽量で長寿命な携帯情報端末を提供できる。例えば、図２４（Ｅ）に示した二次電池７１０４を、筐体７２０１の内部に湾曲した状態で、またはバンド７２０３の内部に湾曲可能な状態で組み込むことができる。

【０３９２】

携帯情報端末７２００はセンサを有することが好ましい。センサとして例えば、指紋センサ、脈拍センサ、体温センサ等の人体センサや、タッチセンサ、加圧センサ、加速度センサ、等が搭載されることが好ましい。

【０３９３】

図２４（Ｇ）は、腕章型の表示装置の一例を示している。表示装置７３００は、表示部７３０４を有し、本発明の一態様の二次電池を有している。また、表示装置７３００は、表示部７３０４にタッチセンサを備えることもでき、また、携帯情報端末として機能させることもできる。

【０３９４】

表示部７３０４はその表示面が湾曲しており、湾曲した表示面に沿って表示を行うことができる。また、表示装置７３００は、通信規格された近距離無線通信などにより、表示状況を変更することができる。

【０３９５】

また、表示装置７３００は入出力端子を備え、他の情報端末とコネクターを介して直接データのやりとりを行うことができる。また入出力端子を介して充電を行うこともできる。なお、充電動作は入出力端子を介さずに無線給電により行ってもよい。

【０３９６】

表示装置７３００が有する二次電池として本発明の一態様の二次電池を用いることで、軽量で長寿命な表示装置を提供できる。

【０３９７】

また、先の実施の形態で示したサイクル特性のよい二次電池を電子機器に実装する例を図２４（Ｈ）、図２５および図２６を用いて説明する。

【０３９８】

日用電子機器に二次電池として本発明の一態様の二次電池を用いることで、軽量で長寿命な製品を提供できる。例えば、日用電子機器として、電動歯ブラシ、電気シェーバー、電動美容機器などが挙げられ、それらの製品の二次電池としては、使用者の持ちやすさを考え、形状をスティック状とし、小型、軽量、且つ、大容量の二次電池が望まれている。

【０３９９】

図２４（Ｈ）はタバコ収容喫煙装置（電子タバコ）とも呼ばれる装置の斜視図である。図２４（Ｈ）において電子タバコ７５００は、加熱素子を含むアトマイザ７５０１と、アトマイザに電力を供給する二次電池７５０４と、液体供給ボトルやセンサなどを含むカートリッジ７５０２で構成されている。安全性を高めるため、二次電池７５０４の過充電や過

放電を防ぐ保護回路を二次電池７５０４に電気的に接続してもよい。図２５（Ｈ）に示した二次電池７５０４は、充電機器と接続できるように外部端子を有している。二次電池７５０４は持った場合に先端部分となるため、トータルの長さが短く、且つ、重量が軽いことが望ましい。本発明の一態様の二次電池は高容量、良好なサイクル特性を有するため、長期間に渡って長時間の使用ができる小型であり、且つ、軽量の電子タバコ７５００を提供できる。

【０４００】

次に、図２５（Ａ）および図２５（Ｂ）に、２つ折り可能なタブレット型端末の一例を示す。図２５（Ａ）および図２５（Ｂ）に示すタブレット型端末９６００は、筐体９６３０ａ、筐体９６３０ｂ、筐体９６３０ａと筐体９６３０ｂを接続する可動部９６４０、表示部９６３１ａと表示部９６３１ｂを有する表示部９６３１、表示モード切り替えスイッチ９６２６、電源スイッチ９６２７、省電力モード切り替えスイッチ９６２５、留め具９６２９、操作スイッチ９６２８、を有する。表示部９６３１には、可撓性を有するパネルを用いることで、より広い表示部を有するタブレット端末とすることができる。図２５（Ａ）は、タブレット型端末９６００を開いた状態を示し、図２５（Ｂ）は、タブレット型端末９６００を閉じた状態を示している。

【０４０１】

また、タブレット型端末９６００は、筐体９６３０ａおよび筐体９６３０ｂの内部に蓄電体９６３５を有する。蓄電体９６３５は、可動部９６４０を通り、筐体９６３０ａと筐体９６３０ｂに渡って設けられている。

【０４０２】

表示部９６３１ａは、一部をタッチパネルの領域９６３２ａとすることができ、表示された操作キー９６３８にふれることでデータ入力をすることができる。なお、表示部９６３１ａにおいては、一例として半分の領域が表示のみの機能を有する構成、もう半分の領域がタッチパネルの機能を有する構成を示しているが該構成に限定されない。表示部９６３１ａの全ての領域がタッチパネルの機能を有する構成としても良い。例えば、表示部９６３１ａの全面をキーボードボタン表示させてタッチパネルとし、表示部９６３１ｂを表示画面として用いることができる。

【０４０３】

また、表示部９６３１ｂにおいても表示部９６３１ａと同様に、表示部９６３１ｂの一部をタッチパネルの領域９６３２ｂとすることができる。また、タッチパネルのキーボード表示切り替えボタン９６３９が表示されている位置に指やスタイラスなどでふれることで表示部９６３１ｂにキーボードボタン表示することができる。

【０４０４】

また、タッチパネルの領域９６３２ａとタッチパネルの領域９６３２ｂに対して同時にタッチ入力することもできる。

【０４０５】

また、表示モード切り替えスイッチ９６２６は、縦表示又は横表示などの表示の向きを切り替え、白黒表示やカラー表示の切り替えなどを選択できる。省電力モード切り替えスイッチ９６２５は、タブレット型端末９６００に内蔵している光センサで検出される使用時の外光の光量に応じて表示の輝度を最適なものとすることができる。タブレット型端末は光センサだけでなく、ジャイロ、加速度センサ等の傾きを検出するセンサなどの他の検出装置を内蔵させてもよい。

【０４０６】

また、図２５（Ａ）では表示部９６３１ｂと表示部９６３１ａの表示面積が同じ例を示しているが特に限定されず、一方のサイズともう一方のサイズが異なっていてもよく、表示の品質も異なっていてもよい。例えば一方が他方よりも高精細な表示を行える表示パネルとしてもよい。

【０４０７】

図２５（Ｂ）は、閉じた状態であり、タブレット型端末は、筐体９６３０、太陽電池９６

３３、ＤＣＤＣコンバータ９６３６を含む充放電制御回路９６３４有する。また、蓄電体９６３５として、本発明の一態様に係る蓄電体を用いる。

【０４０８】

なお、タブレット型端末９６００は２つ折り可能なため、未使用時に筐体９６３０ａおよび筐体９６３０ｂを重ね合せるように折りたたむことができる。折りたたむことにより、表示部９６３１ａ、表示部９６３１ｂを保護できるため、タブレット型端末９６００の耐久性を高めることができる。また、本発明の一態様の二次電池を用いた蓄電体９６３５は高容量、良好なサイクル特性を有するため、長期間に渡って長時間の使用ができるタブレット型端末９６００を提供できる。

【０４０９】

また、この他にも図２５（Ａ）および図２５（Ｂ）に示したタブレット型端末は、様々な情報（静止画、動画、テキスト画像など）を表示する機能、カレンダー、日付又は時刻などを表示部に表示する機能、表示部に表示した情報をタッチ入力操作又は編集するタッチ入力機能、様々なソフトウェア（プログラム）によって処理を制御する機能、等を有することができる。

【０４１０】

タブレット型端末の表面に装着された太陽電池９６３３によって、電力をタッチパネル、表示部、又は映像信号処理部等に供給することができる。なお、太陽電池９６３３は、筐体９６３０の片面又は両面に設けることができ、蓄電体９６３５の充電を効率的に行う構成とすることができる。なお蓄電体９６３５としては、リチウムイオン電池を用いると、小型化を図れる等の利点がある。

【０４１１】

また、図２５（Ｂ）に示す充放電制御回路９６３４の構成、および動作について図２５（Ｃ）にブロック図を示し説明する。図２５（Ｃ）には、太陽電池９６３３、蓄電体９６３５、ＤＣＤＣコンバータ９６３６、コンバータ９６３７、スイッチＳＷ１乃至ＳＷ３、表示部９６３１について示しており、蓄電体９６３５、ＤＣＤＣコンバータ９６３６、コンバータ９６３７、スイッチＳＷ１乃至ＳＷ３が、図２５（Ｂ）に示す充放電制御回路９６３４に対応する箇所となる。

【０４１２】

まず外光により太陽電池９６３３により発電がされる場合の動作の例について説明する。太陽電池で発電した電力は、蓄電体９６３５を充電するための電圧となるようＤＣＤＣコンバータ９６３６で昇圧又は降圧がなされる。そして、表示部９６３１の動作に太陽電池９６３３からの電力が用いられる際にはスイッチＳＷ１をオンにし、コンバータ９６３７で表示部９６３１に必要な電圧に昇圧又は降圧をすることとなる。また、表示部９６３１での表示を行わない際には、ＳＷ１をオフにし、ＳＷ２をオンにして蓄電体９６３５の充電を行う構成とすればよい。

【０４１３】

なお太陽電池９６３３については、発電手段の一例として示したが、特に限定されず、圧電素子（ピエゾ素子）や熱電変換素子（ペルティエ素子）などの他の発電手段による蓄電体９６３５の充電を行う構成であってもよい。例えば、無線（非接触）で電力を送受信して充電する無接点電力伝送モジュールや、また他の充電手段を組み合わせて行う構成としてもよい。

【０４１４】

図２６に、他の電子機器の例を示す。図２６において、表示装置８０００は、本発明の一態様に係る二次電池８００４を用いた電子機器の一例である。具体的に、表示装置８０００は、ＴＶ放送受信用の表示装置に相当し、筐体８００１、表示部８００２、スピーカ部８００３、二次電池８００４等を有する。本発明の一態様に係る二次電池８００４は、筐体８００１の内部に設けられている。表示装置８０００は、商用電源から電力の供給を受けることもできるし、二次電池８００４に蓄積された電力を用いることもできる。よって、停電などにより商用電源から電力の供給が受けられない時でも、本発明の一態様に係る

二次電池８００４を無停電電源として用いることで、表示装置８０００の利用が可能となる。

【０４１５】

表示部８００２には、液晶表示装置、有機ＥＬ素子などの発光素子を各画素に備えた発光装置、電気泳動表示装置、ＤＭＤ（Ｄｉｇｉｔａｌ　Ｍｉｃｒｏｍｉｒｒｏｒ　Ｄｅｖｉｃｅ）、ＰＤＰ（Ｐｌａｓｍａ　Ｄｉｓｐｌａｙ　Ｐａｎｅｌ）、ＦＥＤ（Ｆｉｅｌｄ　Ｅｍｉｓｓｉｏｎ　Ｄｉｓｐｌａｙ）などの、半導体表示装置を用いることができる。

【０４１６】

なお、表示装置には、ＴＶ放送受信用の他、パーソナルコンピュータ用、広告表示用など、全ての情報表示用表示装置が含まれる。

【０４１７】

図２６において、据え付け型の照明装置８１００は、本発明の一態様に係る二次電池８１０３を用いた電子機器の一例である。具体的に、照明装置８１００は、筐体８１０１、光源８１０２、二次電池８１０３等を有する。図２６では、二次電池８１０３が、筐体８１０１及び光源８１０２が据え付けられた天井８１０４の内部に設けられている場合を例示しているが、二次電池８１０３は、筐体８１０１の内部に設けられていても良い。照明装置８１００は、商用電源から電力の供給を受けることもできるし、二次電池８１０３に蓄積された電力を用いることもできる。よって、停電などにより商用電源から電力の供給が受けられない時でも、本発明の一態様に係る二次電池８１０３を無停電電源として用いることで、照明装置８１００の利用が可能となる。

【０４１８】

なお、図２６では天井８１０４に設けられた据え付け型の照明装置８１００を例示しているが、本発明の一態様に係る二次電池は、天井８１０４以外、例えば側壁８１０５、床８１０６、窓８１０７等に設けられた据え付け型の照明装置に用いることもできるし、卓上型の照明装置などに用いることもできる。

【０４１９】

また、光源８１０２には、電力を利用して人工的に光を得る人工光源を用いることができる。具体的には、白熱電球、蛍光灯などの放電ランプ、ＬＥＤや有機ＥＬ素子などの発光素子が、上記人工光源の一例として挙げられる。

【０４２０】

図２６において、室内機８２００及び室外機８２０４を有するエアコンディショナーは、本発明の一態様に係る二次電池８２０３を用いた電子機器の一例である。具体的に、室内機８２００は、筐体８２０１、送風口８２０２、二次電池８２０３等を有する。図２６では、二次電池８２０３が、室内機８２００に設けられている場合を例示しているが、二次電池８２０３は室外機８２０４に設けられていても良い。或いは、室内機８２００と室外機８２０４の両方に、二次電池８２０３が設けられていても良い。エアコンディショナーは、商用電源から電力の供給を受けることもできるし、二次電池８２０３に蓄積された電力を用いることもできる。特に、室内機８２００と室外機８２０４の両方に二次電池８２０３が設けられている場合、停電などにより商用電源から電力の供給が受けられない時でも、本発明の一態様に係る二次電池８２０３を無停電電源として用いることで、エアコンディショナーの利用が可能となる。

【０４２１】

なお、図２６では、室内機と室外機で構成されるセパレート型のエアコンディショナーを例示しているが、室内機の機能と室外機の機能とを１つの筐体に有する一体型のエアコンディショナーに、本発明の一態様に係る二次電池を用いることもできる。

【０４２２】

図２６において、電気冷凍冷蔵庫８３００は、本発明の一態様に係る二次電池８３０４を用いた電子機器の一例である。具体的に、電気冷凍冷蔵庫８３００は、筐体８３０１、冷蔵室用扉８３０２、冷凍室用扉８３０３、二次電池８３０４等を有する。図２６では、二次電池８３０４が、筐体８３０１の内部に設けられている。電気冷凍冷蔵庫８３００は、

商用電源から電力の供給を受けることもできるし、二次電池８３０４に蓄積された電力を用いることもできる。よって、停電などにより商用電源から電力の供給が受けられない時でも、本発明の一態様に係る二次電池８３０４を無停電電源として用いることで、電気冷凍冷蔵庫８３００の利用が可能となる。

【０４２３】

なお、上述した電子機器のうち、電子レンジ等の高周波加熱装置、電気炊飯器などの電子機器は、短時間で高い電力を必要とする。よって、商用電源では賄いきれない電力を補助するための補助電源として、本発明の一態様に係る二次電池を用いることで、電子機器の使用時に商用電源のブレーカーが落ちるのを防ぐことができる。

【０４２４】

また、電子機器が使用されない時間帯、特に、商用電源の供給元が供給可能な総電力量のうち、実際に使用される電力量の割合（電力使用率と呼ぶ）が低い時間帯において、二次電池に電力を蓄えておくことで、上記時間帯以外において電力使用率が高まるのを抑えることができる。例えば、電気冷凍冷蔵庫８３００の場合、気温が低く、冷蔵室用扉８３０２、冷凍室用扉８３０３の開閉が行われない夜間において、二次電池８３０４に電力を蓄える。そして、気温が高くなり、冷蔵室用扉８３０２、冷凍室用扉８３０３の開閉が行われる昼間において、二次電池８３０４を補助電源として用いることで、昼間の電力使用率を低く抑えることができる。

【０４２５】

本発明の一態様により、二次電池のサイクル特性が良好となり、信頼性を向上させることができる。また、本発明の一態様によれば、高容量の二次電池とすることができ、よって、二次電池の特性を向上することができ、よって、二次電池自体を小型軽量化することができる。そのため本発明の一態様である二次電池を、本実施の形態で説明した電子機器に搭載することで、より長寿命で、より軽量な電子機器とすることができる。本実施の形態は、他の実施の形態と適宜組み合わせて実施することが可能である。

【０４２６】

（実施の形態５）

本実施の形態では、車両に本発明の一態様である二次電池を搭載する例を示す。

【０４２７】

二次電池を車両に搭載すると、ハイブリッド車（ＨＥＶ）、電気自動車（ＥＶ）、又はプラグインハイブリッド車（ＰＨＥＶ）等の次世代クリーンエネルギー自動車を実現できる。

【０４２８】

図２７において、本発明の一態様である二次電池を用いた車両を例示する。図２７（Ａ）に示す自動車８４００は、走行のための動力源として電気モーターを用いる電気自動車である。または、走行のための動力源として電気モーターとエンジンを適宜選択して用いることが可能なハイブリッド自動車である。本発明の一態様を用いることで、航続距離の長い車両を実現することができる。また、自動車８４００は二次電池を有する。二次電池は、車内の床部分に対して、図１２（Ｃ）および図１２（Ｄ）に示した二次電池のモジュールを並べて使用すればよい。また、図１７に示す二次電池を複数組み合わせた電池パックを車内の床部分に対して設置してもよい。二次電池は電気モーター８４０６を駆動するだけでなく、ヘッドライト８４０１やルームライト（図示せず）などの発光装置に電力を供給することができる。

【０４２９】

また、二次電池は、自動車８４００が有するスピードメーター、タコメーターなどの表示装置に電力を供給することができる。また、二次電池は、自動車８４００が有するナビゲーションシステムなどの半導体装置に電力を供給することができる。

【０４３０】

図２７（Ｂ）に示す自動車８５００は、自動車８５００が有する二次電池にプラグイン方式や非接触給電方式等により外部の充電設備から電力供給を受けて、充電することができ

る。図２７（Ｂ）に、地上設置型の充電装置８０２１から自動車８５００に搭載された二次電池８０２４に、ケーブル８０２２を介して充電を行っている状態を示す。充電に際しては、充電方法やコネクターの規格等はＣＨＡｄｅＭＯ（登録商標）やコンボ等の所定の方式で適宜行えばよい。充電装置８０２１は、商用施設に設けられた充電ステーションでもよく、また家庭の電源であってもよい。例えば、プラグイン技術によって、外部からの電力供給により自動車８５００に搭載された二次電池８０２４を充電することができる。充電は、ＡＣＤＣコンバータ等の変換装置を介して、交流電力を直流電力に変換して行うことができる。

【０４３１】

また、図示しないが、受電装置を車両に搭載し、地上の送電装置から電力を非接触で供給して充電することもできる。この非接触給電方式の場合には、道路や外壁に送電装置を組み込むことで、停車中に限らず走行中に充電を行うこともできる。また、この非接触給電の方式を利用して、車両どうしで電力の送受信を行ってもよい。さらに、車両の外装部に太陽電池を設け、停車時や走行時に二次電池の充電を行ってもよい。このような非接触での電力の供給には、電磁誘導方式や磁界共鳴方式を用いることができる。

【０４３２】

また、図２７（Ｃ）は、本発明の一態様の二次電池を用いた二輪車の一例である。図２７（Ｃ）に示すスクータ８６００は、二次電池８６０２、サイドミラー８６０１、方向指示灯８６０３を備える。二次電池８６０２は、方向指示灯８６０３に電気を供給することができる。

【０４３３】

また、図２７（Ｃ）に示すスクータ８６００は、座席下収納８６０４に、二次電池８６０２を収納することができる。二次電池８６０２は、座席下収納８６０４が小型であっても、座席下収納８６０４に収納することができる。二次電池８６０２は、取り外し可能となっており、充電時には二次電池８６０２を屋内に持って運び、充電し、走行する前に収納すればよい。

【０４３４】

本発明の一態様によれば、二次電池のサイクル特性が良好となり、二次電池の容量を大きくすることができる。よって、二次電池自体を小型軽量化することができる。二次電池自体を小型軽量化できれば、車両の軽量化に寄与するため、航続距離を向上させることができる。また、車両に搭載した二次電池を車両以外の電力供給源として用いることもできる。この場合、例えば電力需要のピーク時に商用電源を用いることを回避することができる。電力需要のピーク時に商用電源を用いることを回避できれば、省エネルギー、および二酸化炭素の排出の削減に寄与することができる。また、サイクル特性が良好であれば二次電池を長期に渡って使用できるため、コバルトをはじめとする希少金属の使用量を減らすことができる。

【０４３５】

本実施の形態は、他の実施の形態と適宜組み合わせて実施することが可能である。

【実施例１】

【０４３６】

本実施例では、本発明の一態様である正極活物質を作製し、該正極活物質についてＳＴＥＭで観察した結果、ＴＥＭ像を高速フーリエ変換した結果、およびエネルギー分散型Ｘ線解析（ＥＤＸ）した結果について説明する。また、該正極活物質を用いた二次電池の特性を評価した結果について説明する。

【０４３７】

［正極活物質の作製］

≪サンプル０１≫

本実施例では、サンプル０１の正極活物質として、第１の領域が有するリチウムと第１の遷移金属の複合酸化物として、コバルト酸リチウムを有し、第２の領域が有する、第２の遷移金属の酸化物として、チタン酸リチウムを有し、第３の領域が有する、典型元素の酸

化物として、酸化マグネシウムを有するものを作製した。
【０４３８】
本実施例では、出発原料としてコバルト酸リチウム粒子（日本化学工業株式会社製、商品名：Ｃ―２０Ｆ）を用いた。そのため本実施例では、実施の形態１で説明したステップ１２およびステップ１３を省略した。なお上記コバルト酸リチウム粒子は、粒径が約２０μｍであり、ＸＰＳで分析可能な領域にフッ素、マグネシウム、カルシウム、ナトリウム、シリコン、硫黄、リンを含むコバルト酸リチウム粒子である。
【０４３９】
次に、ステップ１４としてマグネシウムとフッ素を含むコバルト酸リチウム粒子にゾルゲル法によりチタンを含む材料を被覆した。具体的には、イソプロパノールにＴＴＩＰを溶解し、ＴＴＩＰのイソプロパノール溶液を作製した。そして該溶液に、コバルト酸リチウム粒子を混合した。ＴＴＩＰがマグネシウムとフッ素を含むコバルト酸リチウムに対して０．０１ｍｌ／ｇとなるように混合した。
【０４４０】
このマグネシウムとフッ素を有するコバルト酸リチウム粒子が混合された混合液を、マグネチックスターラーで４時間、２５℃、湿度９０％ＲＨの条件下で撹拌した。この処理により、雰囲気中の水とＴＴＩＰで加水分解および重縮合反応を起こさせ、マグネシウムとフッ素を有するコバルト酸リチウム粒子の表面に、チタンを含む層を形成させた。
【０４４１】
上記の処理を終えた混合液をろ過し、残渣を回収した。ろ過のフィルターには、桐山ろ紙（Ｎｏ．４）を用いた。
【０４４２】
回収した残渣を、７０℃で１時間、真空乾燥した。
【０４４３】
次に、チタンを有する材料で被覆されたコバルト酸リチウム粒子を加熱した。マッフル炉を用いて、乾燥空気の流量は１０Ｌ／ｍｉｎとし、８００℃（昇温２００℃／時間）、保持時間２時間で加熱した。乾燥空気は、露点―１０９℃以下のものを用いた。
【０４４４】
次に、加熱された粒子を室温まで冷却した。保持温度から室温までの降温時間は１０～１５時間とした。その後、解砕処理を行った。解砕処理は、ふるいにかけることにより行い、ふるいは目開きが５３μｍのものを用いた。
【０４４５】
最後に冷却された粒子を回収し、サンプル０１の正極活物質を得た。
【０４４６】
≪サンプル０２≫
サンプル０２は比較例として、チタンを含む材料を被覆せずに、マグネシウムとフッ素を有するコバルト酸リチウム粒子を加熱して作製した。
【０４４７】
マグネシウムとフッ素を有するコバルト酸リチウム粒子は、日本化学工業製（製品名；Ｃ―２０Ｆ）を用いた。
【０４４８】
このマグネシウムとフッ素を有するコバルト酸リチウム粒子を加熱した。加熱は、８００℃（昇温２００℃／時間）、保持時間２時間、酸素の流量を１０Ｌ／ｍｉｎとして行った。
【０４４９】
加熱した粉末をサンプル０１と同様に冷却しふるいにかけたものを、サンプル０２の正極活物質とした。
【０４５０】
サンプル０２は、内部にコバルト酸リチウムを有し、表層部にマグネシウムを含む領域を有する正極活物質であることが推測された。

【０４５１】
≪サンプル０３≫
サンプル０３は、比較例として、マグネシウムを有さないコバルト酸リチウム粒子に、ゾルゲル法でチタンを含む領域を形成した後、加熱を行って作製した。
【０４５２】
コバルト酸リチウム粒子は、日本化学工業製（製品名；Ｃ―１０Ｎ）を用いた。これはＸＰＳでマグネシウムが検出されず、フッ素が１原子％程度検出されるコバルト酸リチウム粒子である。
【０４５３】
このコバルト酸リチウム粒子に対して、サンプル０１と同様にゾルゲル法によりチタンを含む領域を形成し、乾燥し、加熱し、冷却してふるいにかけた。これをサンプル０３の正極活物質とした。
【０４５４】
サンプル０３は、内部にコバルト酸リチウムを有し、表層部にチタンを含む領域を有する正極活物質であることが推測された。
【０４５５】
≪サンプル０４≫
サンプル０４は、比較例として、コバルト酸リチウム粒子を加熱せずにそのまま用いた。
【０４５６】
コバルト酸リチウム粒子は、日本化学工業製（製品名；Ｃ―１０Ｎ）を用いた。
【０４５７】
サンプル０４は、被覆層をもたない正極活物質である。
【０４５８】
≪サンプル０５≫
サンプル０５は、比較例として、マグネシウムとフッ素を有するコバルト酸リチウム粒子を加熱せずにそのまま用いた。
【０４５９】
マグネシウムとフッ素を有するコバルト酸リチウム粒子は、日本化学工業製（製品名；Ｃ―２０Ｆ）を用いた。つまり、サンプル０５はサンプル０１で出発原料として用いたものである。
【０４６０】
サンプル０１からサンプル０５までの条件を表１に示す。
【０４６１】
【表１】

	条件
Sample01	$LiCoO_2$+Mg+F、Tiを含む材料で被覆、加熱あり
Sample02	$LiCoO_2$+Mg+F、加熱あり
Sample03	$LiCoO_2$、Tiを含む材料で被覆、加熱あり
Sample04	$LiCoO_2$、加熱なし
Sample05	LiCoO2+Mg+F、加熱なし

【０４６２】
［ＳＴＥＭ］
得られたサンプル０１の正極活物質を、電子顕微鏡（日本電子株式会社製ＪＥＭ―ＡＲＭ２００Ｆ、加速電圧２００ｋＶ）で観察した。得られた電子顕微鏡像を図２８に示す。図２８に示すように、正極活物質は３つの異なる領域、第１の領域１０１と、第２の領域１０２と、第３の領域１０３を有していると考えられた。第３の領域１０３は、第１の領域１０１と第２の領域１０２よりも明るい領域として観察された。また第１の領域１０１と第２の領域１０２の結晶の配向が一部で一致し、第２の領域１０２と第３の領域１０３の

結晶の配向が一部で一致した。
【０４６３】
［ＳＴＥＭ―ＦＦＴ］
図２８に示すＳＴＥＭ像中の１０３ＦＦＴで示す領域の、ＦＦＴ（高速フーリエ変換）像を図２９（Ａ１）に示した。図２９（Ａ２）は、図２９（Ａ１）の中心点Ｏを十字で示し、輝点Ａ、Ｂ、Ｃを丸で囲って示した図である。同様に、１０２ＦＦＴで示す領域のＦＦＴ像を図２９（Ｂ１）に示した。図２９（Ｂ２）は、図２９（Ｂ１）の中心点Ｏを十字で示し、輝点Ａ、Ｂ、Ｃを丸で囲って示した図である。また１０１ＦＦＴで示す領域のＦＦＴ像を図２９（Ｃ１）に示した。図２９（Ｃ２）は、図２９（Ｃ１）の中心点Ｏを十字で示し、輝点Ａ、Ｂ、Ｃを丸で囲って示した図である。
【０４６４】
図２９（Ａ２）に示した輝点Ａと中心点Ｏ間の距離は、ｄ　＝　０．２５６ｎｍであった。輝点Ｂと中心点Ｏ間の距離は、ｄ　＝　０．２４１ｎｍであった。輝点Ｃと中心点Ｏ間の距離は、ｄ　＝　０．２０９ｎｍであった。また、∠ＣＯＡ＝１２１°、∠ＣＯＢ＝５２°、∠ＡＯＢ＝６９°であった。これらの結果から、１０３ＦＦＴで示す領域は酸化マグネシウム（ＭｇＯ、立方晶）であることが推察された。
【０４６５】
同様に、図２９（Ｂ２）に示した輝点Ａと中心点Ｏ間の距離は、ｄ＝　０．２３８ｎｍであった。輝点Ｂと中心点Ｏ間の距離は、ｄ＝０．２２５ｎｍであった。輝点Ｃと中心点Ｏ間の距離は、ｄ＝０．１９８ｎｍであった。また、∠ＣＯＡ＝１２３°、∠ＣＯＢ＝５２°、∠ＡＯＢ＝７１°であった。これらの結果から、１０２ＦＦＴで示す領域はチタン酸リチウム（$LiTiO_2$、立方晶）であることが推察された。
【０４６６】
図２９（Ｃ２）に示した輝点Ａと中心点Ｏ間の距離は、ｄ＝０．２４０ｎｍであった。輝点Ｂと中心点Ｏ間の距離は、ｄ＝０．２３５ｎｍであった。輝点Ｃと中心点Ｏ間の距離は、ｄ＝０．１９６ｎｍであった。また、∠ＣＯＡ＝１２６°、∠ＣＯＢ＝５２°、∠ＡＯＢ＝７４°であった。これらの結果から、１０１ＦＦＴで示す領域はコバルト酸リチウム（$LiCoO_2$、Ｒｈｏｍｂｏｈｅｄｒａｌ）であることが推察された。
【０４６７】
［ＥＤＸ］
またサンプル０１の正極活物質の、高角散乱環状暗視野走査透過顕微鏡法（ＨＡＡＤＦ―ＳＴＥＭ）像およびＥＤＸを用いた元素マッピング像を図３０に示す。図３０（Ａ１）はＨＡＡＤＦ―ＳＴＥＭ像、図３０（Ａ２）は酸素原子マッピング像、図３０（Ｂ１）はコバルト原子マッピング像、図３０（Ｂ２）はフッ素原子マッピング像、図３０（Ｃ１）はチタン原子マッピング像、図３０（Ｃ２）はマグネシウム原子マッピング像である。なお、図３０（Ａ２）乃至図３０（Ｃ２）および図３１（Ａ２）乃至図３１（Ｃ２）のＥＤＸ元素マッピング像では、検出下限以下の場合は白で示し、カウントが増えるほど黒に近づくように示している。
【０４６８】
図３０（Ａ２）および図３０（Ｂ１）に示すように、酸素原子およびコバルト原子は正極活物質粒子全体に分布していることが明らかとなった。一方、図３０（Ｂ２）、図３０（Ｃ１）および図３０（Ｃ２）に示すようにフッ素原子、チタン原子およびマグネシウム原子は、正極活物質の表面に近い領域に偏在していることが明らかとなった。
【０４６９】
次にサンプル０５の比較例の正極活物質の、ＨＡＡＤＦ―ＳＴＥＭ像およびＥＤＸを用いた元素マッピング像を図３１に示す。図３１（Ａ１）はＨＡＡＤＦ―ＳＴＥＭ像、図３１（Ａ２）は酸素原子マッピング像、図３１（Ｂ１）はコバルト原子マッピング像、図３１（Ｂ２）はフッ素原子マッピング像、図３１（Ｃ１）はチタン原子マッピング像、図３１（Ｃ２）はマグネシウム原子マッピング像である。
【０４７０】

図３１（Ｂ２）および図３１（Ｃ２）に示すように、加熱を行っていないサンプル０５でも、マグネシウムおよびフッ素がある程度表面近傍に偏在していることが明らかとなった。

【０４７１】

［ＥＤＸ線状分析］

また、サンプル０１の正極活物質の表面近傍の断面について、ＴＥＭ―ＥＤＸで線状分析を行った結果を図３２に示す。図３２はサンプル０１の正極活物質の外と正極活物質の内部を結ぶ線上で検出されたデータをグラフにしたもので、距離０ｎｍは正極活物質の外、距離１４ｎｍは粒子の内部である。ＥＤＸは分析領域が広がりがちなため、電子線照射の中心だけでなくその周囲の元素も検出される場合がある。

【０４７２】

図３２に示すように、サンプル０１の正極活物質の表面近傍にはマグネシウムとチタンのピークが存在し、マグネシウムの分布の方が、チタンの分布よりも表面に近いことが明らかとなった。またマグネシウムのピークの方が、チタンのピークよりも表面に近いことが明らかとなった。また、コバルトと酸素は正極活物質粒子の最表面から存在していることが推測された。

【０４７３】

なお図３２ではフッ素はほとんど検出されなかったが、これはＥＤＸでは軽元素であるフッ素が検出されにくいためと考えられた。

【０４７４】

上記のＳＴＥＭ像、ＦＦＴ像およびＥＤＸを用いた元素マッピング像、ＥＤＸ線状分析から、サンプル０１は、本発明の一態様である、第１の領域としてコバルト酸リチウムを有し、第２の領域としてリチウムと、チタンと、コバルトと、酸素と、を有し、第３の領域としてマグネシウムと、酸素と、を有する正極活物質であることが確認された。また、サンプル０１では第２の領域の一部と第３の領域の一部が重なっていることが明らかとなった。

【０４７５】

また図３２のグラフでは、酸素の検出量は距離４ｎｍ以上で安定している。そこで本実施例では、この安定した領域の酸素の検出量の平均値O_{ave}を求め、平均値O_{ave}の５０％の値、０．５O_{ave}に最も近い測定値を示した測定点の距離ｘを、正極活物質の粒子の表面であると推定することとした。

【０４７６】

本実施例において、距離４ｎｍ以上１４ｎｍ以下の範囲の酸素の検出量の平均O_{ave}は６７４．２であった。６７４．２の５０％である３３７．１に最も近い測定値を示した測定点のｘ軸は、距離１．７１ｎｍであった。そこで、本実施例では図３２のグラフにおける距離１．７１ｎｍが、正極活物質の粒子の表面であると推定することとした。

【０４７７】

正極活物質粒子の表面が図３２中では距離１．７１ｎｍであるとすると、マグネシウムのピークは正極活物質粒子の表面から０．７２ｎｍ、チタンのピークは表面から１．００ｎｍであった。

【０４７８】

また、マグネシウム濃度がピークの１／５以上であるのは距離４．４２ｎｍ、つまり正極活物質粒子の表面から２．７１ｎｍまででであった。距離４．５７ｎｍ以上、つまり正極活物質粒子の表面から２．８６ｎｍ以上の深さでは、マグネシウムの測定値はピークの１／５未満であった。そのため、サンプル０１は、表面から深さ方向に２．７１ｎｍまでは第３の領域であることが明らかとなった。

【０４７９】

また、チタン濃度がピークの１／２以上であるのは距離２．１４ｎｍから距離３．４２ｎｍまででであった。つまり正極活物質粒子の表面から０．４３ｎｍ以上１．７１ｎｍ以下の範囲が第２の領域であることが明らかとなった。

【０４８０】
次に、上記で作製したサンプル０１～サンプル０５の正極活物質を用いた二次電池を作製し、該二次電池の充放電特性について評価した結果について説明する。
【０４８１】
［二次電池の作製］
上記で作製したサンプル０１～サンプル０５の正極活物質を用いた、ＣＲ２０３２タイプ（直径２０ｍｍ高さ３．２ｍｍ）のコイン型の二次電池を作製した。
【０４８２】
正極には、正極活物質（ＬＣＯ）と、アセチレンブラック（ＡＢ）と、ポリフッ化ビニリデン（ＰＶＤＦ）をＬＣＯ：ＡＢ：ＰＶＤＦ＝９５：２．５：２．５（重量比）で混合したスラリーを集電体に塗工したものを用いた。
【０４８３】
対極にはリチウム金属を用いた。
【０４８４】
電解液が有する電解質には、１ｍｏｌ／Ｌの六フッ化リン酸リチウム（ＬｉＰＦ$_6$）を用い、電解液には、エチレンカーボネート（ＥＣ）とジエチルカーボネート（ＤＥＣ）がＥＣ：ＤＥＣ＝３：７（体積比）、ビニレンカーボネート（ＶＣ）が２重量％で混合されたものを用いた。
【０４８５】
正極缶及び負極缶には、ステンレス（ＳＵＳ）で形成されているものを用いた。
【０４８６】
［充放電特性の評価］
次に、上記で作製したサンプル０１およびサンプル０５の二次電池の充放電特性の評価を行った。測定温度は２５℃とした。充電は４．６Ｖ（ＣＣＣＶ，０．５Ｃ―０．０１Ｃ）、放電は２．５Ｖ（ＣＣ，０．５Ｃ）で、それぞれ２０サイクル充放電を行った。なお、ここでの１Ｃは、正極活物質重量あたりの電流値で１３７ｍＡ／ｇとした。
【０４８７】
図３３に、サンプル０１の正極活物質を用いた二次電池の充放電特性のグラフを示す。図３３に示すように、広いプラトーを有する良好な充放電特性を示した。また２０サイクルの充放電のグラフはほぼ重なり、サイクル特性は良好であった。
【０４８８】
図３４に、比較例のサンプル０５の二次電池の充放電特性のグラフを示す。初期のサイクルでは良好な充放電特性を示したが、図中の矢印で示すように、サイクルを経るにつれて充放電容量が減少した。
【０４８９】
［サイクル特性の評価］
≪充電４．４Ｖ≫
サンプル０１およびサンプル０５の二次電池について、４．４Ｖ充電の場合のサイクル特性を評価した。サイクル特性測定温度は２５℃とした。充電は４．４Ｖ（ＣＣＣＶ，０．５Ｃ―０．０１Ｃ）、放電は２．５Ｖ（ＣＣ，０．５Ｃ）で行った。
【０４９０】
図３５に、充電４．４Ｖの場合のサイクル特性のグラフを示す。図中の実線はサンプル０１、点線はサンプル０５の正極活物質を有する二次電池のグラフである。図３５に示すように、サンプル０１を有する二次電池は、５０サイクルを経てもエネルギー密度の維持率は９９．５％であり、極めて良好なサイクル特性を示した。一方、サンプル０５を有する二次電池は、５０サイクルを経た時点のエネルギー密度の維持率は９４．３％であった。
【０４９１】
≪充電４．６Ｖ≫
サンプル０１～サンプル０４の二次電池について、充電４．６Ｖの場合のサイクル特性を評価した。測定温度は２５℃とした。充電は４．６Ｖ（ＣＣＣＶ，０．５Ｃ―０．０１Ｃ

）、放電は２．５Ｖ（ＣＣ，０．５Ｃ）で行った。
【０４９２】
図３６に、充電４．６Ｖの場合のサイクル特性のグラフを示す。図３６に示すように、本発明の一態様の正極活物質であるサンプル０１を有する二次電池は、４．６Ｖという高い電圧の充放電で５０サイクルを経てもエネルギー密度の維持率は９４．１％であり、極めて良好なサイクル特性を示した。一方、比較例のサンプル０２～サンプル０４の正極活物質を有する二次電池はサンプル０１より劣り、たとえばサンプル０４の場合、５０サイクルを経た時点のエネルギー密度の維持率は３３．２％であった。
【０４９３】
このように、本発明の一態様の正極活物質の構成は、４．４Ｖを超えるような高電圧で充放電を行った場合に顕著な効果を発揮することが明らかとなった。
【実施例２】
【０４９４】
本実施例では、本発明の一態様である正極活物質を作製し、実施例１と異なる分析を行った結果について説明する。また該正極活物質を用いた二次電池の特性を、実施例１と異なる条件で評価した結果について説明する。
【０４９５】
本実施例では正極活物質として、第１の領域が有するリチウムと第１の遷移金属の複合酸化物として、コバルト酸リチウムを有し、第２の領域が有する第２の遷移金属の酸化物として、チタン酸リチウムを有し、第３の領域が有する典型元素の酸化物として、酸化マグネシウムを有するものを作製した。
【０４９６】
［正極活物質の作製、二次電池の作製］
≪サンプル０６、サンプル０７≫
本実施例では、出発原料としてコバルト酸リチウム粒子（日本化学工業株式会社製、商品名：Ｃ―２０Ｆ）を用いた。
【０４９７】
次に、ステップ１４としてコバルト酸リチウム粒子にゾルゲル法によりチタン酸化物を被覆し、乾燥した。ＴＴＩＰをコバルト酸リチウムに対して０．００４ｍｌ／ｇとなるように混合したこと以外は実施例１と同様に行った。この、チタン酸化物で被覆した後、加熱する前のコバルト酸リチウム粒子を、サンプル０６ということとする。
【０４９８】
次に、サンプル０６のチタン酸化物で被覆されたコバルト酸リチウム粒子を加熱した。マッフル炉を用いて、酸素雰囲気、８００℃、保持時間２時間で加熱し、酸素の流量は１０Ｌ／ｍｉｎとした。
【０４９９】
その後実施例１と同様に冷却および回収して正極活物質を得た。加熱後の該正極活物質を、サンプル０７ということとする。
【０５００】
［ＴＥＭ―ＥＤＸ］
サンプル０６およびサンプル０７について、特に粒子に生じたクラックおよびその周辺について、ＴＥＭ―ＥＤＸを用いて分析を行った。
【０５０１】
まずチタンについてのＴＥＭ―ＥＤＸ面分析の結果を、図３７および図３８に示す。
【０５０２】
図３７は、加熱前のサンプル０６のＴＥＭ―ＥＤＸ分析結果である。図３７（Ａ）は、粒子表面とクラック部を含む断面ＴＥＭ像である。図３７（Ａ）中の１と付した丸で示す、粒子表面を含む領域の、ＨＡＡＤＦ―ＳＴＥＭ像を図３７（Ｂ１）に、Ｔｉマッピング像を図３７（Ｂ２）に示す。同様に、図３７（Ａ）中の２と付した丸で示す、クラック部のうち表面からの深さが約２０ｎｍの領域の、ＨＡＡＤＦ―ＳＴＥＭ像を図３７（Ｃ１）に

、Ｔｉマッピング像を図３７（Ｃ２）に示す。図３７（Ａ）中の３と付した丸で示す、クラック部のうち表面からの深さが約５００ｎｍの領域の、ＨＡＡＤＦ―ＳＴＥＭ像を図３７（Ｄ１）に、Ｔｉマッピング像を図３７（Ｄ２）に示す。図３７（Ａ）中の４と付した丸で示す、クラック部のうち表面からの深さが約１０００ｎｍの領域の、ＨＡＡＤＦ―ＳＴＥＭ像を図３７（Ｅ１）に、Ｔｉマッピング像を図３７（Ｅ２）に示す。なお、図３７乃至図４０のＥＤＸ元素マッピング像では、検出下限以下の場合は黒で示し、カウントが増えるほど白に近づくように示している。

【０５０３】

図３８は、加熱後のサンプル０７のＴＥＭ―ＥＤＸ分析結果である。図３８（Ａ）は、粒子表面とクラック部を含む断面ＴＥＭ像である。図３８（Ａ）中の１と付した丸で示す、粒子表面を含む領域の、ＨＡＡＤＦ―ＳＴＥＭ像を図３８（Ｂ１）に、Ｔｉマッピング像を図３８（Ｂ２）に示す。同様に、図３８（Ａ）中の２と付した丸で示す、クラック部のうち表面からの深さが約２０ｎｍの領域の、ＨＡＡＤＦ―ＳＴＥＭ像を図３８（Ｃ１）に、Ｔｉマッピング像を図３８（Ｃ２）に示す。図３８（Ａ）中の３と付した丸で示す、クラック部のうち表面からの深さが約５００ｎｍの領域の、ＨＡＡＤＦ―ＳＴＥＭ像を図３８（Ｄ１）に、Ｔｉマッピング像を図３８（Ｄ２）に示す。図３８（Ａ）中の４と付した丸で示す、クラック部のうち表面からの深さが約１０００ｎｍの領域の、ＨＡＡＤＦ―ＳＴＥＭ像を図３８（Ｅ１）に、Ｔｉマッピング像を図３８（Ｅ２）に示す。

【０５０４】

図３７と図３８に示すように、チタンは、加熱前のサンプル０６では粒子表面に偏析している様子が観察されたものの、クラック部では偏析は確認されなかった。一方、加熱後のサンプル０７では、チタンは粒子表面とクラック部の両方において、偏析が観察された。つまり加熱により、チタンがクラック部の界面に偏析することが明らかとなった。

【０５０５】

次にマグネシウムについてのＴＥＭ―ＥＤＸ面分析の結果を、図３９および図４０に示す。

【０５０６】

図３９（Ａ）は、図３７（Ａ）と同じサンプル０６の断面ＴＥＭ像である。図３９（Ｂ１）、図３９（Ｃ１）、図３９（Ｄ１）および図３９（Ｅ１）は、図３７（Ｂ１）、図３７（Ｃ１）、図３７（Ｄ１）および図３７（Ｅ１）と同じＨＡＡＤＦ―ＳＴＥＭ像である。図３９（Ｂ１）と同じ領域のＭｇマッピング像を図３９（Ｂ２）に示す。図３９（Ｃ１）と同じ領域のＭｇマッピング像を図３９（Ｃ２）に示す。図３９（Ｄ１）と同じ領域のＭｇマッピング像を図３９（Ｄ２）に示す。図３９（Ｅ１）と同じ領域のＭｇマッピング像を図３９（Ｅ２）に示す。

【０５０７】

図４０（Ａ）は、図３８（Ａ）と同じサンプル０７の断面ＴＥＭ像である。図４０（Ｂ１）、図４０（Ｃ１）、図４０（Ｄ１）および図４０（Ｅ１）は、図３８（Ｂ１）、図３８（Ｃ１）、図３８（Ｄ１）および図３８（Ｅ１）と同じＨＡＡＤＦ―ＳＴＥＭ像である。図４０（Ｂ１）と同じ領域のＭｇマッピング像を図４０（Ｂ２）に示す。図４０（Ｃ１）と同じ領域のＭｇマッピング像を図４０（Ｃ２）に示す。図４０（Ｄ１）と同じ領域のＭｇマッピング像を図４０（Ｄ２）に示す。図４０（Ｅ１）と同じ領域のＭｇマッピング像を図４０（Ｅ２）に示す。

【０５０８】

図３９と図４０に示すように、マグネシウムは、加熱前のサンプル０６では粒子表面、クラック部ともに偏析は確認されなかった。一方、加熱後のサンプル０７では、マグネシウムは粒子表面とクラック部の両方において、偏析が観察された。

【０５０９】

次に、チタンとマグネシウムを定量するため、図３７（Ａ）中で１～６を付した丸で示す領域、および図３８（Ａ）中で１～６を付した丸で示す領域について、ＥＤＸ点分析を行った。それぞれの領域の範囲内で、２箇所測定を行った。

【０５１０】
図４１に、ＥＤＸ点分析の結果を、チタンとコバルトの原子数比で示す。図４１（Ａ）は加熱前のサンプル０６の結果である。図４１（Ａ）における１～６の検出箇所は、それぞれ図３７（Ａ）中で１～６を付した丸で示す領域内である。図４１（Ｂ）は、加熱後のサンプル０７の結果である。図４１（Ｂ）における１～６の検出箇所は、それぞれ図３８（Ａ）中で１～６を付した丸で示す領域内である。
【０５１１】
図４１に示すように、サンプル０６のクラック部はいずれの測定箇所でもＴｉ／Ｃｏが０．０１以下であった。一方、サンプル０７のクラック部ではチタンが増加した箇所が多く、Ｔｉ／Ｃｏが０．０５以上の測定箇所もあった。また、サンプル０７の粒子表面におけるＴｉ／Ｃｏは０．１０～０．１８の間であった。
【０５１２】
次に、図４２にＥＤＸ点分析の結果を、マグネシウムとコバルトの原子数比で示す。検出箇所は図４１と同じである。
【０５１３】
図４２に示すように、サンプル０６では、粒子表面、クラック部いずれにおいてもＭｇ／Ｃｏが０．０３以下であった。一方サンプル０７では粒子表面、クラック部ともにマグネシウムが増加した箇所が多かった。粒子表面におけるＭｇ／Ｃｏは０．１５～０．５０の間であり、クラック部では０～０．２２の範囲であった。
【０５１４】
［二次電池の作製］
次に、加熱後のサンプル０７の正極活物質を用いてＣＲ２０３２タイプのコイン型の二次電池を作製した。正極には、サンプル０２の正極活物質（ＬＣＯ）と、ＡＢと、ポリフッ化ビニリデン（ＰＶＤＦ）をＬＣＯ：ＡＢ：ＰＶＤＦ＝９５：３：２（重量比）で混合したスラリーを正極集電体に塗工したものを用いた。正極集電体には厚さ２０μｍのアルミニウム箔を用いた。正極活物質と、ＡＢと、ＰＶＤＦを含む正極活物質層の担持量は７．６ｍｇ／ｃｍ^{2}とした。
【０５１５】
対極にはリチウム金属を用いた。
【０５１６】
電解液には、エチレンカーボネート（ＥＣ）とジエチルカーボネート（ＤＥＣ）がＥＣ：ＤＥＣ＝３：７（体積比）で混合されたものに、１ｍｏｌ／ＬのＬｉＰＦ$_6$を溶解し、ビニレンカーボネート（ＶＣ）が２ｗｔ％で添加されたものを用いた。
【０５１７】
［初期特性、レート特性］
上記で作製したサンプル０７の正極活物質を用いた二次電池について、初期特性およびレート特性を測定した。
【０５１８】
初期特性の測定は、充電をＣＣＣＶ、０．２Ｃ、４．６Ｖ、カットオフ電流０．０５Ｃで行った。放電をＣＣ、０．２Ｃ、カットオフ電圧３．０Ｖで行った。なおここでは１Ｃは正極活物質の容量を１６０ｍＡｈ／ｇとして算出した。測定温度は２５℃とした。初期特性を測定した結果を表２に示す。
【０５１９】
【表２】

初回充電容量 [mAh/g]	初回放電容量 [mAh/g]	初回充放電効率 [%]
221.2	217.5	98.3

【０５２０】

初期特性測定後にレート特性を測定した。放電レートを変化させ、放電レート以外は初期特性測定と同じ条件で、０．２Ｃ充電／０．２Ｃ放電、０．２Ｃ充電／０．５Ｃ放電、０．２Ｃ充電／１．０Ｃ放電、０．２Ｃ充電／２．０Ｃ放電、０．２Ｃ充電／３．０Ｃ、０．２Ｃ充電／４．０Ｃ、０．２Ｃ充電／５．０Ｃの順で測定した。測定温度は２５℃とした。

【０５２１】

初期特性およびレート特性を測定した結果を表３に示す。また各レートの放電カーブを図４３に示す。

【０５２２】

【表３】

レート [C]	放電容量 [mAh/g]	放電容量維持率 [%]
0.2	218.2	100.0
0.5	215.8	98.9
1	213.1	97.7
2	207.9	95.3
3	204.0	93.5
4	198.8	91.1
5	184.3	84.5

【０５２３】

［温度特性］

次に、正極活物質層の担持量を８．２ｍｇ／ｃｍ^{2}とした他はレートを評価したセルと同様の条件のセルを作製し、温度特性を評価した。充電は全て２５℃で、ＣＣＣＶ、０．２Ｃ、４．６Ｖ、カットオフ電流０．０５Ｃで行った。放電は２５℃、０℃、－１０℃、－２０℃、または４５℃で、ＣＣ、０．２Ｃ、カットオフ電圧３．０Ｖで行った。温度特性の結果を図４４に示す。

【０５２４】

［サイクル特性］

次に、温度特性を測定したセルと同様の条件のセルを作製し、サイクル特性を測定した。サイクル特性では、充電をＣＣＣＶ、１．０Ｃ、４．５５Ｖ、カットオフ電流０．０５Ｃ、放電をＣＣ、１．０Ｃ、カットオフ電圧３．０Ｖで行った。サイクル特性の測定温度は４５℃とし、１００サイクル測定した。１００サイクル後の放電容量維持率は８６％であった。測定したサイクル特性について放電容量維持率でグラフにしたものを図４５に示す。

【０５２５】

また、サンプル０７の正極活物質の比表面積を測定した結果は、０．１３ｍ^2／ｇであった。

【０５２６】

また、サンプル０７の正極活物質の粒度分布を測定した結果、平均粒径は２１．５μｍ，１０％Ｄは１３．１μｍ、５０％Ｄは２２．０μｍ、９０％Ｄは３４．４μｍであった。

【０５２７】

また、サンプル０７の正極活物質のタップ密度は２．２１ｇ／ｃｍ^{3}であった。タップ密度の測定には、ＭＵＬＴＩ　ＴＥＳＴＥＲ　ＭＴ－１０００（セイシン企業製）を用いた。

【０５２８】

以上のように、本発明の一態様であるサンプル０７の正極活物質は良好な初期特性、レート特性、サイクル特性および温度特性を示すことが明らかになった。特に初回充放電効率

が９８％以上と高く、副反応が抑制されていることが推測された。また、２Ｃという高い放電レートにおいても、０．２Ｃを基準として９６．１％という良好な容量を示した。
【実施例３】
【０５２９】
本実施例では、表層部にチタンとマグネシウムを含む領域を有する正極活物質について、出発原料のＬｉ／第１の遷移金属比を変化させて作製し、特性を評価した結果を示す。
【０５３０】
［正極活物質の作製］
本実施例では、第１の遷移金属としてコバルトを用いた、サンプル１１～サンプル１７、サンプル２１～サンプル２８、サンプル３１～サンプル４０の正極活物質を用意した。各サンプルの作製方法および条件は以下の通りとした。
【０５３１】
≪サンプル１１～１７≫
まず、出発原料となるリチウム源、コバルト源、マグネシウム源およびフッ素源をそれぞれ秤量した。本実施例ではリチウム源として炭酸リチウム、コバルト源として酸化コバルト、マグネシウム源として酸化マグネシウム、フッ素源およびリチウム源としてフッ化リチウムを用いた。
【０５３２】
このとき、
サンプル１１は、出発原料のＬｉ／Ｃｏ比が１．００となるように秤量した。
サンプル１２は、出発原料のＬｉ／Ｃｏ比が１．０３となるように秤量した。
サンプル１３は、出発原料のＬｉ／Ｃｏ比が１．０５となるように秤量した。
サンプル１４は、出発原料のＬｉ／Ｃｏ比が１．０６となるように秤量した。
サンプル１５は、出発原料のＬｉ／Ｃｏ比が１．０７となるように秤量した。
サンプル１６は、出発原料のＬｉ／Ｃｏ比が１．０８となるように秤量した。
サンプル１７は、出発原料のＬｉ／Ｃｏ比が１．１３となるように秤量した。
【０５３３】
また、サンプル１１～サンプル１７に共通して、出発原料に含まれるコバルトの原子数を１としたときに、マグネシウムの原子数が０．０１、フッ素の原子数が０．０２となるように秤量した。
【０５３４】
次に、秤量した出発原料を、サンプルごとにボールミルを用いて混合した。
【０５３５】
次に、混合した出発材料を焼成した。焼成は１０００℃で１０時間、昇温は２００℃／ｈ、乾燥空気の流量は１０Ｌ／ｍｉｎとした。
【０５３６】
上記の工程で、リチウム、コバルト、フッ素、マグネシウムを含む複合酸化物の粒子を合成した。
【０５３７】
次に、２－プロパノールに、正極活物質重量当たりのＴＴＩＰが０．０１ｍｌ／ｇとなるようＴＴＩＰを加えて混合し、テトラ－ｉ－プロポキシチタンの２－プロパノール溶液を作製した。
【０５３８】
このＴＴＩＰの２－プロパノール溶液に、リチウム、コバルト、フッ素、マグネシウムを含む複合酸化物の粒子を加え、混合した。
【０５３９】
上記の混合液を、マグネチックスターラーで４時間、２５℃、湿度９０％ＲＨの条件下で撹拌した。この処理により、雰囲気中の水とＴＴＩＰで加水分解および重縮合反応を起こさせ、マグネシウムとフッ素を有するコバルト酸リチウム粒子の表面に、チタンを含む層を形成させた。

【０５４０】
上記の処理を終えた混合液をろ過し、残渣を回収した。ろ過のフィルターには、桐山ろ紙（Ｎｏ．４）を用いた。
【０５４１】
回収した残渣を、７０℃で１時間、真空乾燥した。
【０５４２】
乾燥させた粉末を加熱した。加熱は、８００℃（昇温２００℃／時間）、保持時間２時間、酸素雰囲気下で行った。
【０５４３】
加熱した粉末を冷却し、解砕処理を行った。解砕処理は、ふるいにかけることにより行い、ふるいは目開きが５３μｍのものを用いた。
【０５４４】
解砕処理を終えた粒子を、サンプル１１～サンプル１７の正極活物質とした。
【０５４５】
≪サンプル２１～２７≫
サンプル２１～サンプル２７は、出発原料はサンプル１１～サンプル１６と同じものを用いた。このとき、
サンプル２１は、出発原料のＬｉ／Ｃｏ比が１．００となるように秤量した。
サンプル２２は、出発原料のＬｉ／Ｃｏ比が１．０３となるように秤量した。
サンプル２３は、出発原料のＬｉ／Ｃｏ比が１．０５となるように秤量した。
サンプル２４は、出発原料のＬｉ／Ｃｏ比が１．０６となるように秤量した。
サンプル２５は、出発原料のＬｉ／Ｃｏ比が１．０７となるように秤量した。
サンプル２６は、出発原料のＬｉ／Ｃｏ比が１．０８となるように秤量した。
サンプル２７は、出発原料のＬｉ／Ｃｏ比が１．１３となるように秤量した。
【０５４６】
サンプル２１～サンプル２７は、ＴＴＩＰの２‐プロパノール溶液の濃度を正極活物質重量当たりのＴＴＩＰが０．０２ｍｌ／ｇとなるようにした他は、サンプル１１～サンプル１７と同様に作製した。
【０５４７】
≪サンプル２８≫
サンプル２８は、出発原料のＬｉ／Ｃｏ比およびＴＴＩＰ量をサンプル２３と同じとした。つまりサンプル２８は、出発原料のＬｉ／Ｃｏ比が１．０５となるように秤量し、正極活物質重量当たりのＴＴＩＰが０．０２ｍｌ／ｇとなるようにした。
【０５４８】
ただしサンプル２８では、出発材料を混合した後、焼成を９５０℃で行った。
【０５４９】
焼成温度の他は、サンプル２３と同様に作製した。
【０５５０】
サンプル１１～１７、サンプル２１～２８は、内部にコバルト酸リチウムを有し、表層部にチタンとマグネシウムを含む領域を有する正極活物質であることが推測された。
【０５５１】
≪サンプル３１～４０≫
サンプル３１～サンプル４０は、比較例として、チタン含む領域を形成せずに作製した。
【０５５２】
サンプル３１は、出発原料のＬｉ／Ｃｏ比が１．００となるように秤量した。サンプル３２は、出発原料のＬｉ／Ｃｏ比が１．０１となるように秤量した。サンプル３３は、出発原料のＬｉ／Ｃｏ比が１．０２となるように秤量した。サンプル３４は、出発原料のＬｉ／Ｃｏ比が１．０３となるように秤量した。サンプル３５は、出発原料のＬｉ／Ｃｏ比が１．０３５となるように秤量した。サンプル３６は、出発原料のＬｉ／Ｃｏ比が１．０４となるように秤量した。サンプル３７は、出発原料のＬｉ／Ｃｏ比が１．０５１となるよ

うに秤量した。サンプル３８は、出発原料のＬｉ／Ｃｏ比が１．０６１となるように秤量した。サンプル３９は、出発原料のＬｉ／Ｃｏ比が１．０８１となるように秤量した。サンプル４０は、出発原料のＬｉ／Ｃｏ比が１．１３０となるように秤量した。

【０５５３】

また、サンプル３１～サンプル４０に共通して、出発原料に含まれるコバルトの原子数を１としたときに、マグネシウムの原子数が０．０１、フッ素の原子数が０．０２となるように秤量した。

【０５５４】

次に、秤量した出発原料を、サンプルごとにボールミルを用いて混合した。

【０５５５】

次に、混合した出発材料を焼成した。焼成は１０００℃で１０時間、昇温は２００℃／ｈ、乾燥空気の流量は１０Ｌ／ｍｉｎとした。

【０５５６】

上記の工程で、リチウム、コバルト、フッ素、マグネシウムを含む複合酸化物の粒子を合成した。

【０５５７】

合成した粒子を冷却してから、加熱した。加熱は、８００℃（昇温２００℃／時間）、保持時間２時間、酸素雰囲気下で行った。

【０５５８】

加熱した粉末を冷却し、解砕処理を行った。解砕処理は、ふるいにかけることにより行い、ふるいは目開きが５３μｍのものを用いた。

【０５５９】

解砕処理を終えた粒子を、サンプル３１～サンプル４０の正極活物質とした。

【０５６０】

サンプル１１～サンプル１７、サンプル２１～サンプル２８、サンプル３１～サンプル４０の作製条件を表４に示す。

【０５６１】

【表４】

	Li/Co	TTIP	焼成温度
Sample11	1.00		
Sample12	1.03		
Sample13	1.05		
Sample14	1.06	0.01ml/g	1000℃
Sample15	1.07		
Sample16	1.08		
Sample17	1.13		
Sample21	1.00		
Sample22	1.03		
Sample23	1.05		
Sample24	1.06	0.02ml/g	1000℃
Sample25	1.07		
Sample26	1.08		
Sample27	1.13		
Sample28	1.05	0.02ml/g	950℃
Sample31	1.00		
Sample32	1.01		
Sample33	1.02		
Sample34	1.03		
Sample35	1.035	–	1000℃
Sample36	1.04		
Sample37	1.051		
Sample38	1.061		
Sample39	1.081		
Sample40	1.130		

【０５６２】

［ＸＰＳ］

サンプル１１～サンプル１７、サンプル２１～サンプル２８、サンプル３１～サンプル４０の正極活物質について、ＸＰＳ分析を行った。サンプル１１～サンプル１６のＸＰＳ分析の結果を表５、サンプル２１～サンプル２８のＸＰＳ分析の結果を表６、サンプル３１～サンプル４０のＸＰＳ分析の結果を表７に示す。なお、表５～表７では各元素の濃度は、コバルトを１としたときの相対値で示した。

【０５６３】

【表５】

	Coを1としたときの相対値											
	Li/Co	Li	Co	O	C	F	Mg	Ti	Si	Ca	Na	S
Sample11	1.00	0.82	1.00	2.87	0.25	0.37	0.57	0.15	0.00	0.04	0.27	0.01
Sample12	1.03	0.69	1.00	2.84	0.93	0.63	0.63	0.14	0.00	0.05	0.04	0.00
Sample13	1.05	0.96	1.00	3.22	0.66	0.47	0.61	0.14	0.00	0.03	0.09	0.03
Sample14	1.06	1.04	1.00	3.13	0.34	0.77	0.77	0.15	0.00	0.05	0.34	0.04
Sample15	1.07	0.84	1.00	3.42	0.58	0.60	0.23	0.18	0.00	0.08	0.27	0.00
Sample16	1.08	1.08	1.00	3.10	0.66	0.59	0.04	0.16	0.00	0.03	0.16	0.02
Sample17	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】

【表６】

	Coを1としたときの相対値											
	Li/Co	Li	Co	O	C	F	Mg	Ti	Si	Ca	Na	S
Sample21	1.00	1.08	1.00	3.23	0.47	0.45	0.49	0.20	0.00	0.06	0.35	0.01
Sample22	1.03	0.73	1.00	3.05	1.06	0.70	0.59	0.17	0.00	0.03	0.09	0.00
Sample23	1.05	1.19	1.00	3.40	0.73	0.48	0.58	0.23	0.00	0.05	0.10	0.04
Sample24	1.06	1.1●	1.00	3.44	0.33	0.92	0.91	0.25	0.00	0.05	0.30	0.04
Sample25	1.07	1.05	1.00	4.26	0.74	0.20	0.25	0.29	0.00	0.07	0.30	0.02
Sample26	1.08	1.12	1.00	3.32	0.74	0.52	0.14	0.21	0.00	0.05	0.15	0.00
Sample27	1.13	1.49	1.00	3.80	0.60	0.41	0.00	0.12	0.00	0.00	0.16	0.02
Sample28	1.05	0.99	1.00	3.11	0.72	0.8●	0.56	0.18	0.00	0.05	0.12	0.02

【０５６５】
【表７】

	Coを1としたときの相対値								
	Li/Co	Li	Co	O	C	F	Mg	Ca	Na
Sample31	1.00	0.51	1.00	2.45	0.69	0.08	0.27	0.03	0.08
Sample32	1.01	0.67	1.00	2.65	0.77	0.08	0.28	0.03	0.08
Sample33	1.02	0.53	1.00	2.51	0.66	0.09	0.27	0.02	0.06
Sample34	1.03	0.79	1.00	2.93	0.92	0.09	0.35	0.04	0.14
Sample35	1.04	0.65	1.00	2.33	0.48	0.11	0.32	0.03	0.11
Sample36	1.04	0.6●	1.00	2.73	0.56	0.11	0.38	0.05	0.16
Sample37	1.05	0.67	1.00	3.04	0.64	0.09	0.35	0.04	0.21
Sample38	1.06	0.83	1.00	2.65	1.03	0.29	0.10	0.05	0.11
Sample39	1.08	0.80	1.00	2.79	1.04	0.26	0.03	0.08	0.10
Sample40	1.13	0.77	1.00	2.72	0.22	0.99	0.00	0.01	0.26

【０５６６】
また表５～表７の分析結果から、マグネシウム濃度とチタン濃度について抽出したグラフを図４６に示す。図４６（Ａ）はＬｉ／Ｃｏ比とマグネシウム濃度のグラフ、図４６（Ｂ）はＬｉ／Ｃｏ比とチタン濃度のグラフである。
【０５６７】
まず図４６（Ａ）のサンプル３１～サンプル４０から、チタンを含む被覆層をもたない場合、Ｌｉ／Ｃｏ比が１．００以上１．０５以下のサンプルでマグネシウム濃度が高くなっていることが明らかとなった。これは、加熱により、出発原料に含まれたマグネシウムがＸＰＳで検出可能な範囲に偏析したためと考えられる。一方、Ｌｉ／Ｃｏ比が１．０６以上ではマグネシウムの濃度が低くなり、リチウムが過剰となりすぎるとマグネシウムの偏析が起こりにくくなることが推測された。
【０５６８】
また図４６（Ａ）のサンプル１１～サンプル１６およびサンプル２１～サンプル２６から、表層部にチタンを含む領域を有すると、有さない場合よりもＸＰＳで検出可能な範囲のマグネシウム濃度が高くなることが明らかとなった。
【０５６９】
さらに、Ｌｉ／Ｃｏ比が１．０６の場合、チタンを含む領域を有さない場合はＸＰＳで検出可能な範囲のマグネシウム濃度が低くなっているのに対して、チタンを含む領域を有するサンプルではＸＰＳで検出可能な範囲のマグネシウム濃度が高かった。つまり、表層部にチタンを含む領域を形成することで、Ｌｉ／Ｃｏ比が高い場合でも、マグネシウムの偏

析が十分に起こることが明らかとなった。
【０５７０】
なおチタンを含む領域を有しても、Ｌｉ／Ｃｏ比が１．０７の場合は、１．０６の場合よりもマグネシウム濃度が低下した。またＬｉ／Ｃｏ比が１．０８以上の場合は、チタンを含む領域を有しても、マグネシウムの偏析が起こりにくくなることが推測された。
【０５７１】
［サイクル特性の評価］
≪エネルギー密度維持率≫
次に、サンプル１１～サンプル１４、サンプル１６およびサンプル２１～サンプル２４、サンプル２６の正極活物質を用いて、実施例１と同様にサイクル特性を評価した。
【０５７２】
二次電池の形状、正極における正極活物質、導電助剤、バインダの材料および混合比、対極、電解液、外装体、サイクル特性試験の条件等は実施例１と同じとした。
【０５７３】
図４７（Ａ）に、正極活物質あたりのＴＴＩＰが０．０１ｍｌ／ｇとなるように作製したサンプル１１～サンプル１４、サンプル１６の正極活物質を用いた二次電池の、４．６Ｖ充電時のエネルギー密度維持率と充放電サイクル数のグラフを示す。図４７（Ｂ）に正極活物質あたりのＴＴＩＰが０．０２ｍｌ／ｇとなるように作製したサンプル２１～サンプル２４、サンプル２６の正極活物質を用いた二次電池の、４．６Ｖ充電時のエネルギー密度維持率と充放電サイクル数のグラフを示す。
【０５７４】
図４７（Ａ）から明らかなように、ＴＴＩＰが０．０１ｍｌ／ｇの場合、サンプル１１～サンプル１４、つまりＬｉ／Ｃｏ比が１．００以上１．０６以下の正極活物質は良好なサイクル特性を示した。特に、サンプル１１およびサンプル１２、つまりＬｉ／Ｃｏ比が１．００以上１．０３以下の正極活物質は極めて良好なサイクル特性を示した。一方、Ｌｉ／Ｃｏ比が１．０８のサンプル１６では比較的早い段階でエネルギー密度維持率が悪化していた。
【０５７５】
また図４７（Ｂ）から明らかなように、ＴＴＩＰが０．０２ｍｌ／ｇの場合、サンプル２１～サンプル２４、つまりＬｉ／Ｃｏ比が１．００以上１．０６以下の正極活物質は良好なサイクル特性を示した。特に、サンプル２３およびサンプル２４、つまりＬｉ／Ｃｏ比が１．０５以上１．０６以下の正極活物質は極めて良好なサイクル特性を示した。
【０５７６】
図４８に、サンプル１１～サンプル１５の中で最も良好なサイクル特性を示したサンプル１１と、サンプル２１～サンプル２５の中で最も良好なサイクル特性を示したサンプル２３を比較したグラフを示す。
【０５７７】
図４８から明らかなように、両者とも極めて良好なサイクル特性を示したが、ＴＴＩＰが０．０２ｍｌ／ｇのサンプル２３の方がより良好なサイクル特性であった。
【０５７８】
≪放電容量維持率≫
次に、サンプル２１～サンプル２６およびサンプル２８について、放電容量維持率でサイクル特性を評価した結果を図４９に示す。
【０５７９】
サンプル２１～サンプル２６の二次電池の形状、正極における正極活物質、導電助剤、バインダの材料および混合比、対極、電解液、外装体、サイクル特性試験の条件等は実施例１と同じとした。
【０５８０】
サンプル２８を用いた二次電池は、バインダとしてＰＶＤＦを用い、正極活物質（ＬＣＯ）と、ＡＢと、ＰＶＤＦをＬＣＯ：ＡＢ：ＰＶＤＦ＝９５：３：２（重量比）で混合した

他は、サンプル２１～サンプル２６を用いた二次電池と同様に作製し評価した。
【０５８１】
図４９から明らかなように、サンプル２１～サンプル２４、およびサンプル２８は良好なサイクル特性を示した。中でも、サンプル２８は極めて良好なサイクル特性を示した。サンプル２８では、５０サイクル後の放電容量維持率は８５％以上であった。
【０５８２】
一方、Ｌｉ／Ｃｏ比が１．０７および１．０８であるサンプル２５およびサンプル２６は、比較的早い段階から放電容量維持率が悪化していた。
【０５８３】
以上の結果から、正極活物質重量あたりのＴＴＩＰが０．０２ｍｌ／ｇの場合、好ましいＬｉ／Ｃｏ比の範囲は１．００以上１．０７未満であることが明らかとなった。さらに、Ｌｉ／Ｃｏ比の範囲が１．０５以上１．０６以下であると極めて良好なサイクル特性を示すことが明らかとなった。
【０５８４】
図４９で極めて良好なサイクル特性を示したサンプル２８、サンプル２４、および比較的早い段階で悪化がみられたサンプル２５を用いた二次電池の、充放電カーブを図５０に示す。
【０５８５】
図５０（Ａ）はサンプル２８、図５０（Ｂ）はサンプル２４、図５０（Ｃ）はサンプル２５を用いた二次電池の充放電カーブである。それぞれ５０回充放電を繰り返した結果を重ねて示した。図中の矢印のように、１サイクル目から５０サイクル目に向かって充放電容量が減少している。
【０５８６】
図５０（Ａ）および図５０（Ｂ）に示すように、本発明の一態様の正極活物質であるサンプル２８およびサンプル２４は、高い充放電容量および良好な充放電特性を示した。また図５０（Ｃ）のサンプル２５と比較して、図５０（Ａ）および図５０（Ｂ）のサンプル２８およびサンプル２４は充放電容量の減少が大幅に抑制されたことが明らかとなった。
【実施例４】
【０５８７】
本実施例では、実施例２で作製したサンプル２４の正極活物質についてＳＥＭ観察およびＳＥＭ―ＥＤＸ分析を行った結果について説明する。
【０５８８】
サンプル２４は、Ｌｉ／Ｃｏ＝１．０６、正極活物質重量当たりのＴＴＩＰが０．０２ｍｌ／ｇとなるよう作製したサンプルである。サンプル２４のＳＥＭ像を図５１（Ａ）に示す。図５１（Ａ）の一部を拡大した像を図５１（Ｂ）および図５１（Ｃ）に示す。
【０５８９】
図５１から明らかなように、正極活物質の表層部には凸状の領域が多数存在した。
【０５９０】
次に、サンプル２４の正極活物質についてＳＥＭ―ＥＤＸを用いて分析した結果を図５２に示す。図５２（Ａ―１）は正極活物質表層部のＳＥＭ像、図５２（Ａ―２）はチタンのマッピング、図５２（Ｂ―１）はマグネシウムのマッピング、図５２（Ｂ―２）は酸素のマッピング、図５２（Ｃ―１）はアルミニウムのマッピング、図５２（Ｃ―２）はコバルトのマッピングである。なお、図５２のＥＤＸ元素マッピング像では、検出下限以下の場合は黒で示し、カウントが増えるほど白に近づくように示している。
【０５９１】
図５２（Ａ―１）、図５２（Ａ―２）、図５２（Ｂ―１）の図中の同じ領域を点線で囲った。点線で囲った領域を比較すると明らかなように、正極活物質表層部の凸状の領域には、チタンおよびマグネシウムが分布していた。
【０５９２】
そのため、サンプル２４は、第３の領域１０３上に、チタンおよびマグネシウムを有する

凸状の第４の領域１０４を有する正極活物質であることが確認された。
【０５９３】
実施例２で示したように、サンプル２４は極めて良好なサイクル特性を示した条件の一つである。そのため表層部に第４の領域が存在しても、または第４の領域が存在することで、良好なサイクル特性を示す正極活物質を得られることが明らかとなった。
【０５９４】
以上の実施例１乃至実施例３の結果から、表層部にチタンを含む領域を形成することで、良好なサイクル特性を示す正極活物質が得られることが明らかとなった。また、正極活物質の粒径を大きくするためにＬｉ／Ｃｏ比を大きくすると、サイクル特性が悪化する懸念があるが、表層部にチタンを含む領域を形成することで、良好なサイクル特性を得られるＬｉ／Ｃｏ比の範囲を広くできることが明らかとなった。また、正極活物質の表層部にチタンおよびマグネシウムを有する第４の領域が存在しても、良好なサイクル特性を示すことが明らかとなった。
【実施例５】
【０５９５】
本実施例では、酸化グラフェンで被覆された正極活物質の製造方法の一例を示し、その方法で作製された正極活物質を電子顕微鏡で観察した結果について説明する。
【０５９６】
図５３の工程フロー図に示すように、正極活物質への被膜形成工程は、（Ｓ１１）酸化グラフェンの秤量と、（Ｓ１２）酸化グラフェンと純水との混合及び攪拌、（Ｓ１３）ｐＨの制御、（Ｓ１４）活物質の投入、（Ｓ１５）懸濁液の完成、（Ｓ１６）スプレードライ装置を用いた懸濁液の噴霧処理、（Ｓ１７）容器への粉末の回収、を包含する。
【０５９７】
なお、（Ｓ１２）において純水を分散媒として用いているが特に限定されずエタノールなどを用いてもよい。また、（Ｓ１４）において活物質は、正極活物質とする。
【０５９８】
スプレードライ装置２８０の模式図を図５４に示す。スプレードライ装置２８０はチャンバー２８１と、ノズル２８２を有する。ノズル２８２には、チューブ２８３を介して懸濁液２８４が供給される。懸濁液２８４はノズル２８２からチャンバー２８１内へ噴霧状に供給され、チャンバー２８１内で乾燥される。ノズル２８２は、ヒーター２８５により加熱されてもよい。ここで、ヒーター２８５により、チャンバー２８１のうちノズル２８２に近い領域、例えば図５４に示す二点鎖線で囲む領域も加熱される。
【０５９９】
ここで懸濁液２８４として正極活物質と酸化グラフェンを含む懸濁液を用いた場合、酸化グラフェンで覆われた正極活物質の粉末がチャンバー２８１を介して回収容器２８６へ回収される。
【０６００】
ここで矢印２８８に示す経路により、チャンバー２８１内の雰囲気がアスピレーター等により吸引されてもよい。
【０６０１】
以下に、被膜形成条件の一例を示す。
【０６０２】
まず、酸化グラフェンを溶媒に分散させて懸濁液を作製した。
【０６０３】
純水は、酸化グラフェンの分散媒として高い分散性を有しているが、後に投入する活物質によっては正極活物質と反応し、Ｌｉが溶出する、または、正極活物質の表面構造を変化させるようなダメージを与える恐れがある。よってエタノールと純水の割合を４：６として酸化グラフェンを液中に分散させた。
【０６０４】
液中に分散させるための攪拌としては、スターラーおよび超音波発生器を用い、回転数は

７５０ｒｐｍとし、超音波を２分照射した。
【０６０５】
次いで、ＬｉＯＨ水溶液を滴下して、ｐＨ７（２５℃）に調整した。
【０６０６】
正極活物質（本実施例では日本化学工業株式会社製の、コバルト酸リチウム粒子（商品名：Ｃ―２０Ｆ））を投入し、攪拌として、スターラーおよび超音波発生器を用い、回転数７５０ｒｐｍ、超音波１分照射した。以上の工程によって懸濁液を用意した。上記の日本化学工業株式会社製コバルト酸リチウム粒子（商品名：Ｃ―２０Ｆ）は、少なくともフッ素、マグネシウム、カルシウム、ナトリウム、シリコン、硫黄、リンを含むコバルト酸リチウム粒子であり、粒径が約２０μｍである。
【０６０７】
次いで、スプレードライ装置を用いて、懸濁液をスプレーノズル（ノズル径２０μｍ）で均一に噴霧して粉末を得た。スプレードライ装置の温風温度においては、入口の温度１６０℃、出口の温度４０℃、N_2ガス流量１０Ｌ／ｍｉｎとした。
【０６０８】
結果として得られた粉体の断面ＴＥＭ写真を図５５に示す。また、ＳＥＭ写真を図５６に示す。また、比較例として、同じ条件の正極活物質を用い、自転公転ミキサーを用いたところ、被覆が不十分であった。その比較例のＳＥＭ写真を図５７に示す。
【０６０９】
図５７と比較して図５６のほうが、粉体の表面に対して被膜が均一になされていることがわかる。
【０６１０】
スプレードライ装置を用いて酸化グラフェンを被覆させた後、酸化グラフェンで覆われた活物質層２００に、さらに導電助剤としてグラフェン化合物を用いる場合の断面構成例を図５８に説明する。
【０６１１】
図５８（Ａ）に、活物質層２００の縦断面図を示す。活物質層２００は、酸化グラフェンで覆われた粒状の正極活物質１００と、導電助剤としてのグラフェン化合物２０１と、バインダ（図示せず）と、を含む。ここで、グラフェン化合物２０１として例えばグラフェンまたはマルチグラフェンを用いればよい。ここで、グラフェン化合物２０１はシート状の形状を有することが好ましい。また、グラフェン化合物２０１は、複数のマルチグラフェン、または（および）複数のグラフェンが部分的に重なりシート状となっていてもよい。
【０６１２】
活物質層２００の縦断面においては、図５８（Ｂ）に示すように、酸化グラフェンからなる被膜１０５で覆われた正極活物質１００とグラフェン化合物２０１が接触している様子を表している。複数のグラフェン化合物２０１は、被膜１０５で覆われた正極活物質１００と一部接し、さらに隣り合う正極活物質１００の被膜１０５に張り付くように形成されて、それぞれと接触している。
【０６１３】
グラフェン化合物２０１と被膜１０５は同じ炭素系の材料であるため、優れた導電パスを形成することができる。
【０６１４】
被膜１０５は、電解液が触れないように正極活物質１００の結晶構造を保護する効果と、優れた導電パスを形成する効果を有している。
【符号の説明】
【０６１５】
１１ａ　正極
１１ｂ　負極
１２ａ　リード

１２ｂ　　リード
１４　　セパレータ
１５ａ　　接合部
１５ｂ　　接合部
１７　　固定部材
２０　　ノズル径
５０　　二次電池
５１　　外装体
６１　　折り曲げ部
６２　　シール部
６３　　シール部
７１　　稜線
７２　　谷線
７３　　空間
１００　　正極活物質
１０１　　領域
１０１ｐ　　結晶面
１０２　　領域
１０２ｐ　　結晶面
１０３　　領域
１０３ｐ　　結晶面
１０４　　領域
１０５　　被膜
１０６　　クラック部
１１０　　粒子
１１１　　領域
１１２　　層
１１４　　酸化コバルト層
１２０　　粒子
１２１　　領域
１２２　　層
１２４　　酸化コバルト層
１２５　　層
２００　　活物質層
２０１　　グラフェン化合物
２１４　　セパレータ
２８０　　スプレードライ装置
２８１　　チャンバー
２８２　　ノズル
２８３　　チューブ
２８４　　懸濁液
２８５　　ヒーター
２８６　　回収容器
２８８　　矢印
３００　　二次電池
３０１　　正極缶
３０２　　負極缶
３０３　　ガスケット
３０４　　正極
３０５　　正極集電体

３０６　正極活物質層
３０７　負極
３０８　負極集電体
３０９　負極活物質層
３１０　セパレータ
５００　二次電池
５０１　正極集電体
５０２　正極活物質層
５０３　正極
５０４　負極集電体
５０５　負極活物質層
５０６　負極
５０７　セパレータ
５０８　電解液
５０９　外装体
５１０　正極リード電極
５１１　負極リード電極
６００　二次電池
６０１　正極キャップ
６０２　電池缶
６０３　正極端子
６０４　正極
６０５　セパレータ
６０６　負極
６０７　負極端子
６０８　絶縁板
６０９　絶縁板
６１１　ＰＴＣ素子
６１２　安全弁機構
６１３　導電板
６１４　導電板
６１５　モジュール
６１６　導線
６１７　温度制御装置
９００　回路基板
９１０　ラベル
９１１　端子
９１２　回路
９１３　二次電池
９１４　アンテナ
９１５　アンテナ
９１６　層
９１７　層
９１８　アンテナ
９２０　表示装置
９２１　センサ
９２２　端子
９３０　筐体
９３０ａ　筐体
９３０ｂ　筐体

９３１　負極
９３２　正極
９３３　セパレータ
９５０　捲回体
９５１　端子
９５２　端子
９８０　二次電池
９８１　フィルム
９８２　フィルム
９９３　捲回体
９９４　負極
９９５　正極
９９６　セパレータ
９９７　リード電極
９９８　リード電極
７１００　携帯表示装置
７１０１　筐体
７１０２　表示部
７１０３　操作ボタン
７１０４　二次電池
７２００　携帯情報端末
７２０１　筐体
７２０２　表示部
７２０３　バンド
７２０４　バックル
７２０５　操作ボタン
７２０６　入出力端子
７２０７　アイコン
７３００　表示装置
７３０４　表示部
７４００　携帯電話機
７４０１　筐体
７４０２　表示部
７４０３　操作ボタン
７４０４　外部接続ポート
７４０５　スピーカ
７４０６　マイク
７４０７　二次電池
７４０８　リード電極
７４０９　集電体
７５００　電子タバコ
７５０１　アトマイザ
７５０２　カートリッジ
７５０４　二次電池
８０００　表示装置
８００１　筐体
８００２　表示部
８００３　スピーカ部
８００４　二次電池
８０２１　充電装置

８０２２　ケーブル
８０２４　二次電池
８１００　照明装置
８１０１　筐体
８１０２　光源
８１０３　二次電池
８１０４　天井
８１０５　側壁
８１０６　床
８１０７　窓
８２００　室内機
８２０１　筐体
８２０２　送風口
８２０３　二次電池
８２０４　室外機
８３００　電気冷凍冷蔵庫
８３０１　筐体
８３０２　冷蔵室用扉
８３０３　冷凍室用扉
８３０４　二次電池
８４００　自動車
８４０１　ヘッドライト
８４０６　電気モーター
８５００　自動車
８６００　スクータ
８６０１　サイドミラー
８６０２　二次電池
８６０３　方向指示灯
８６０４　座席下収納
９６００　タブレット型端末
９６２５　スイッチ
９６２６　スイッチ
９６２７　電源スイッチ
９６２８　操作スイッチ
９６２９　留め具
９６３０　筐体
９６３０ａ　筐体
９６３０ｂ　筐体
９６３１　表示部
９６３１ａ　表示部
９６３１ｂ　表示部
９６３２ａ　領域
９６３２ｂ　領域
９６３３　太陽電池
９６３４　充放電制御回路
９６３５　蓄電体
９６３６　ＤＣＤＣコンバータ
９６３７　コンバータ
９６３８　操作キー
９６３９　ボタン

９６４０　　可動部

【書類名】特許請求の範囲
【請求項１】
正極活物質であって、
前記正極活物質は、第１の領域と、第２の領域と、第３の領域と、を有し、
前記第１の領域は前記正極活物質の内部に存在し、前記第２の領域および前記第３の領域は前記正極活物質の表層部に存在し、
前記第３の領域は、前記第２の領域よりも、前記正極活物質の表面に近い領域に存在し、
前記第１の領域はリチウムと第１の遷移金属の酸化物を有し、層状岩塩型の結晶構造を有し、
前記第２の領域は第２の遷移金属の酸化物を有する不定比化合物を有し、前記不定比化合物は岩塩型の結晶構造を有し、
前記第３の領域は典型元素の化合物を有し、前記典型元素の化合物は岩塩型の結晶構造を有する正極活物質。
【請求項２】
請求項１において、
前記第１の領域が有する前記層状岩塩型の結晶構造と、前記第２の領域が有する前記岩塩型の結晶構造の不整合度が０．１２以下、
前記第２の領域が有する前記岩塩型の結晶構造と、前記第３の領域が有する前記岩塩型の結晶構造の不整合度が０．１２以下である正極活物質。
【請求項３】
請求項２において、
前記第１の領域が有する前記層状岩塩型の結晶構造と、前記第２の領域が有する前記岩塩型の結晶構造の不整合度が０．０４以下、
前記第２の領域が有する前記岩塩型の結晶構造と、前記第３の領域が有する前記岩塩型の結晶構造の不整合度が０．０４以下である正極活物質。
【請求項４】
請求項１乃至請求項３のいずれか一において、
前記第１の領域が有する前記層状岩塩型の結晶構造の（１―１―４）面、または前記（１―１―４）面と直交する面と、前記第２の領域が有する前記岩塩型の結晶構造の｛１００｝面の不整合度が０．１２以下であり、
前記第２の領域が有する前記岩塩型の結晶構造の｛１００｝面と、前記第３の領域が有する前記岩塩型の結晶構造の｛１００｝面の不整合度が０．１２以下である正極活物質。
【請求項５】
請求項４において、
前記第１の領域が有する前記層状岩塩型の結晶構造の（１―１―４）面、または前記（１―１―４）面と直交する面と、前記第２の領域が有する前記岩塩型の結晶構造の｛１００｝面の不整合度が０．０４以下であり、
前記第２の領域が有する前記岩塩型の結晶構造の｛１００｝面と、前記第３の領域が有する前記岩塩型の結晶構造の｛１００｝面の不整合度が０．０４以下である正極活物質。
【請求項６】
請求項１乃至請求項５のいずれか一において、
前記第１の領域が有するリチウムと第１の遷移金属の酸化物は多結晶である正極活物質。
【請求項７】
請求項６において、
前記第１の領域が有するリチウムと第１の遷移金属の酸化物の、結晶子サイズの平均は２８０ｎｍ以上６３０ｎｍ以下である正極活物質。
【請求項８】
正極活物質であって、

前記正極活物質は、第１の領域と、第２の領域と、第３の領域と、を有し、
前記第１の領域は前記正極活物質の内部に存在し、前記第２の領域および前記第３の領域は前記正極活物質の表層部に存在し、
前記第３の領域は、前記第２の領域よりも、前記正極活物質の表面に近い領域に存在し、
前記第１の領域はコバルト酸リチウムを有し、
前記第２の領域はチタン酸リチウムを有し、
前記第３の領域は酸化マグネシウムを有する正極活物質。
【請求項９】
正極活物質であって、
前記正極活物質は、リチウムと、チタンと、コバルトと、マグネシウムと、酸素と、フッ素と、を有し、
前記正極活物質の表層部に存在し、Ｘ線光電子分光で測定されるコバルトの濃度を１としたとき、チタン濃度が０．０５以上０．４以下であり、マグネシウム濃度が０．４以上１．５以下であり、フッ素濃度が０．０５以上１．５以下である、正極活物質。
【請求項１０】
請求項１乃至請求項９のいずれか一に記載の前記正極活物質を有する正極と、負極と、を有する二次電池。
【請求項１１】
リチウム源、コバルト源、マグネシウム源およびフッ素源を混合する工程と、
前記リチウム源、前記コバルト源、前記マグネシウム源および前記フッ素源の混合物を、８００℃以上１１００℃以下で２時間以上２０時間以下加熱し、リチウムと、コバルトと、マグネシウムと、酸素と、フッ素と、を有する粒子を得る工程と、
第２の遷移金属のアルコキシドをアルコールに溶解する工程と、
第２の遷移金属のアルコキシドのアルコール溶液に、前記リチウムと、コバルトと、マグネシウムと、酸素と、フッ素と、を有する粒子を混合し、混合液を水蒸気を含む雰囲気中で撹拌する工程と、
混合液から沈殿物を回収する工程と、
回収された前記沈殿物を、酸素を有する雰囲気で５００℃以上１２００℃以下、保持時間５０時間以下で加熱する工程と、を有する、正極活物質の作製方法。

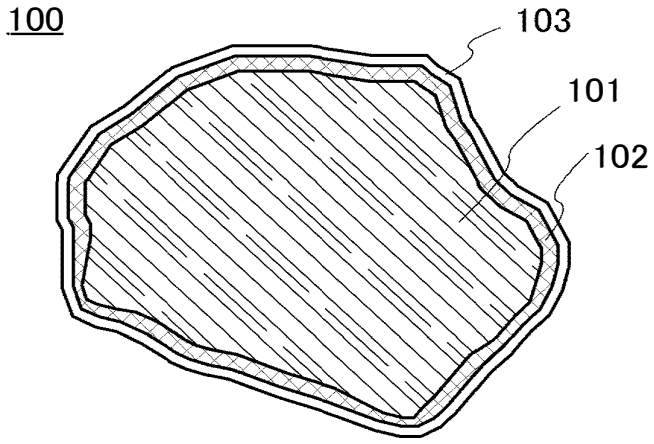
【請求項１２】
請求項１１に記載の正極活物質の作製方法において、
前記リチウム源が有するリチウムの原子数と、前記コバルト源が有する第１の遷移金属の原子数の比が、１．００≦Ｌｉ／第１の遷移金属＜１．０７である、正極活物質の作製方法。
【請求項１３】
請求項１１または請求項１２において、
前記フッ素源に含まれるフッ素と、前記マグネシウム源に含まれるマグネシウムの原子数比は、
Ｍｇ：Ｆ＝１：ｘ（１．５≦ｘ≦４）
である、正極活物質の作製方法。
【請求項１４】
請求項１１乃至請求項１３のいずれか一において、
前記マグネシウム源に含まれるマグネシウムは、
前記コバルト源に含まれる第１の遷移金属の０．５原子％以上１．５原子％以下である正極活物質の作製方法。
【請求項１５】
請求項１１乃至請求項１３のいずれか一において、
前記正極活物質と、グラフェン化合物と、を混合した後、

スプレードライ法により処理を行う、正極活物質の作製方法。
【請求項１６】
請求項１５において、前記グラフェン化合物は導電助剤である正極活物質の作製方法。

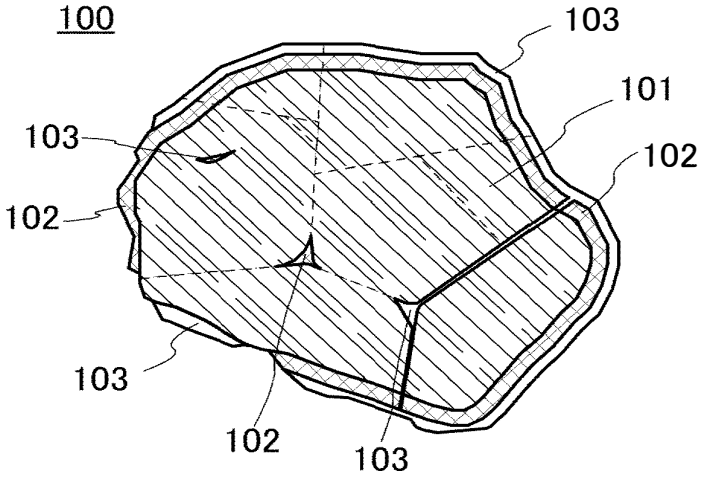
【書類名】要約書
【要約】
【課題】二次電池のサイクル特性を向上できる正極活物質を提供する。
【解決手段】層状岩塩型の結晶構造を有する、コバルト酸リチウム等の正極活物質の表層部に、２種の領域を設ける。内側の領域はチタン等の遷移金属を有する不定比化合物、外側の領域は酸化マグネシウム等の典型元素の化合物とする。２種の領域はどちらも岩塩型の結晶構造を有する。また内部の層状岩塩型の結晶構造と、表層部の２種の領域はトポタキシである。これらがトポタキシであるため、充放電によって生じる正極活物質の結晶構造の変化を効果的に抑制できる。また電解液と接触する外側の被覆層が、化学的に安定した典型元素の化合物であるため、サイクル特性の優れた二次電池とすることができる。
【選択図】図１

【書類名】図面
【図１】

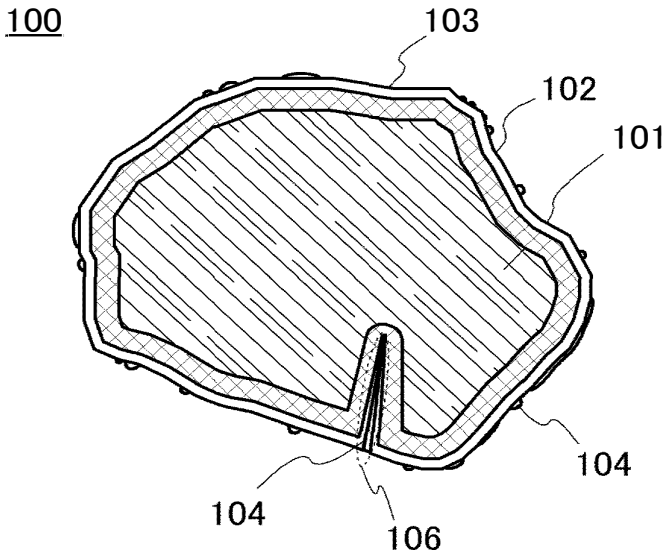
(A)


100
103
101
102

(B)

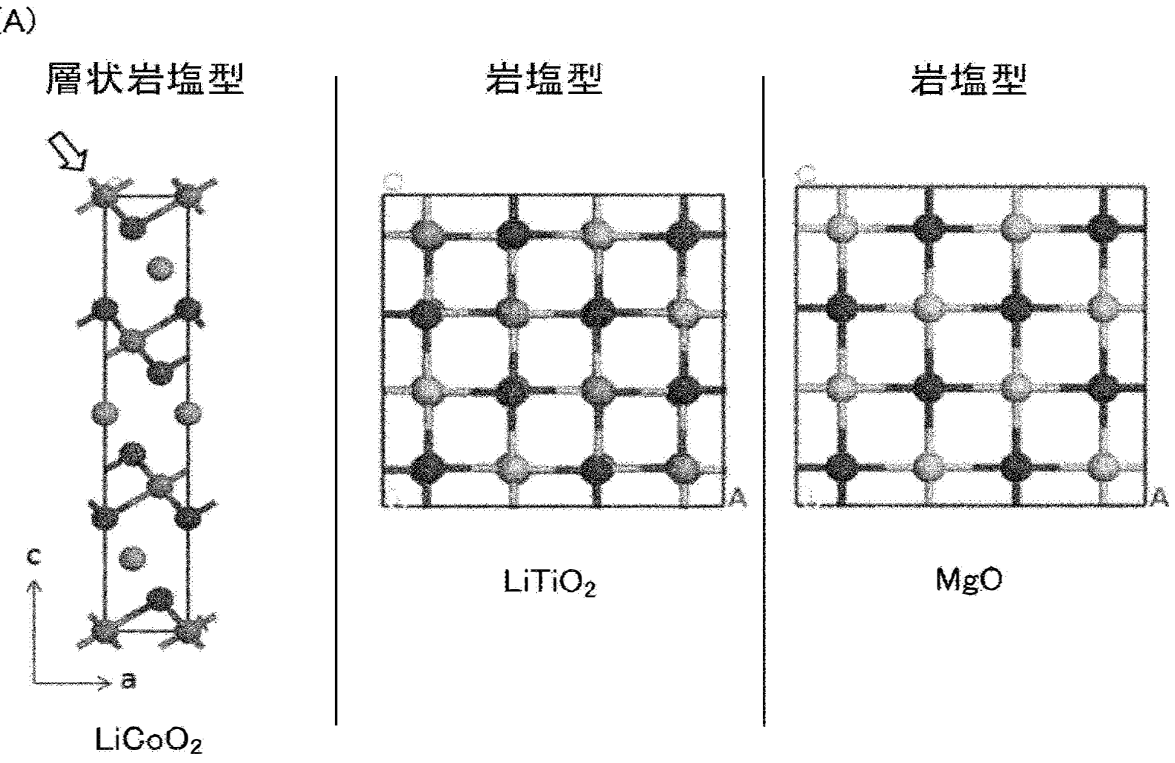

100
103
101
103
102
102
103
102
103

(C)

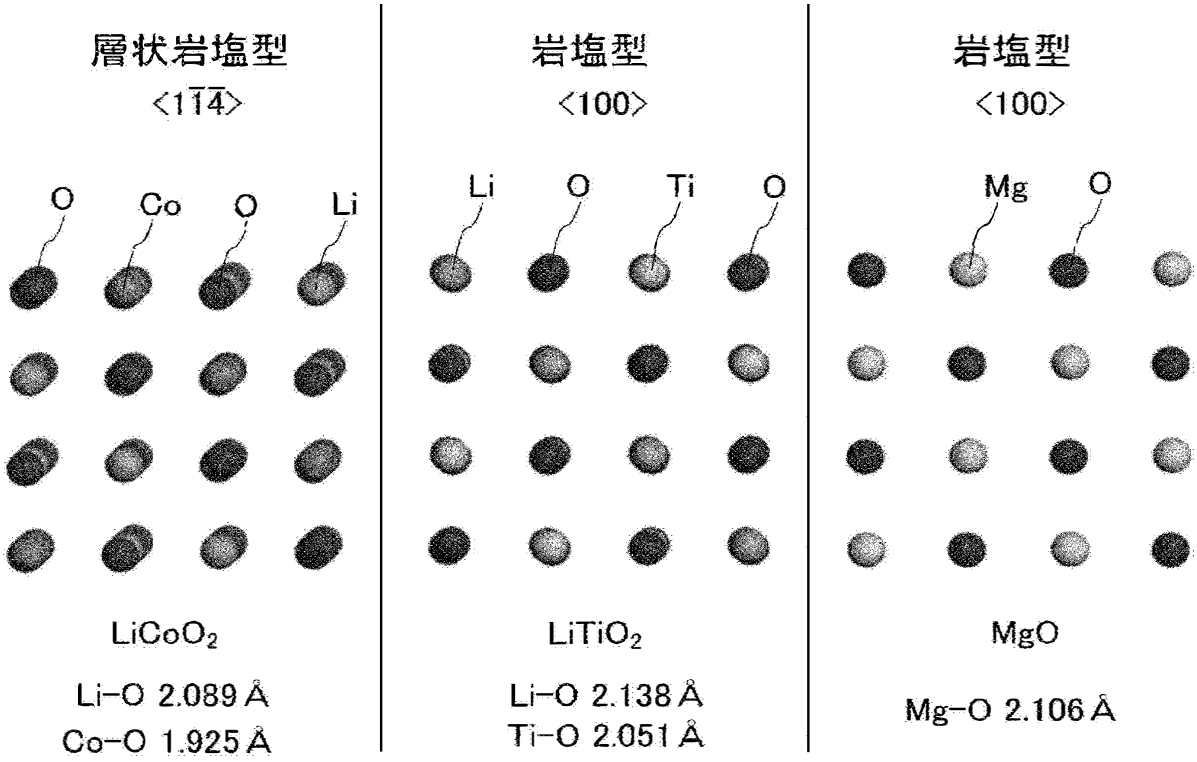

100
103
102
101
104
104
106

【図２】

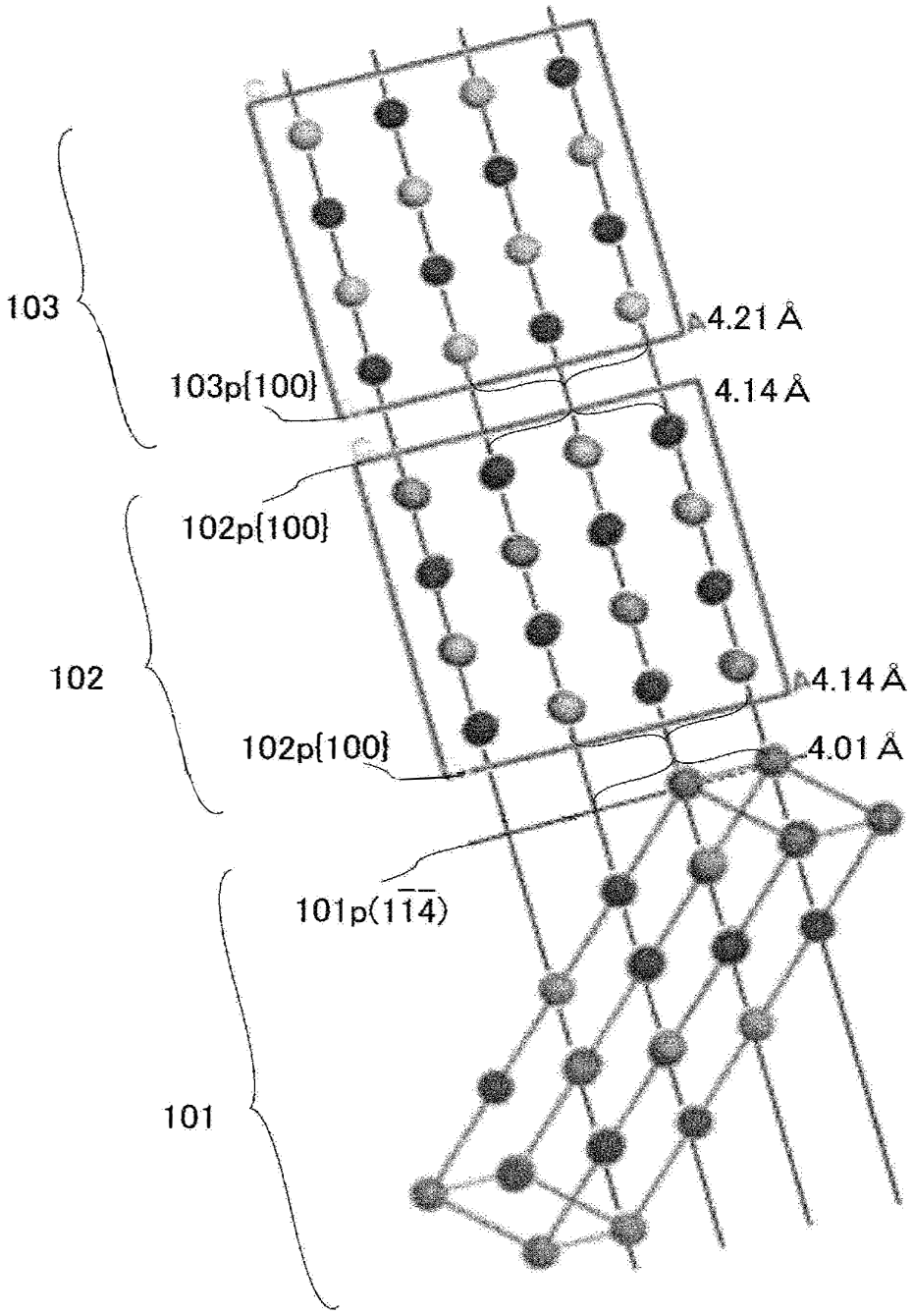
(A)


層状岩塩型
岩塩型
岩塩型
c
a
LiCoO2
LiTiO2
MgO

(B)

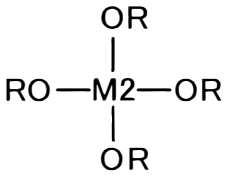

層状岩塩型
<1$\bar{1}\bar{4}$>
岩塩型
<100>
岩塩型
<100>
O
Co
O
Li
Li
O
Ti
O
Mg
O
LiCoO2
Li-O 2.089 Å
Co-O 1.925 Å
LiTiO2
Li-O 2.138 Å
Ti-O 2.051 Å
MgO
Mg-O 2.106 Å

【図３】


103
4.21 Å
103p{100}
4.14 Å
102p{100}
102
4.14 Å
102p{100}
4.01 Å
101p(1$\bar{1}\bar{4}$)
101

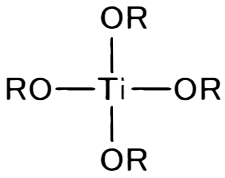
【図４】

(A-1)



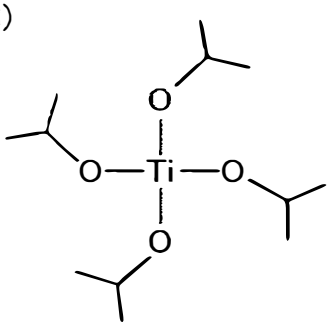
$M2(OR)_4$

(A-2)



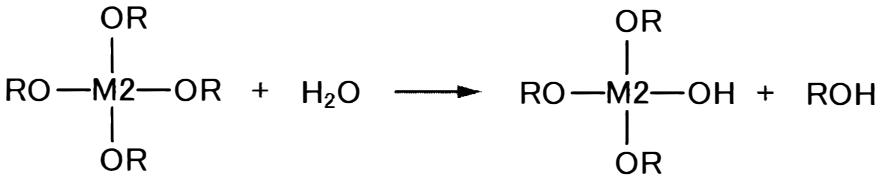
$Ti(OR)_4$

(A-3)

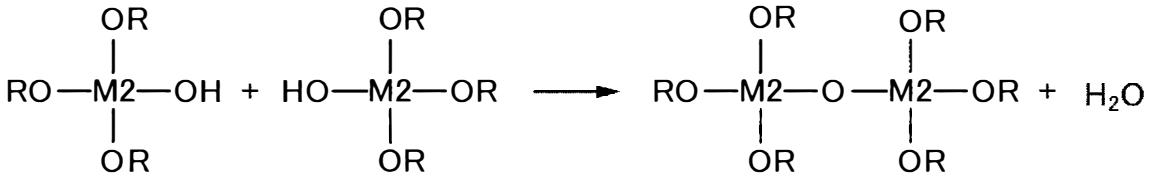


TTIP

(B)

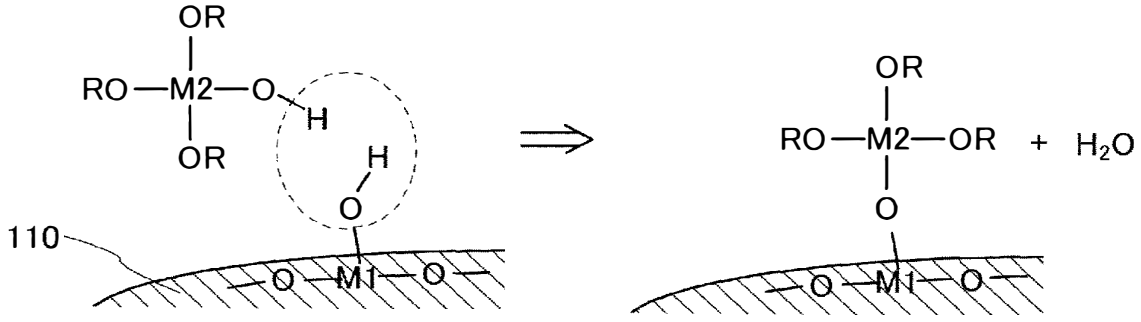


(C)

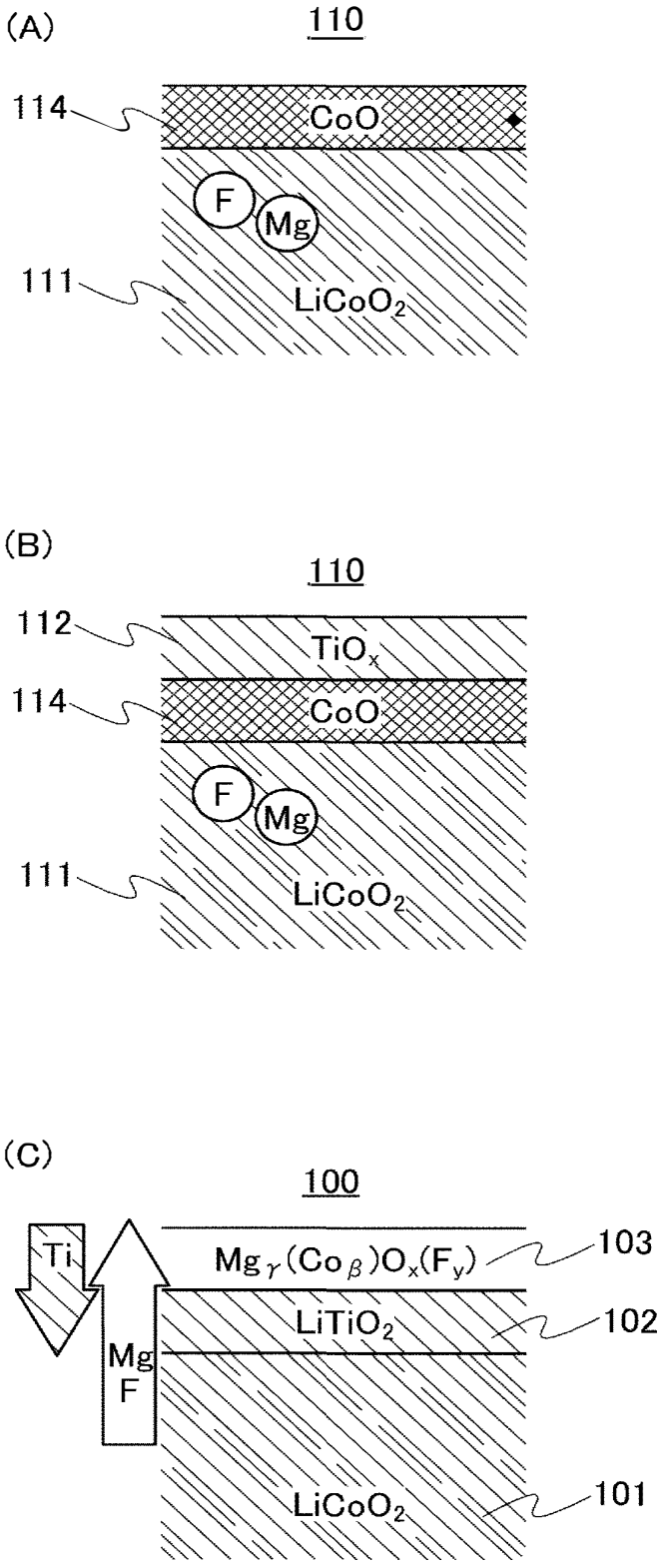


(D-1)

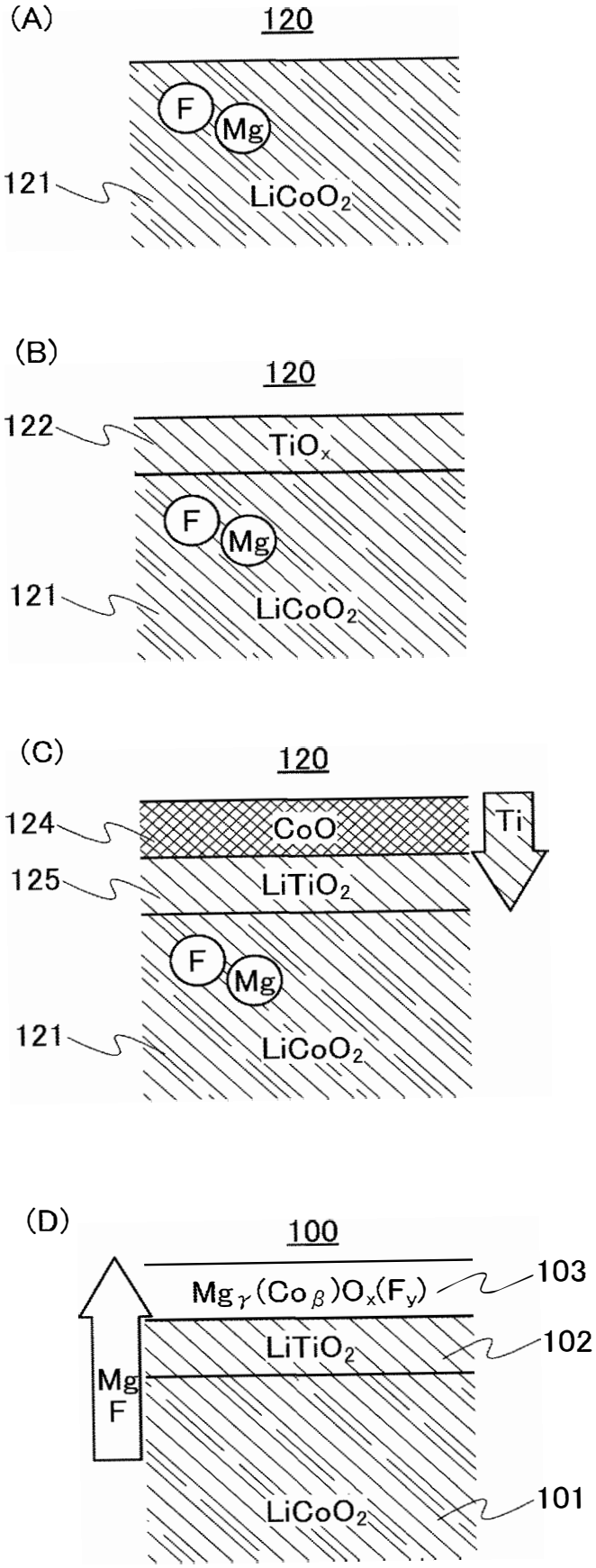
(D-2)



【図５】

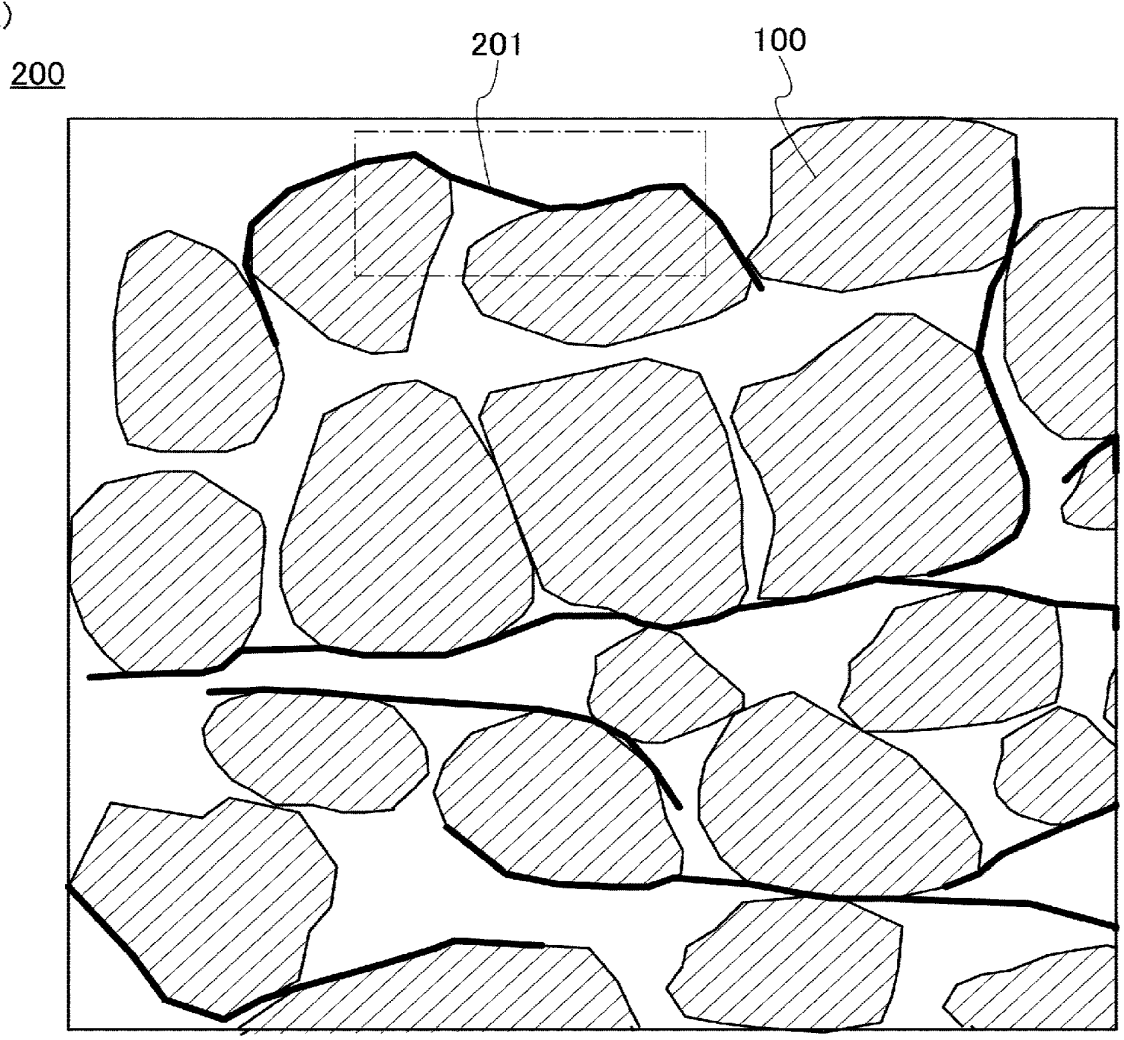

(A)
110
114
CoO
F
Mg
111
$LiCoO_2$
(B)
110
112
TiO_x
114
CoO
F
Mg
111
$LiCoO_2$
(C)
100
Ti
Mg
F
$Mg_\gamma(Co_\beta)O_x(F_y)$
103
$LiTiO_2$
102
$LiCoO_2$
101

【図６】

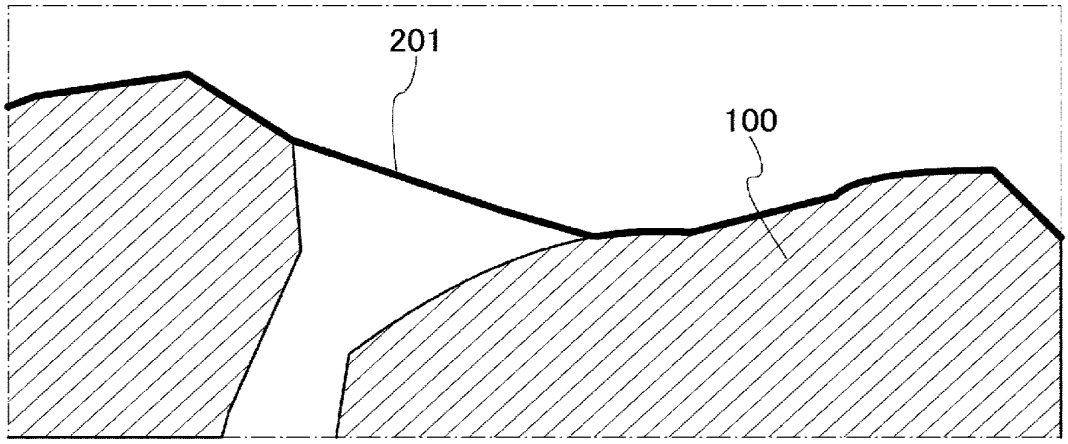

(A)
120
F
Mg
121
$LiCoO_2$
(B)
120
122
TiO_x
F
Mg
121
$LiCoO_2$
(C)
120
124
CoO
Ti
125
$LiTiO_2$
F
Mg
121
$LiCoO_2$
(D)
100
$Mg_\gamma(Co_\beta)O_x(F_y)$
103
$LiTiO_2$
102
Mg
F
$LiCoO_2$
101

【図７】

(A)

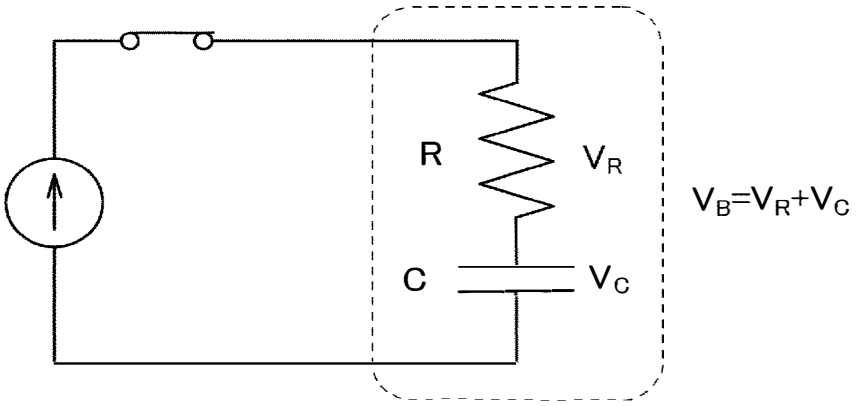


(B)

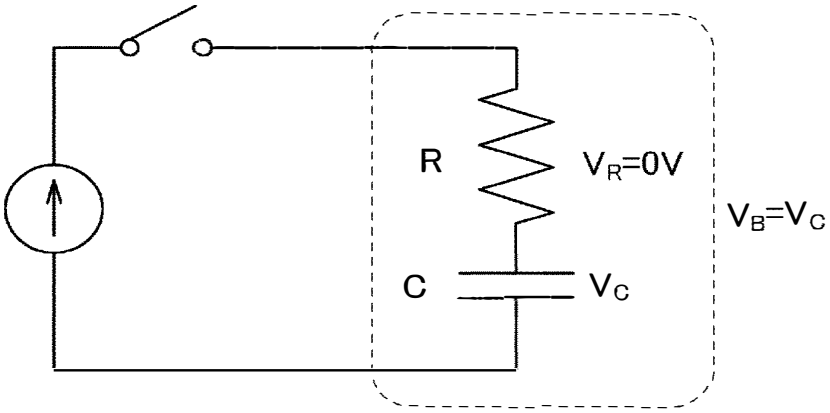


【図８】

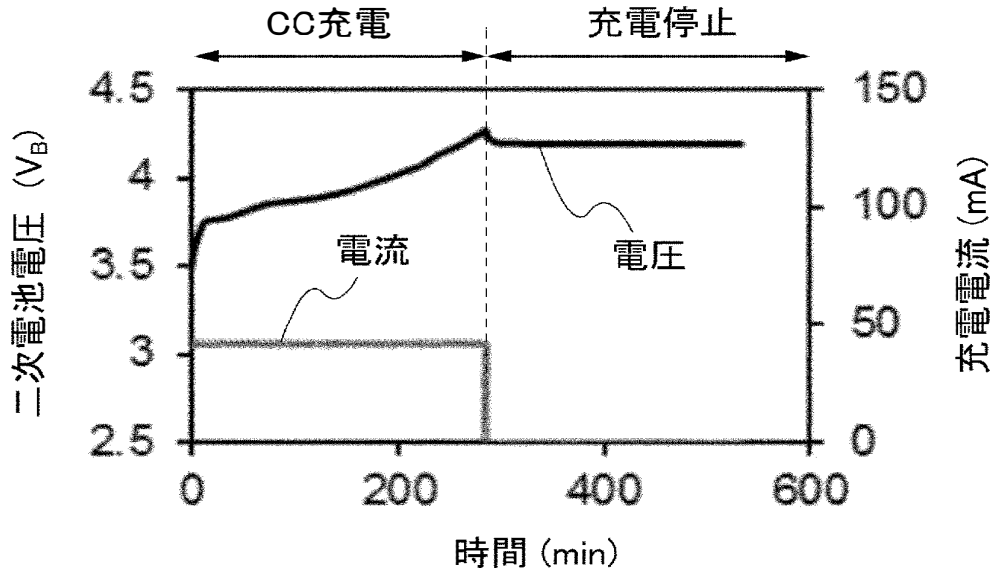
(A)


R
V_R
C
V_C
$V_B=V_R+V_C$

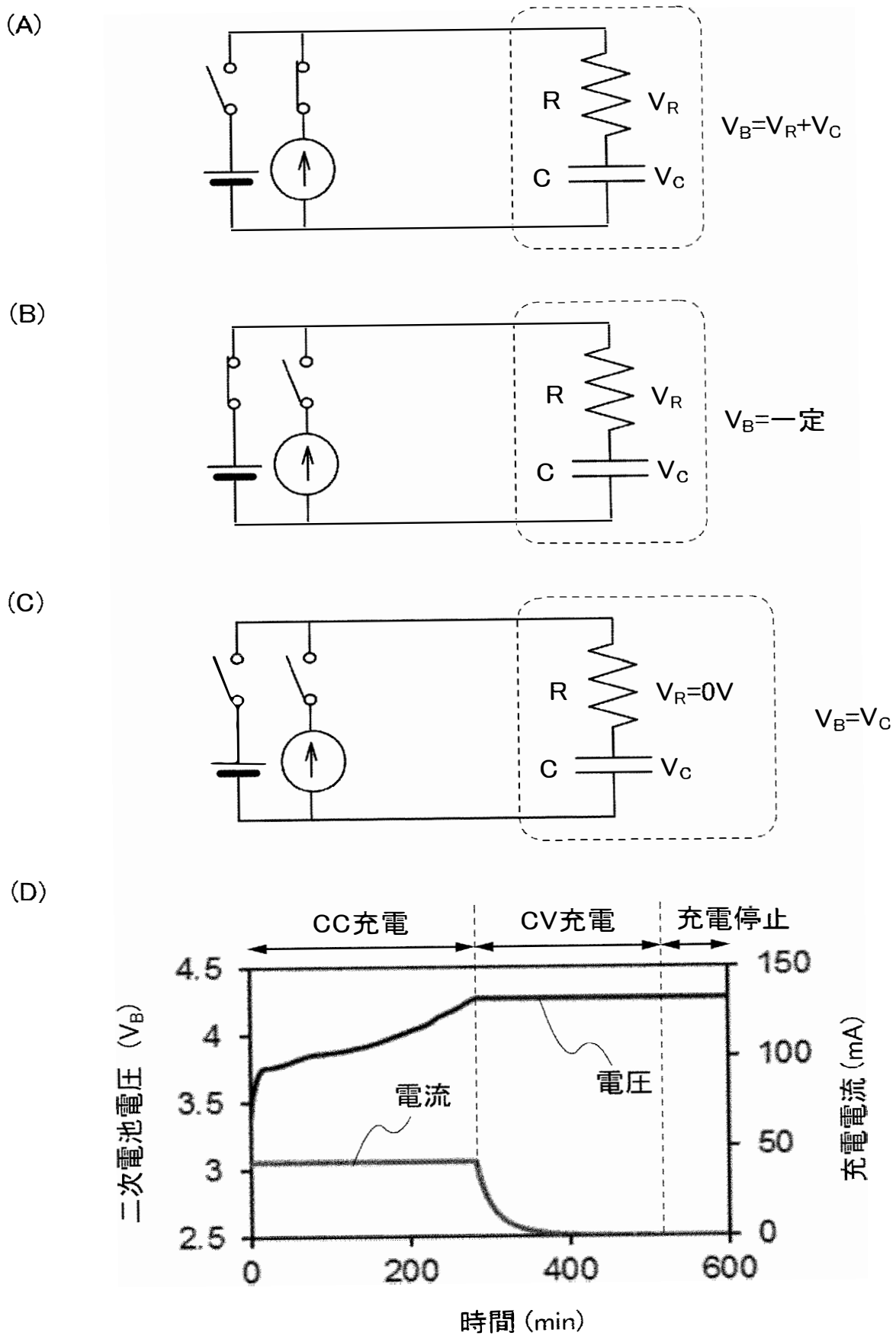
(B)


R
V_R=0V
C
V_C
$V_B=V_C$

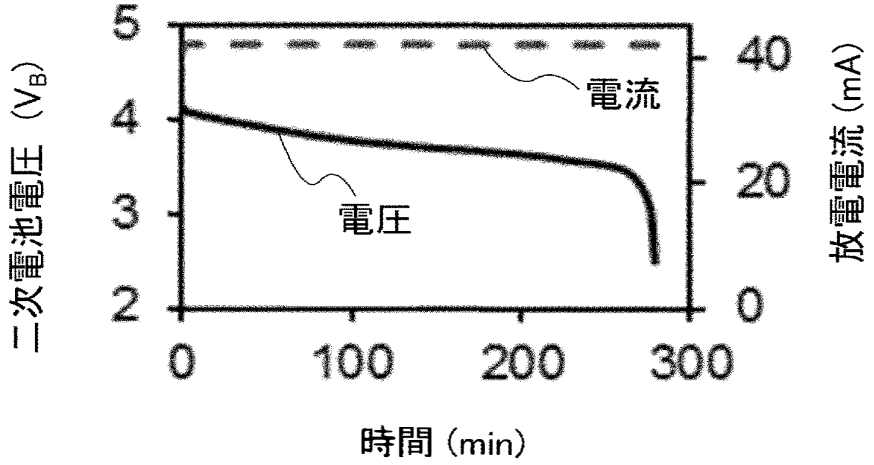
(C)


CC充電
充電停止
4.5
4
3.5
3
2.5
150
100
50
0
0
200
400
600
電流
電圧
二次電池電圧（V_B）
充電電流（mA）
時間（min）

【図９】

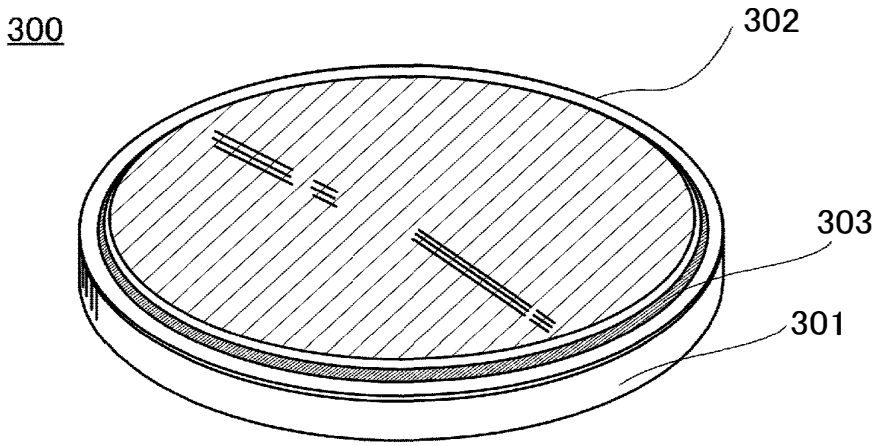

(A)
R
V_R
C
V_C
$V_B=V_R+V_C$
(B)
R
V_R
C
V_C
V_B=一定
(C)
R
V_R=0V
C
V_C
$V_B=V_C$
(D)
CC充電
CV充電
充電停止
4.5
4
3.5
3
2.5
150
100
50
0
0
200
400
600
電流
電圧
二次電池電圧（V_B）
充電電流（mA）
時間（min）

【図１０】

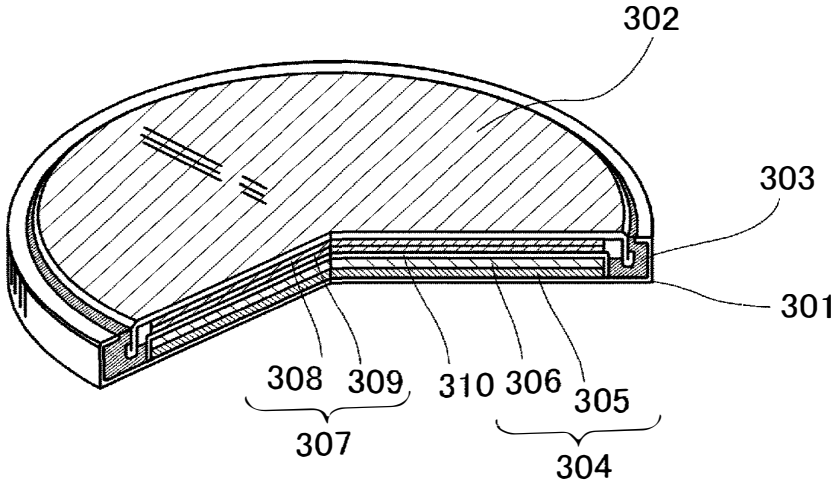


【図１１】

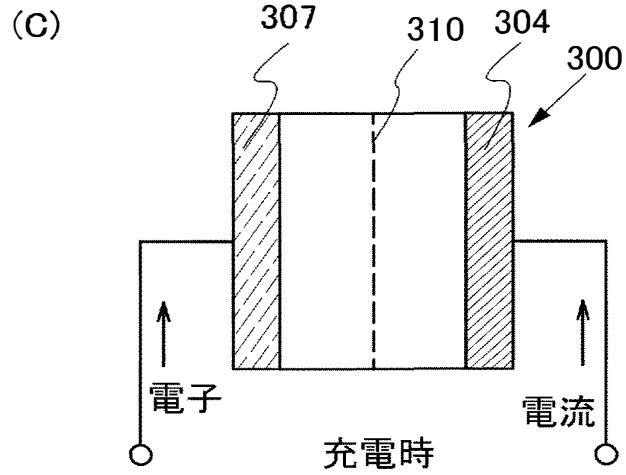
(A)


300
302
303
301

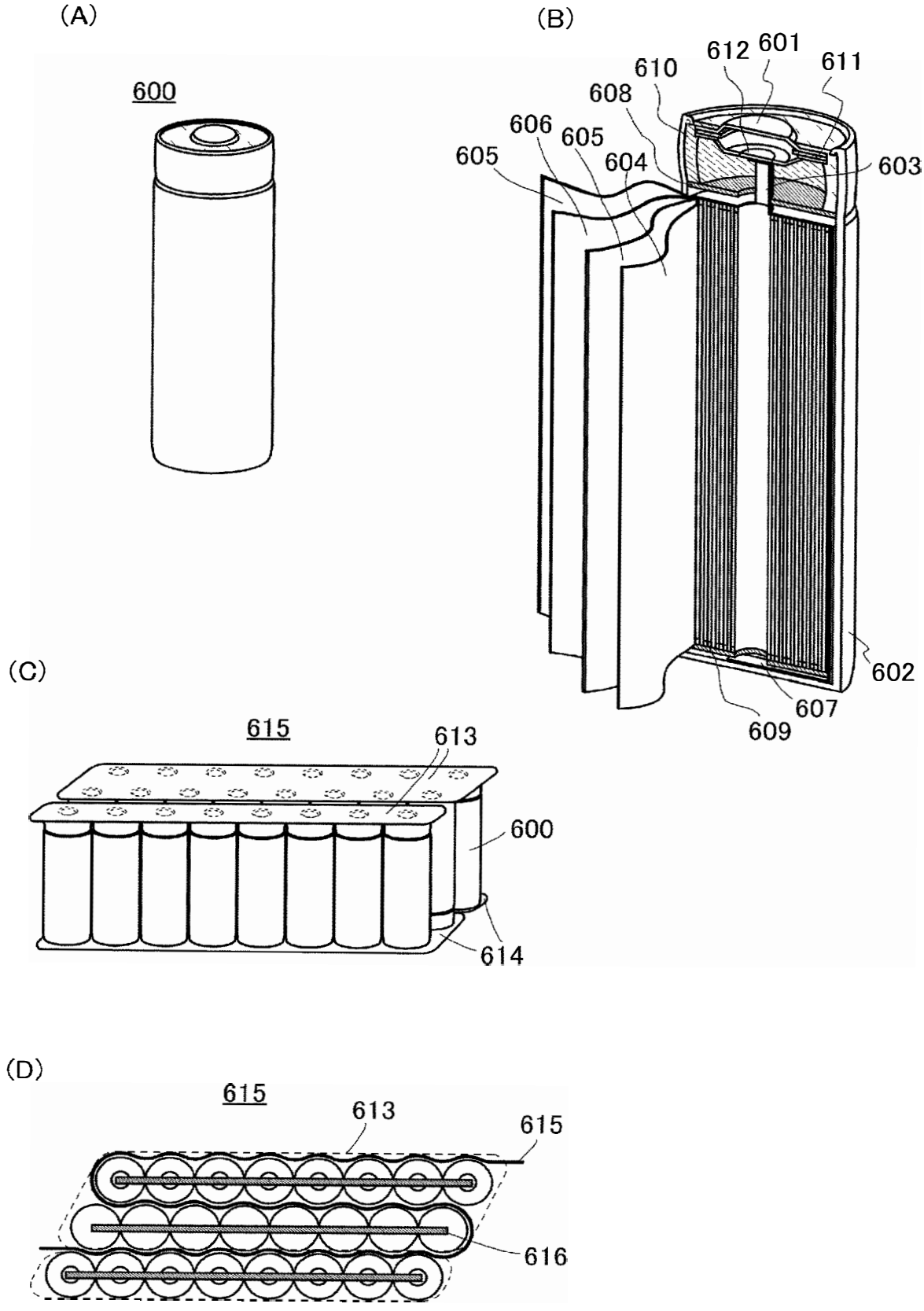
(B)


302
303
301
308
309
310
306
305
307
304

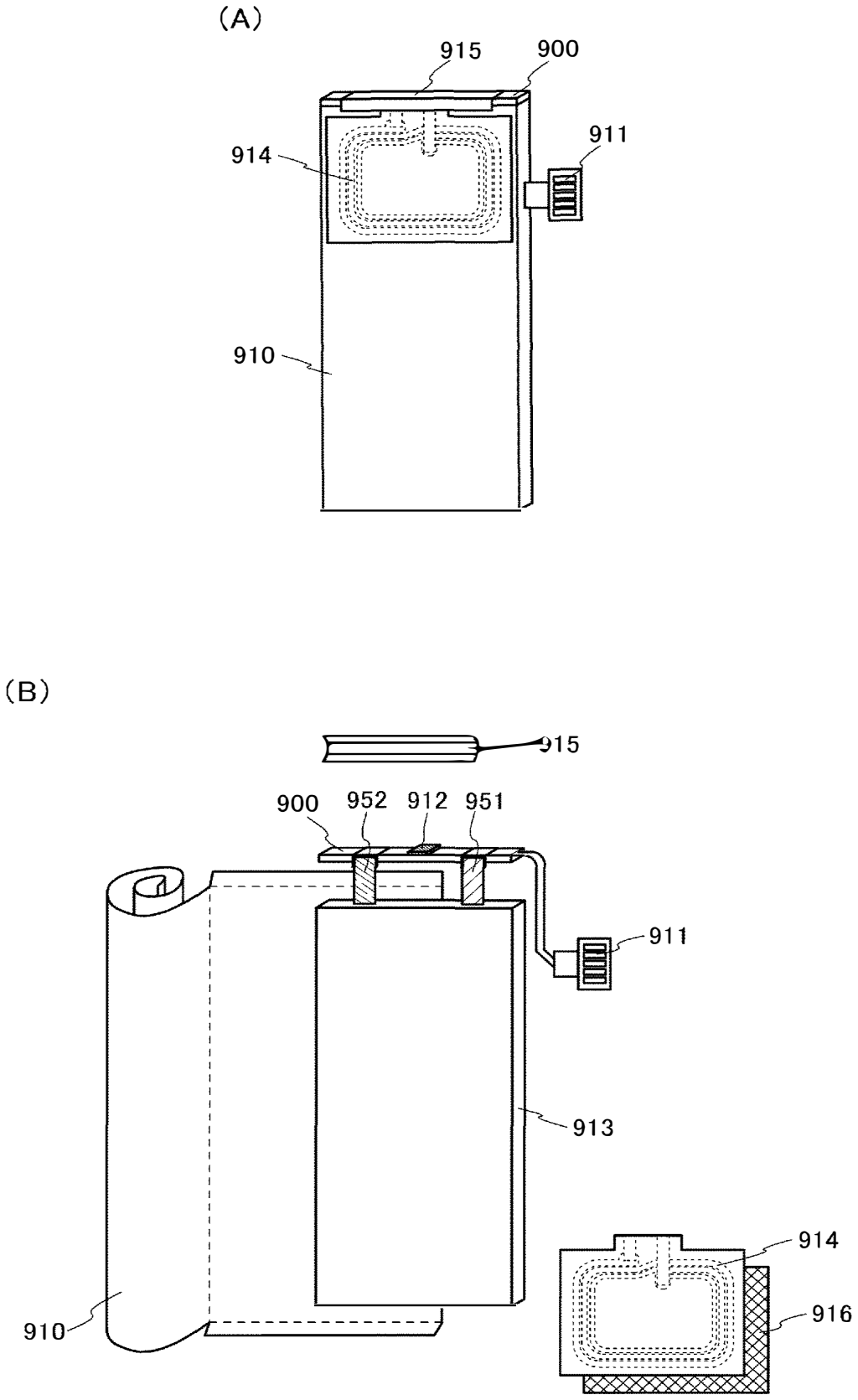
(C)


307
310
304
300
電子
電流
充電時

【図１２】

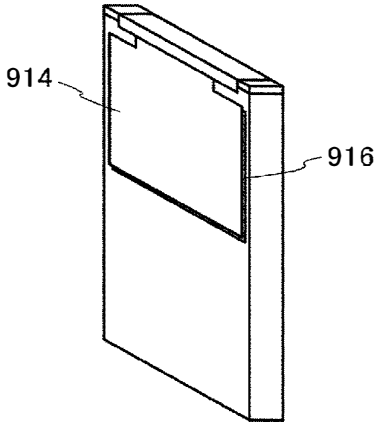

(A)
600
(B)
612
601
611
610
608
606
605
605
604
603
602
607
609
(C)
615
613
600
614
(D)
615
613
615
616

【図１３】

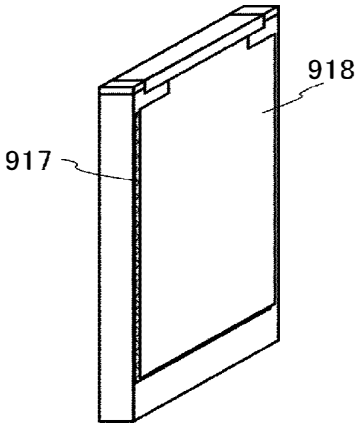


【図１４】

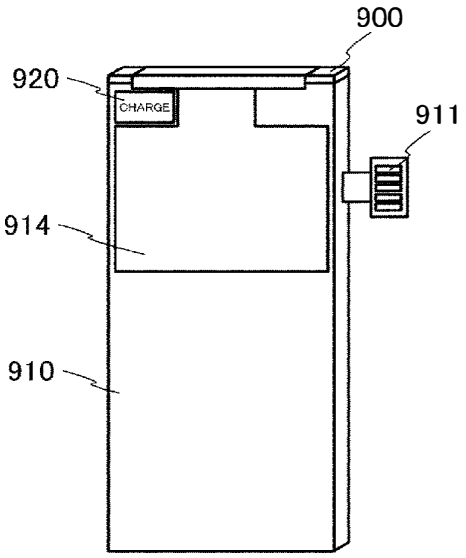
（A-1）



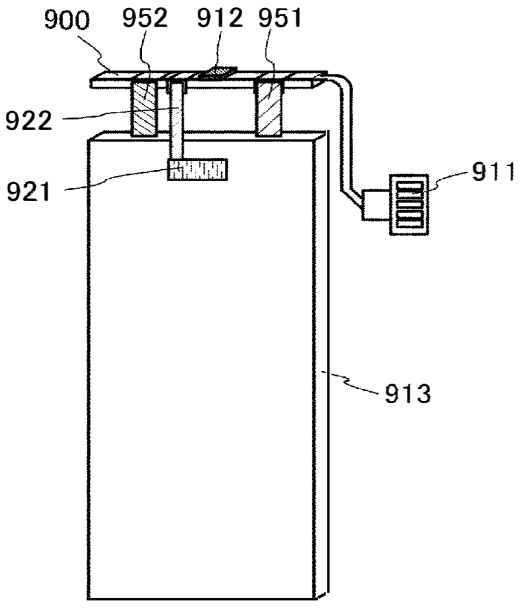
（A-2）



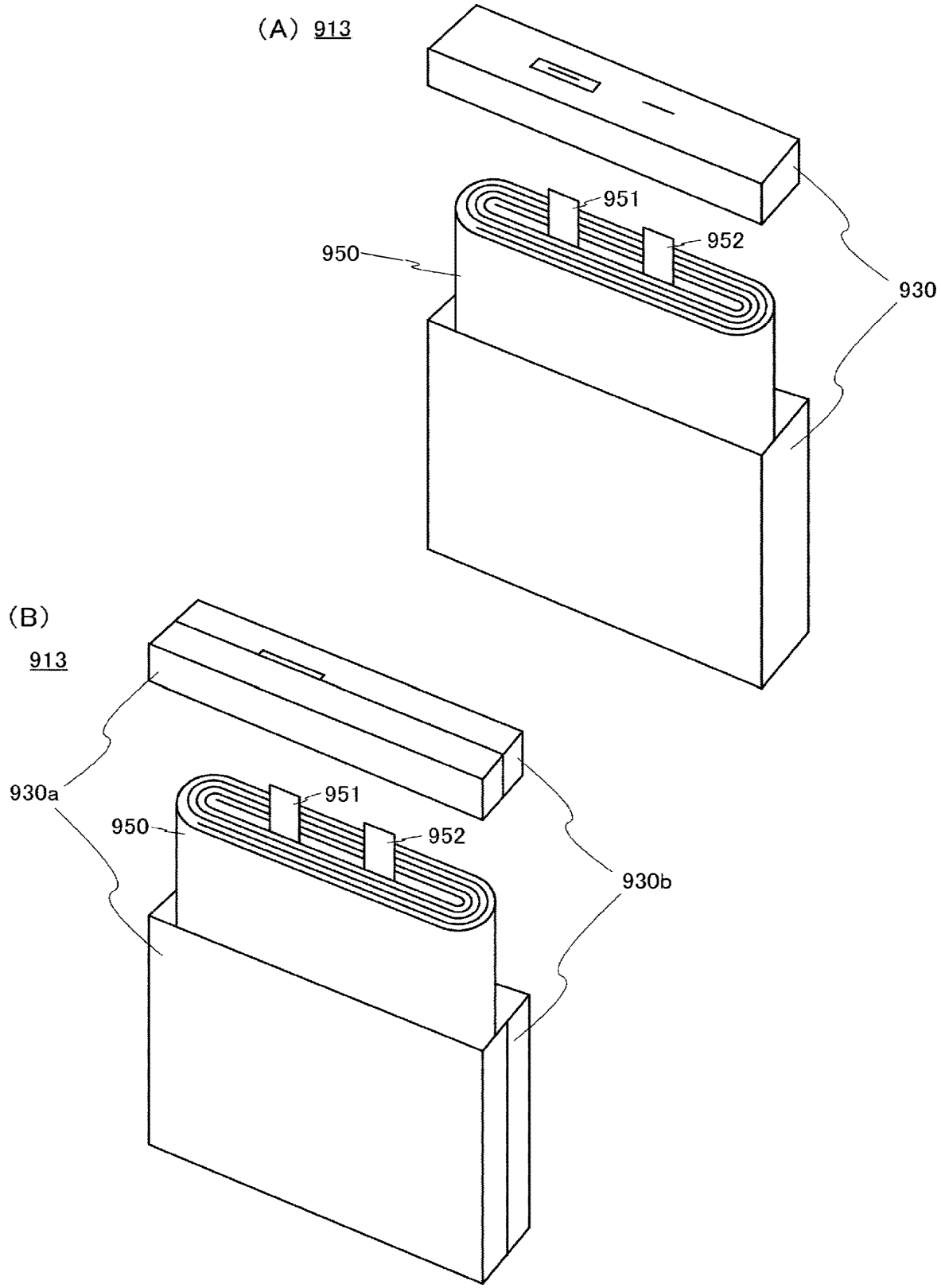
（B-1）



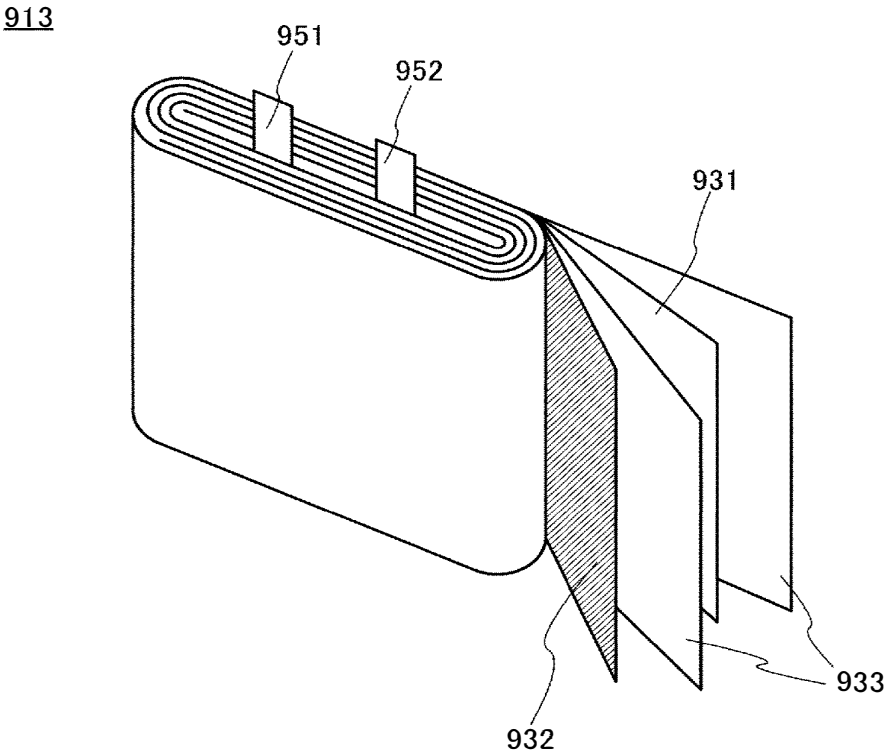
（B-2）



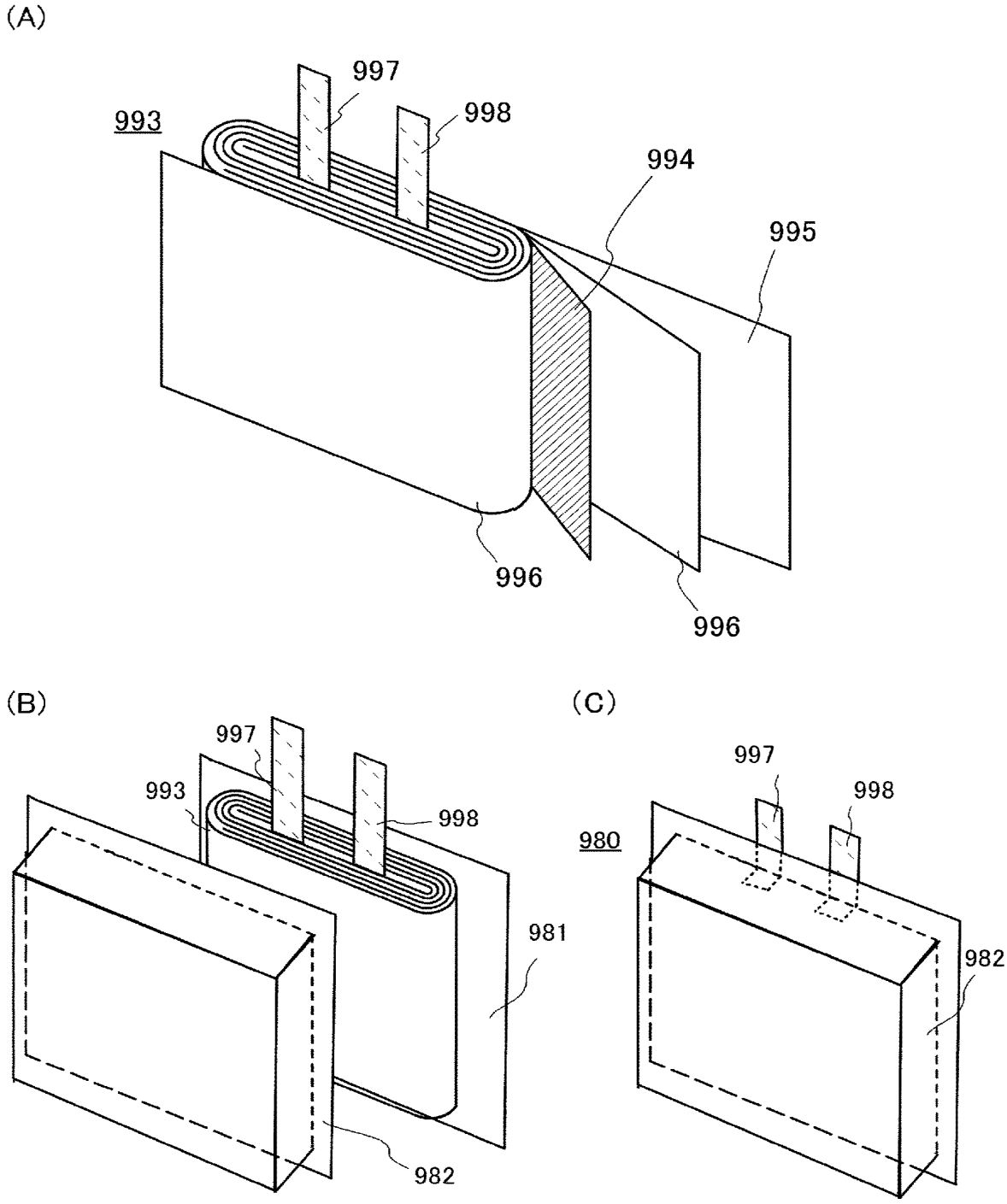
【図１５】



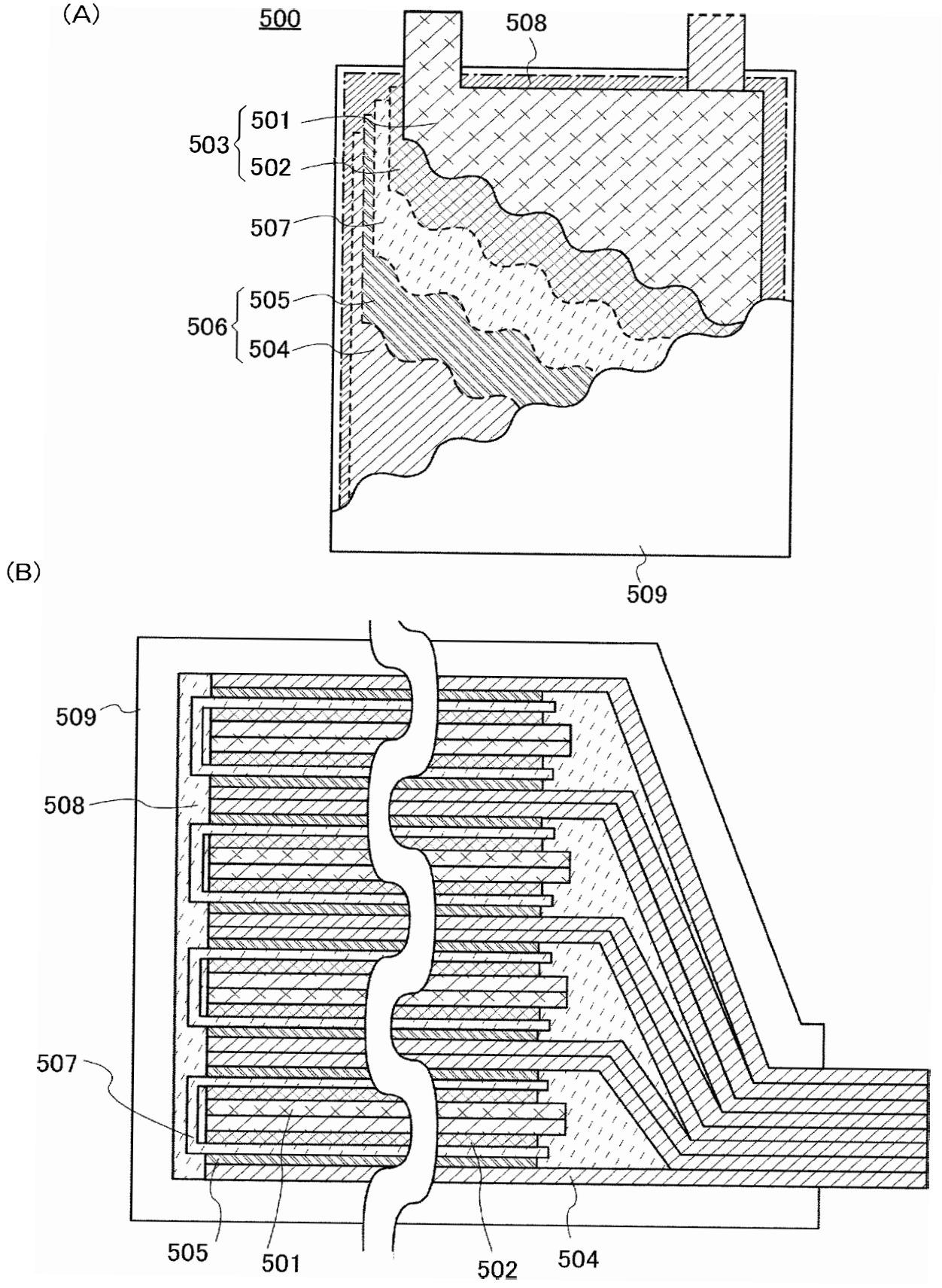
【図１６】



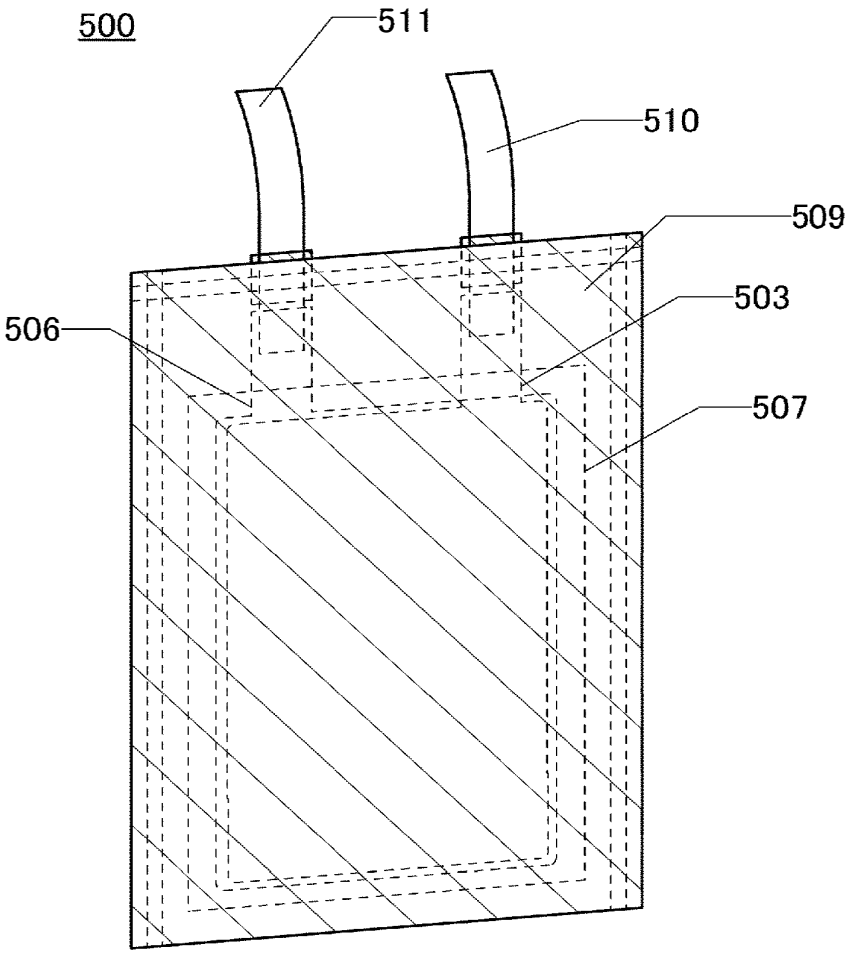
【図１７】


(A)
997
998
993
994
995
996
996
(B)
997
993
998
981
982
(C)
997
998
980
982

【図１８】

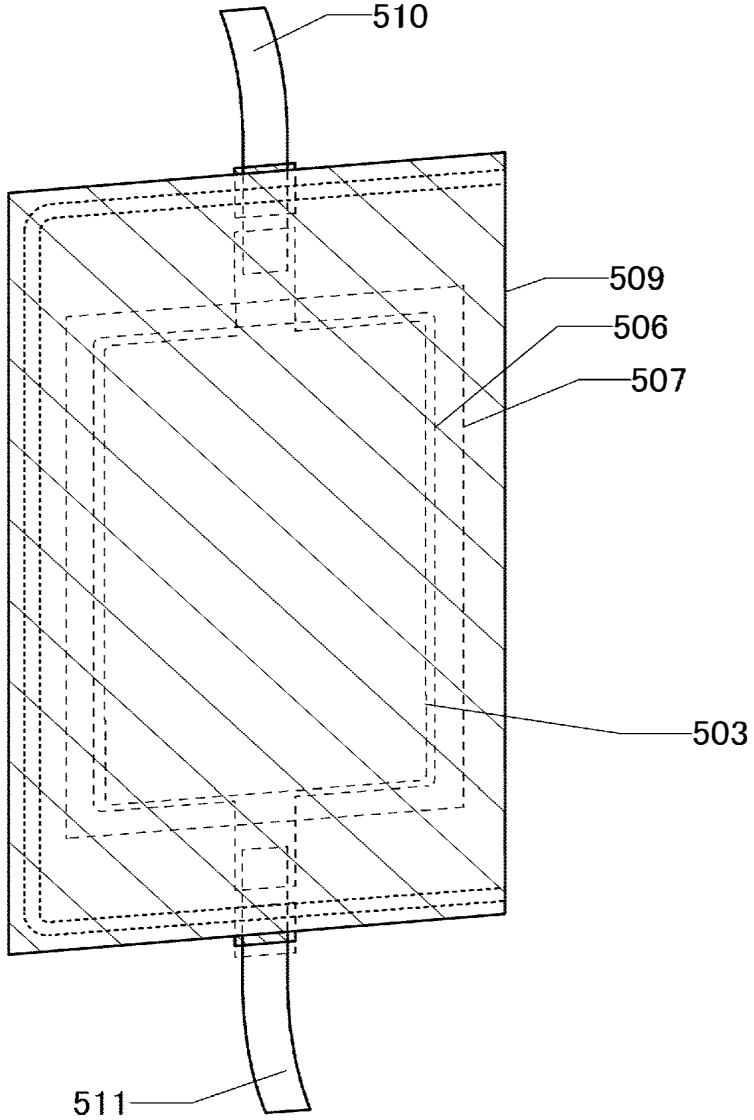

(A)
500
508
503
501
502
507
506
505
504
509
(B)
509
508
507
505
501
502
504

【図１９】

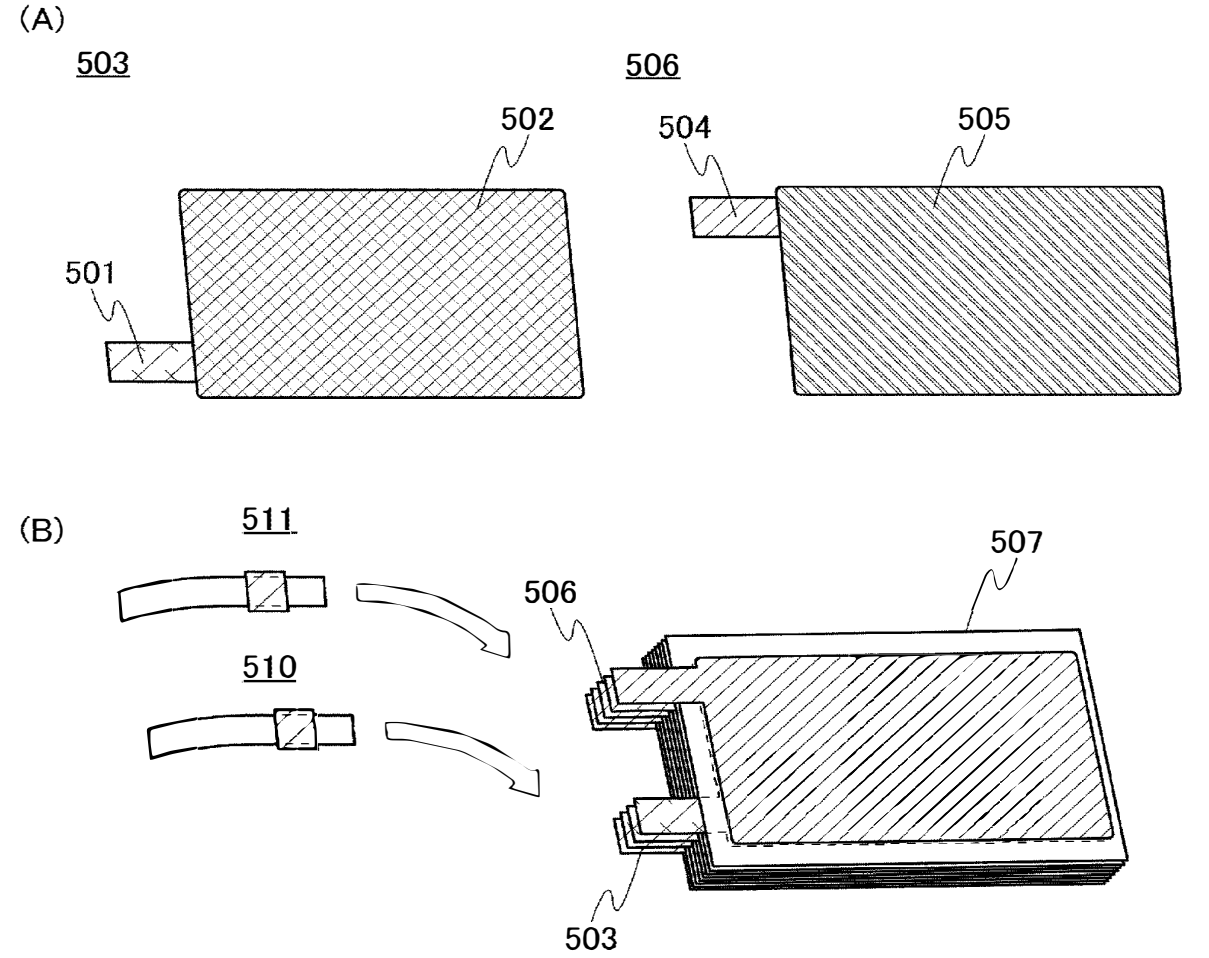


【図２０】

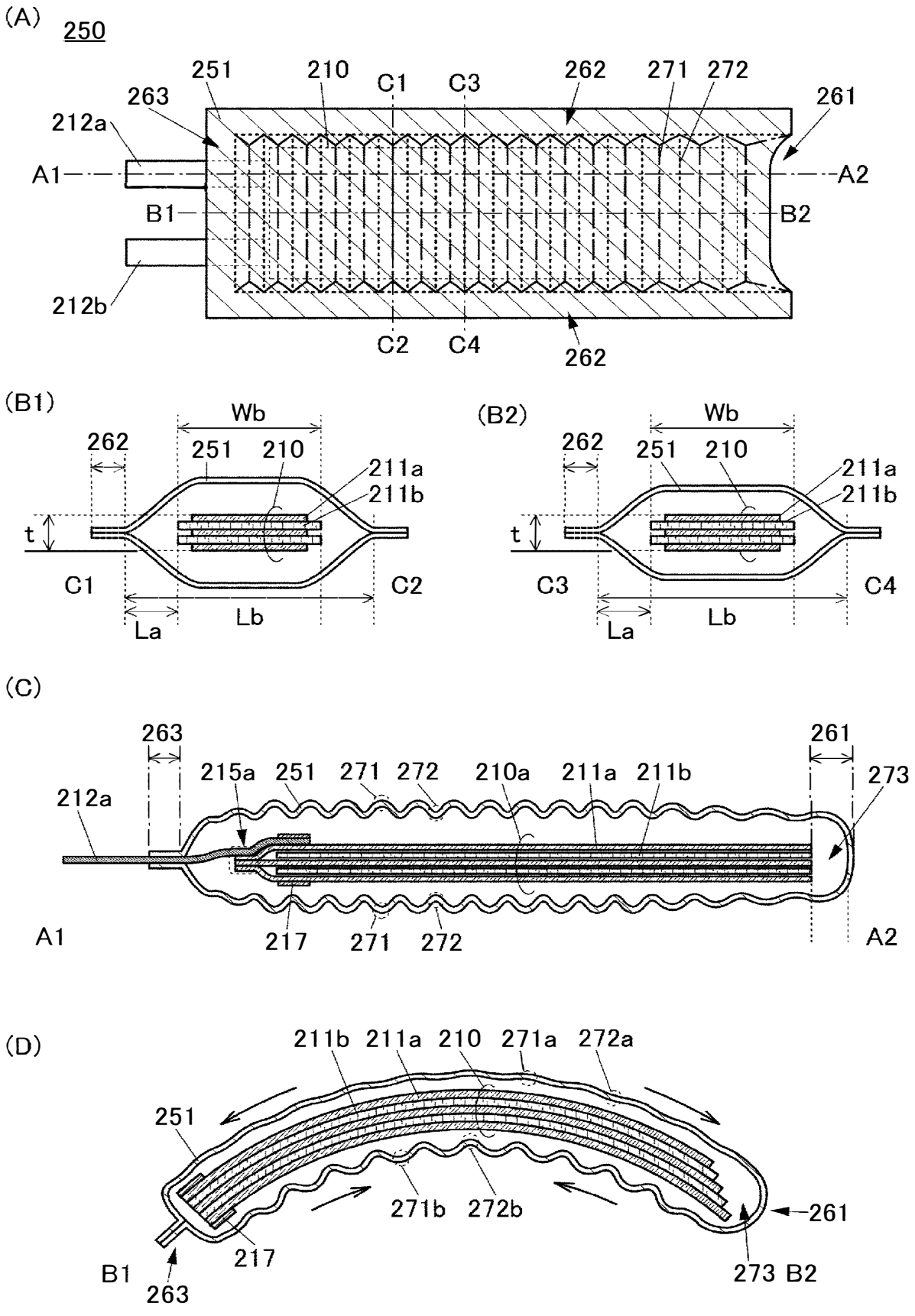
500



【図２１】

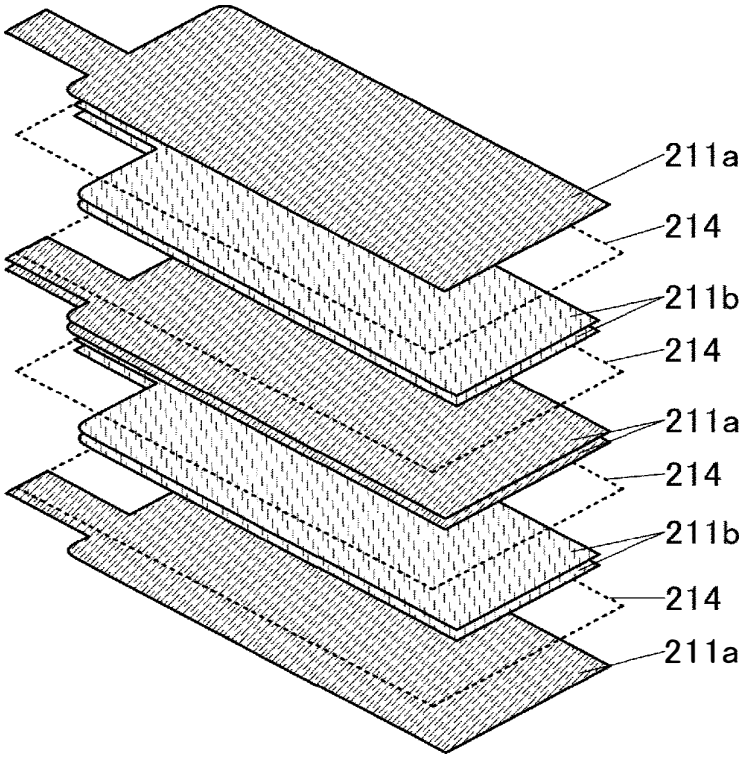


【図２２】

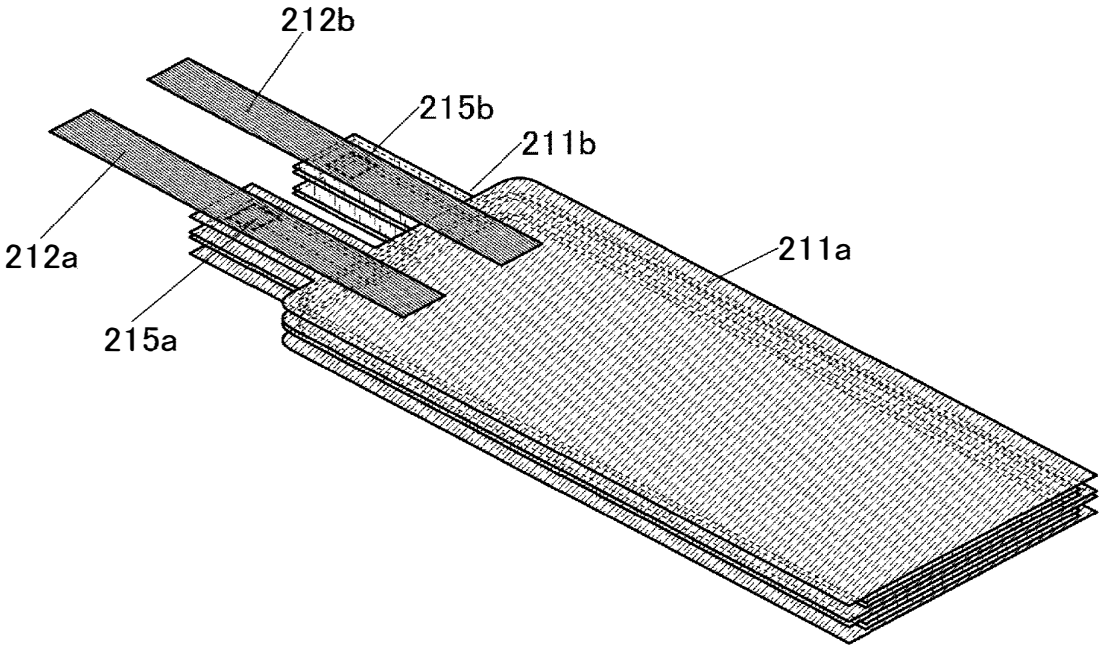

(A)
250
251
210
C1
C3
262
271
272
263
261
212a
A1
A2
B1
B2
212b
C2
C4
262
(B1)
Wb
262
251
210
211a
211b
t
C1
C2
La
Lb
(B2)
Wb
262
251
210
211a
211b
t
C3
C4
La
Lb
(C)
263
261
215a
251
271
272
210a
211a
211b
273
212a
A1
217
271
272
A2
(D)
211b
211a
210
271a
272a
251
271b
272b
261
217
B1
263
273
B2

【図２３】

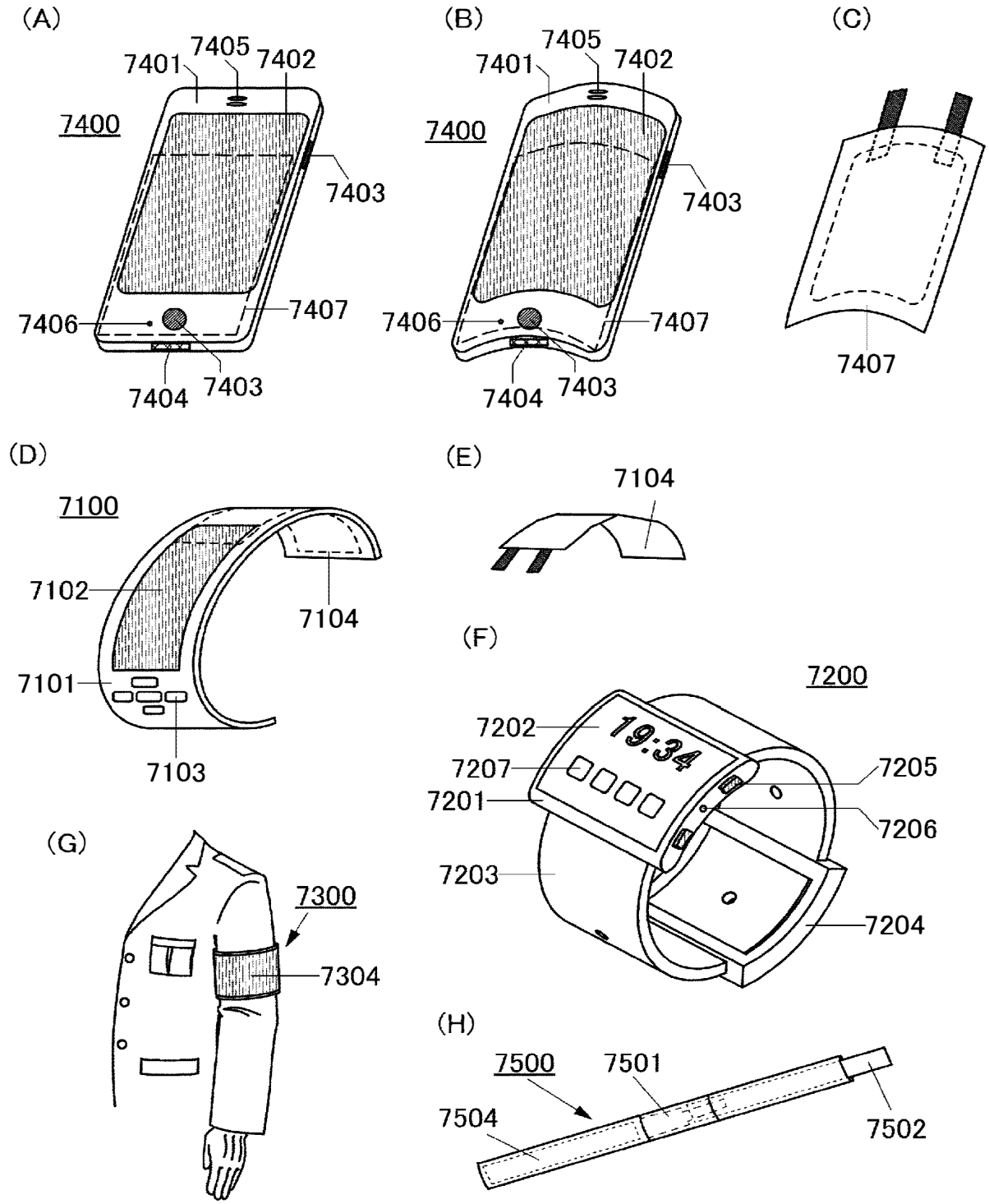
(A)


211a
214
211b
214
211a
214
211b
214
211a

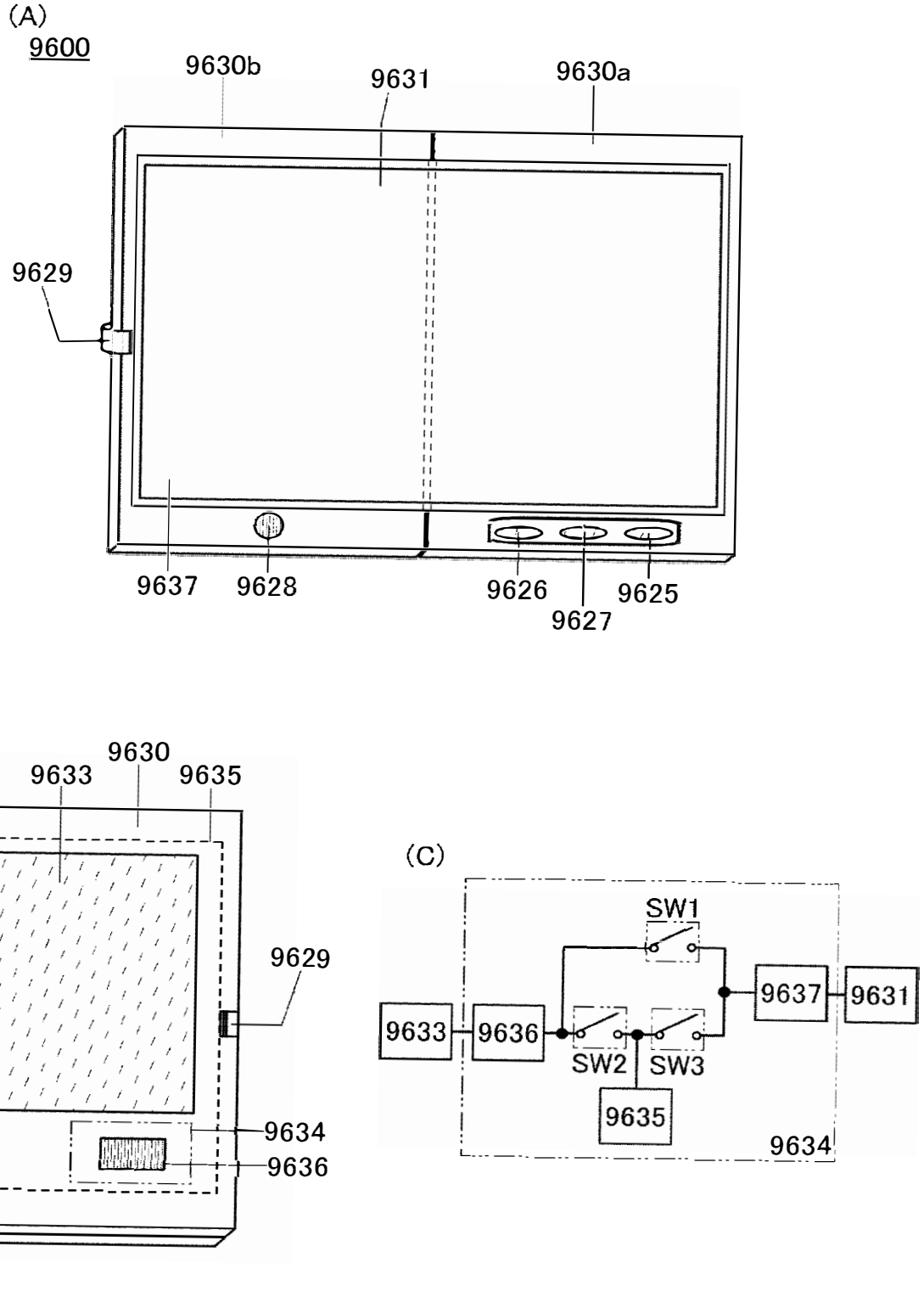
(B)


212b
215b
211b
212a
211a
215a

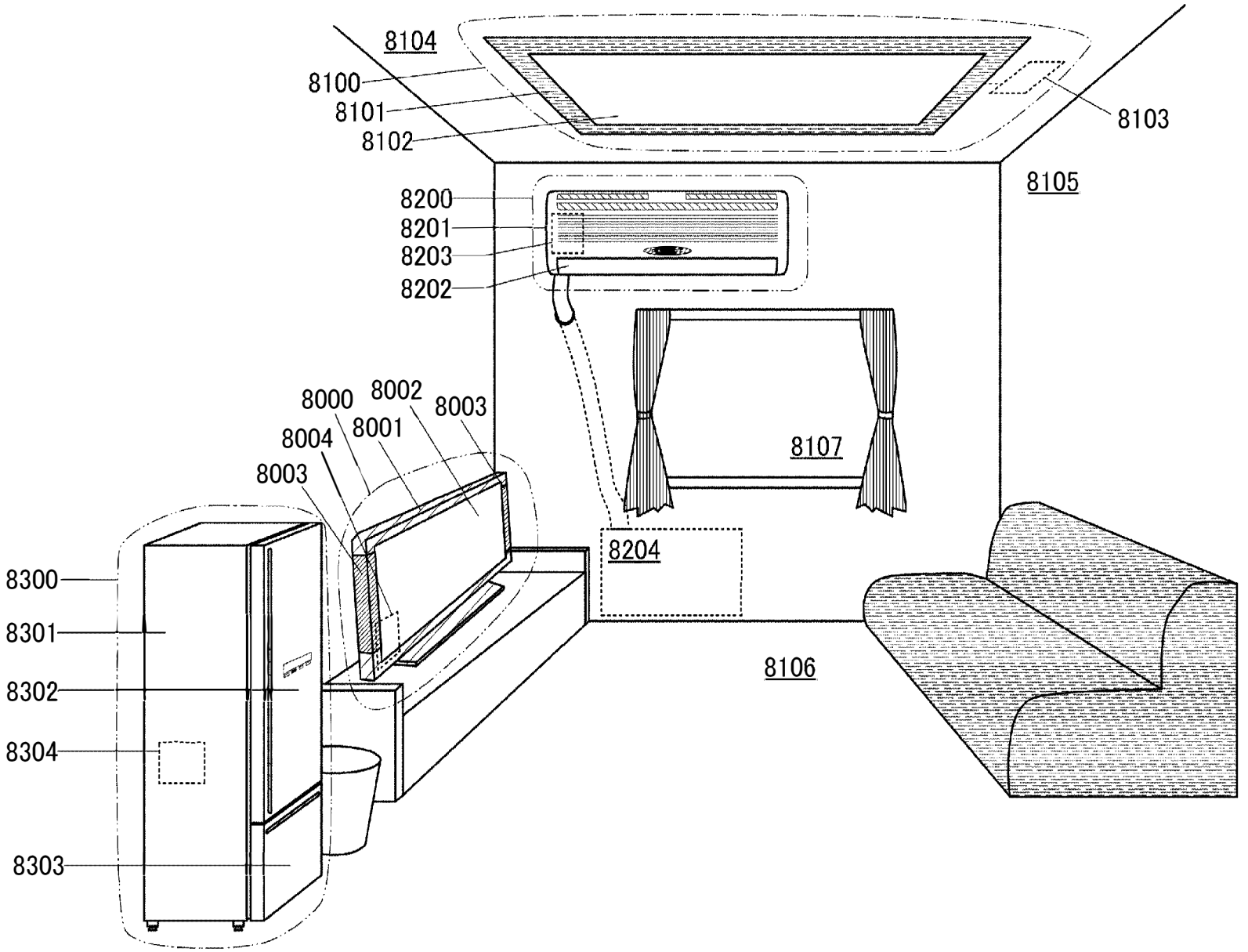
【図２４】


(A)
7401
7405
7402
7400
7403
7406
7407
7404
7403
(B)
7401
7405
7402
7400
7403
7406
7407
7404
7403
(C)
7407
(D)
7100
7102
7104
7101
7103
(E)
7104
(F)
7200
7202
19:34
7207
7205
7201
7206
7203
7204
(G)
7300
7304
(H)
7500
7501
7504
7502

【図２５】

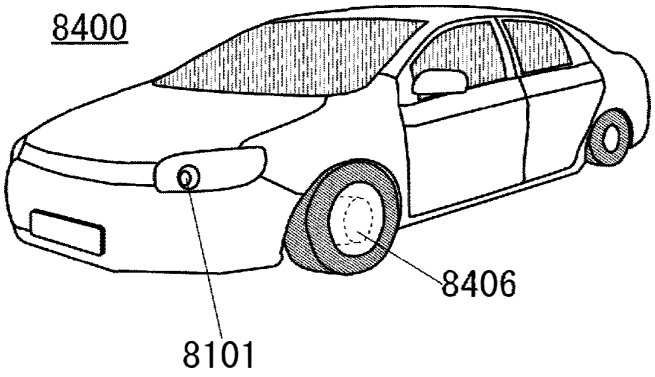

(A)
9600
9630b
9631
9630a
9629
9637
9628
9626
9627
9625
(B)
9600
9633
9630
9635
9629
9634
9636
9640
(C)
SW1
9633
9636
SW2
SW3
9637
9631
9635
9634

【図２６】

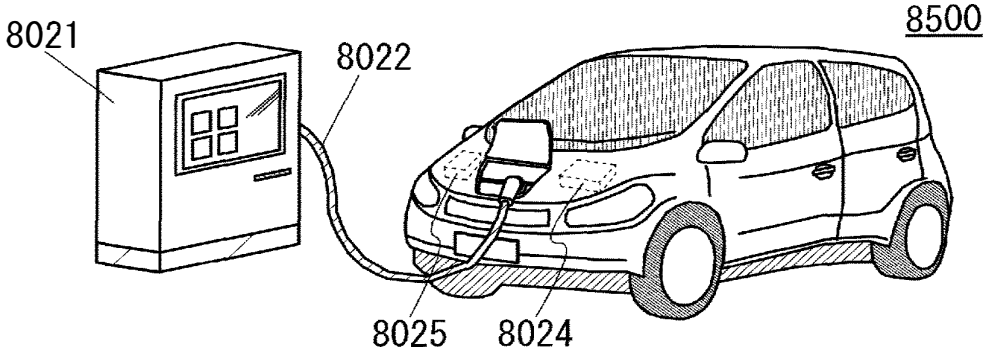

8104
8100
8101
8102
8103
8105
8200
8201
8203
8202
8000
8002
8003
8004
8001
8003
8107
8204
8300
8301
8302
8304
8303
8106

【図２７】

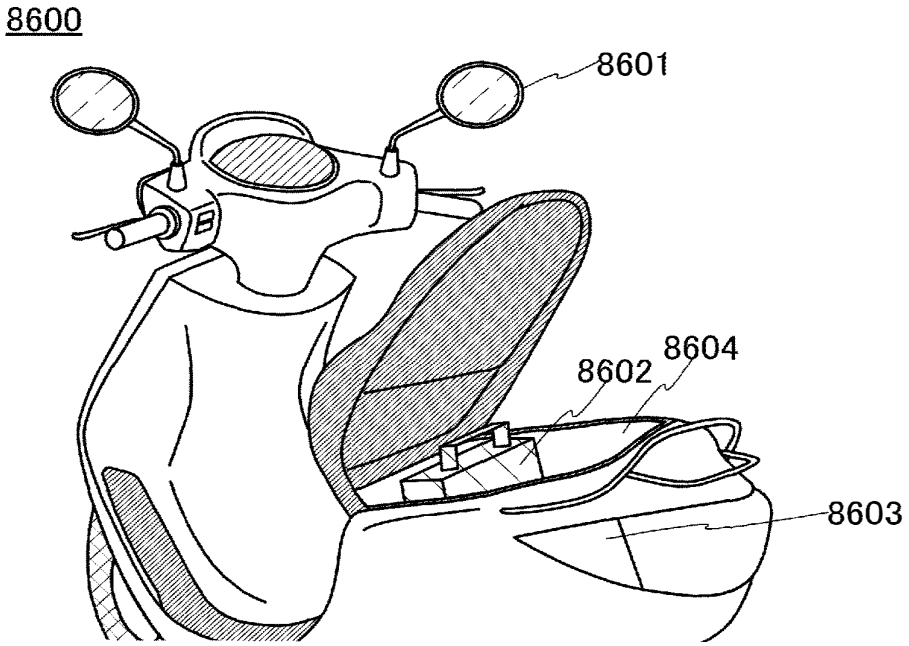
(A)


8400
8406
8101

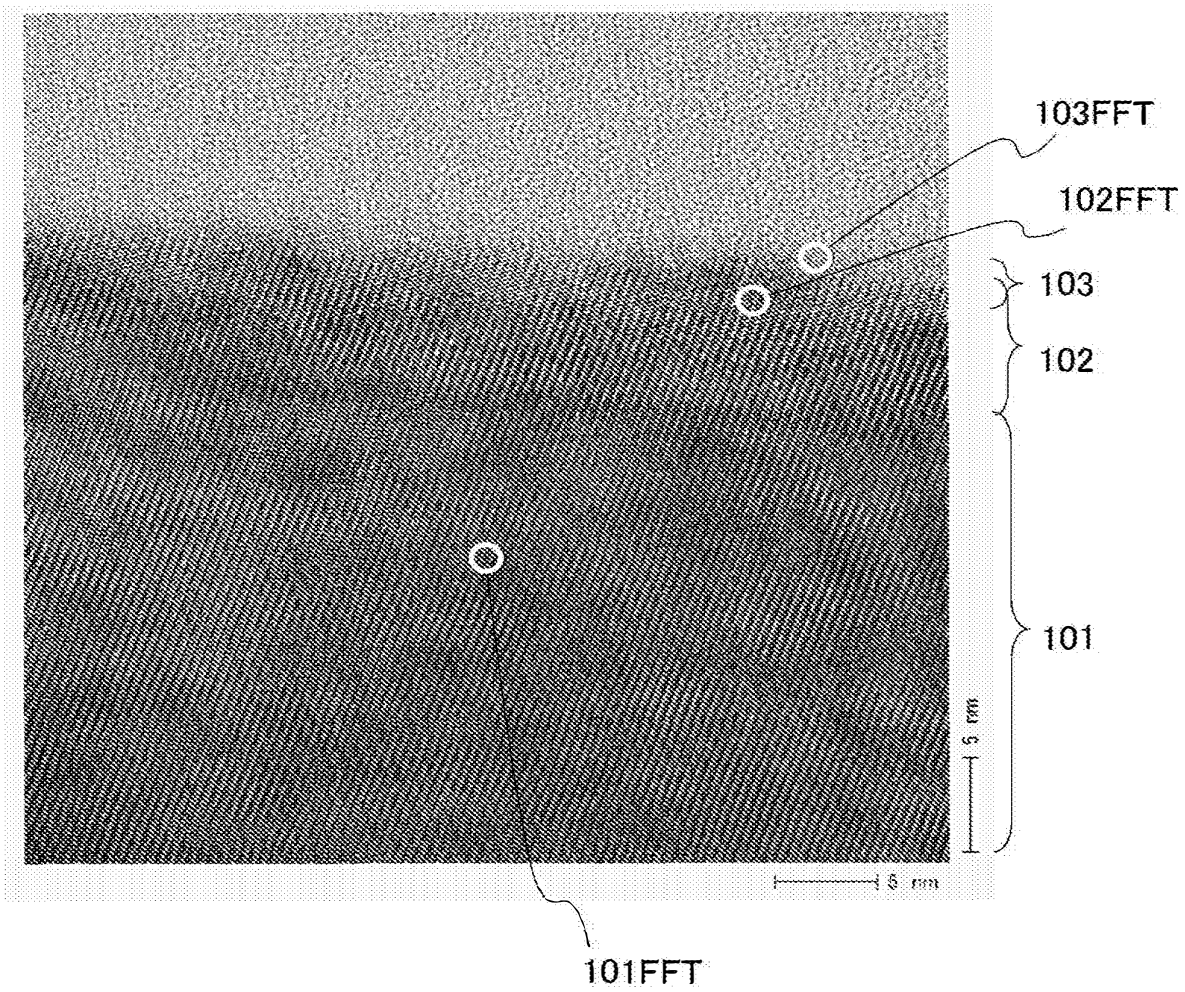
(B)


8021
8022
8500
8025
8024

(C)

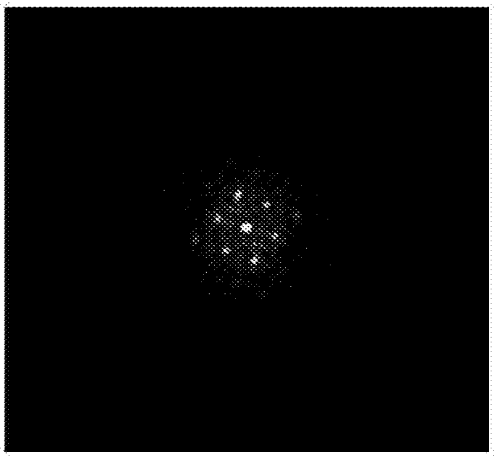

8600
8601
8602
8604
8603

【図２８】

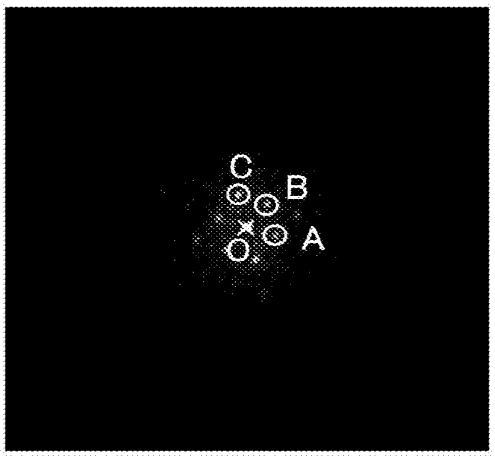


【図２９】

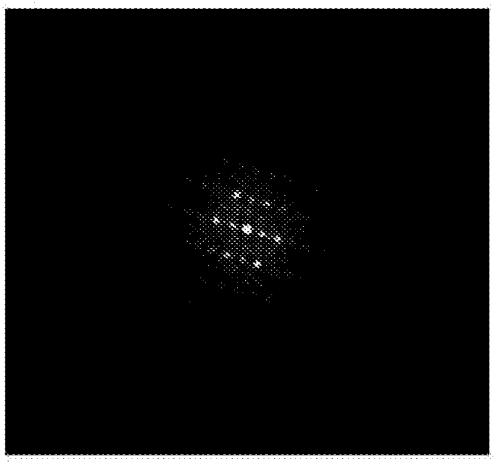
(A1)



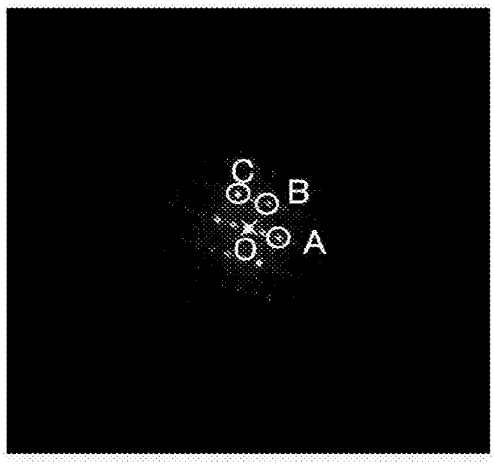
(A2)



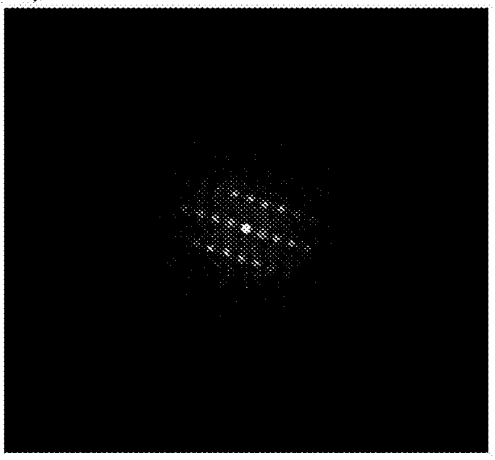
(B1)



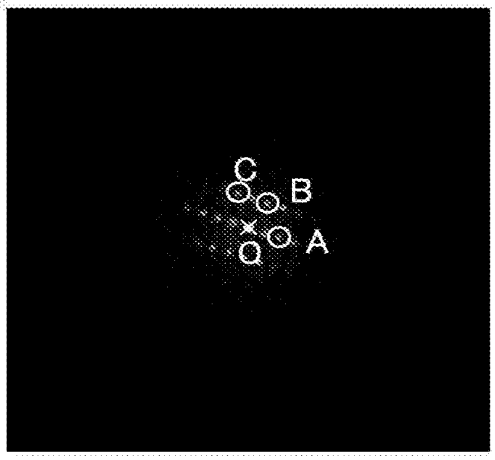
(B2)



(C1)

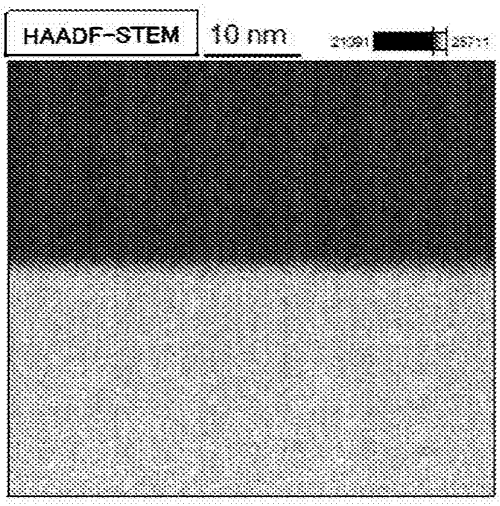


(C2)

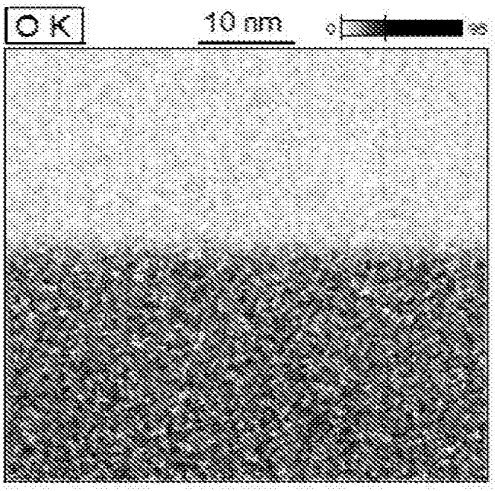


【図３０】

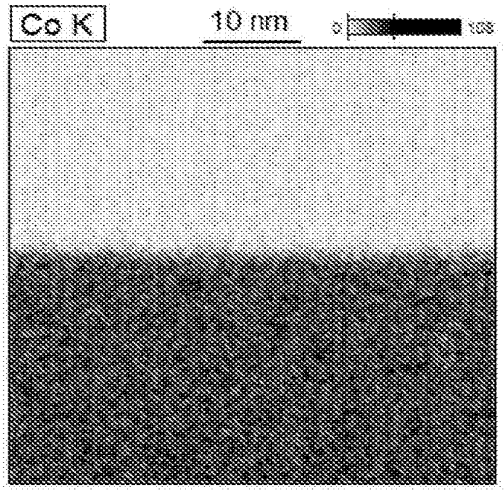
(A1)



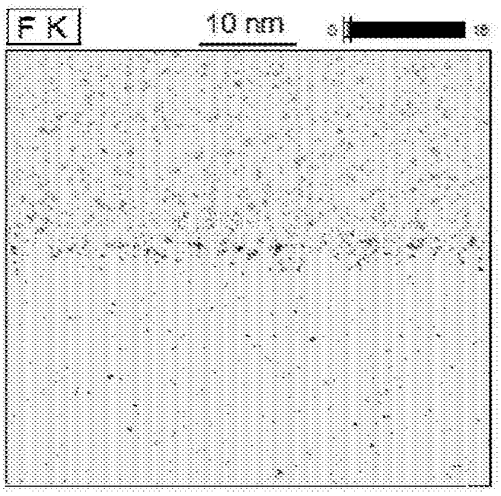
(A2)



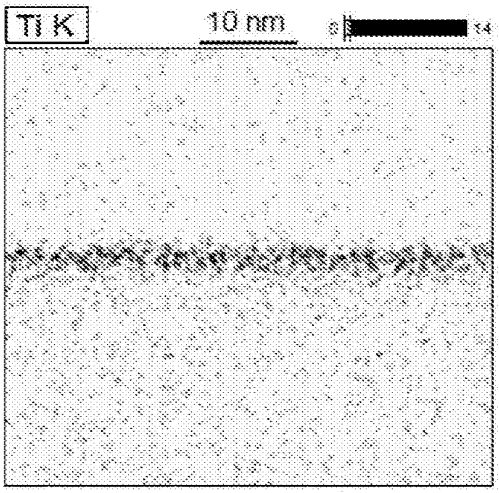
(B1)



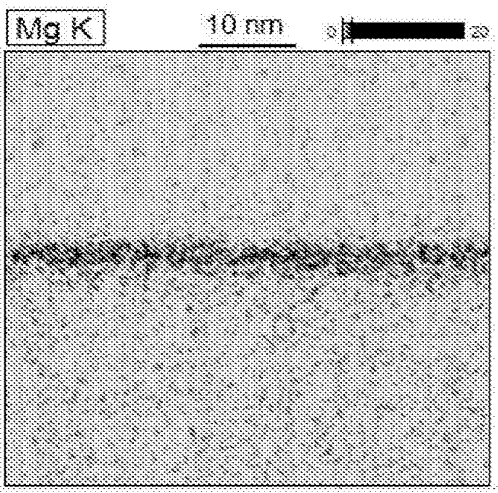
(B2)



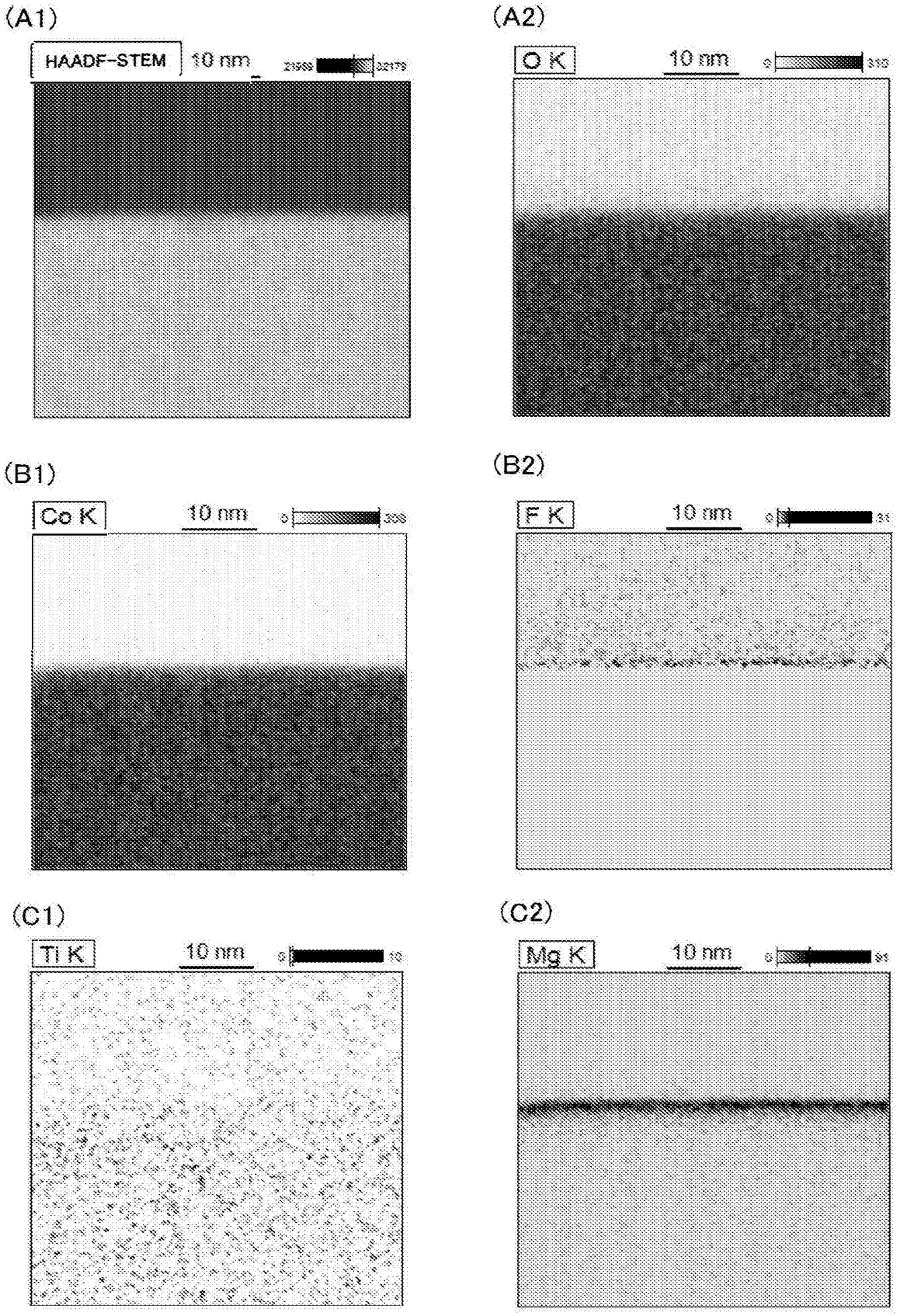
(C1)



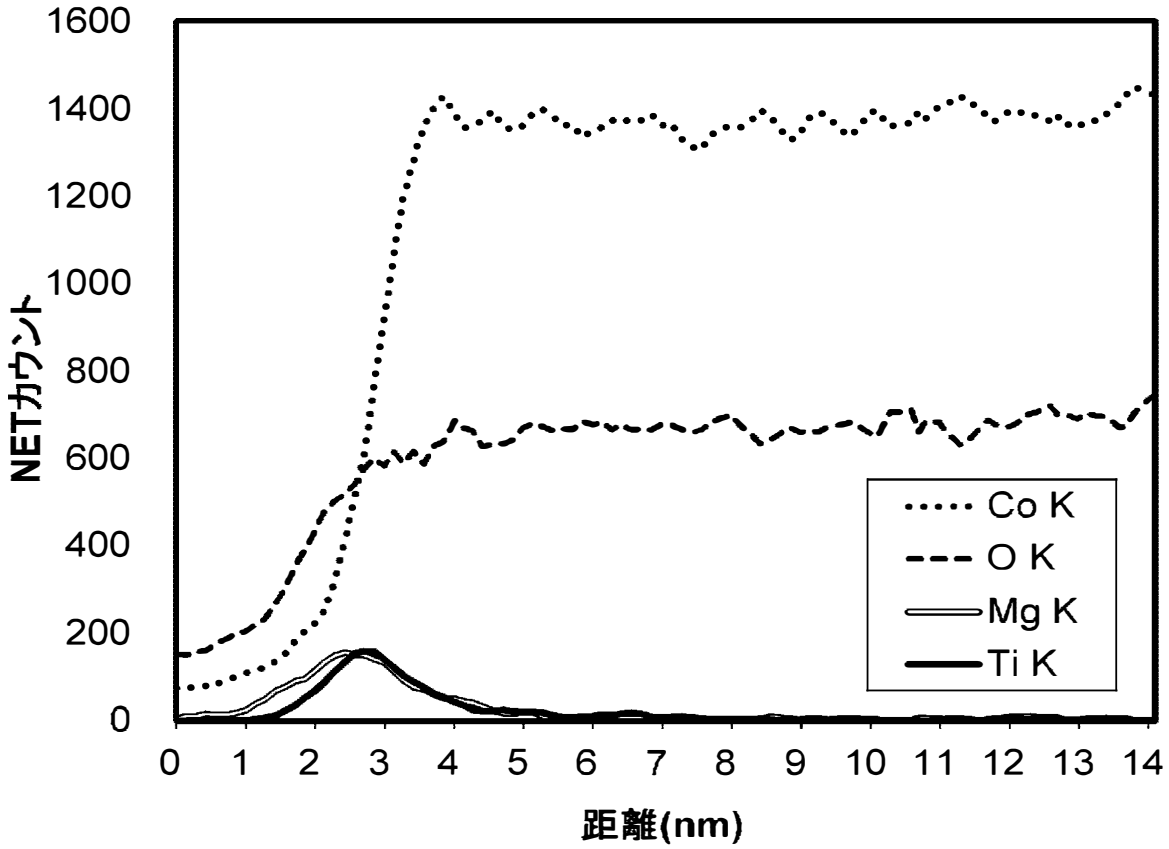
(C2)



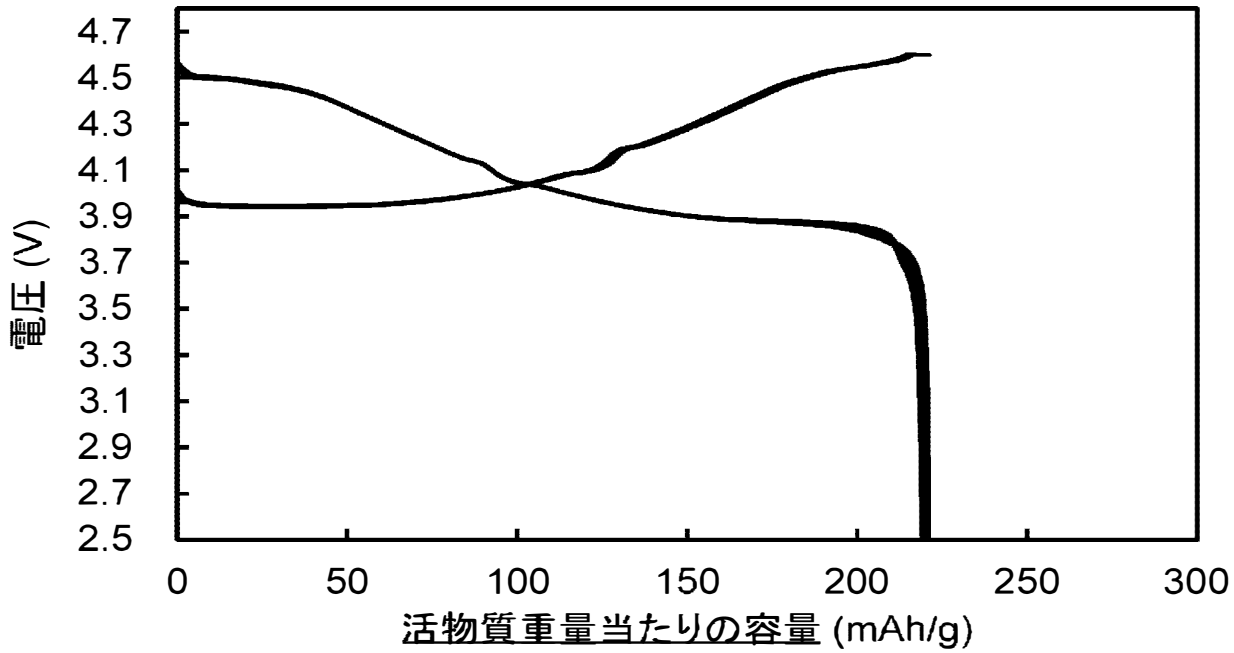
【図３１】


(A1)
HAADF-STEM
10 nm
(A2)
O K
10 nm
(B1)
Co K
10 nm
(B2)
F K
10 nm
(C1)
Ti K
10 nm
(C2)
Mg K
10 nm

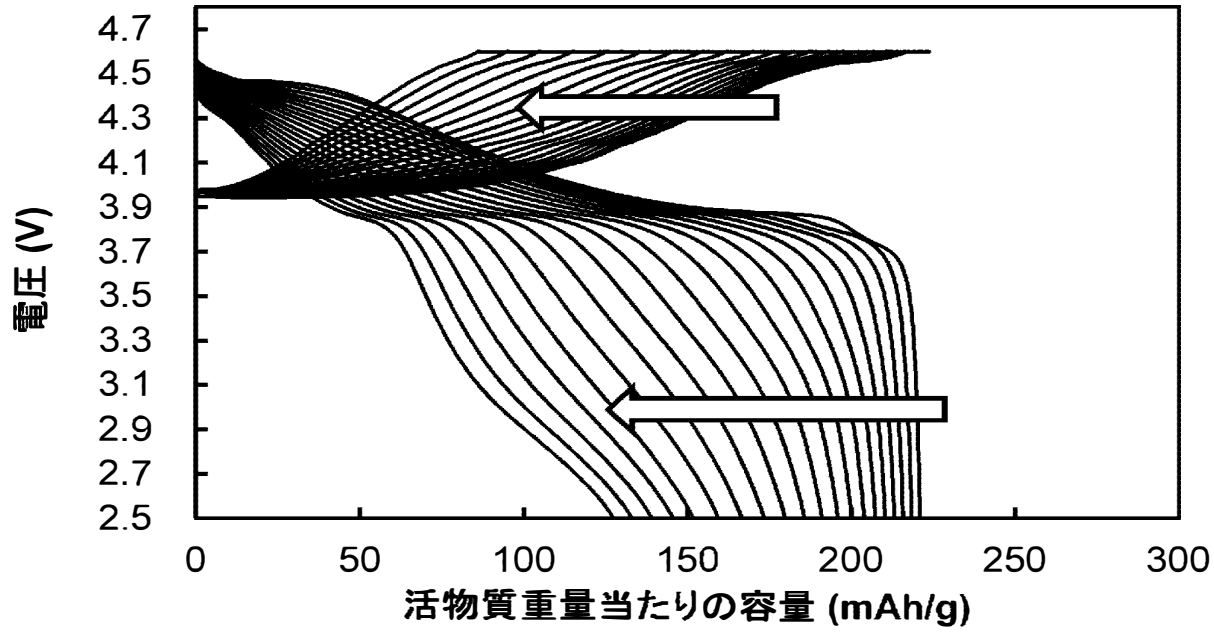
【図３２】


1600
1400
1200
1000
800
600
400
200
0
NETカウント
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14
距離(nm)
Co K
O K
Mg K
Ti K

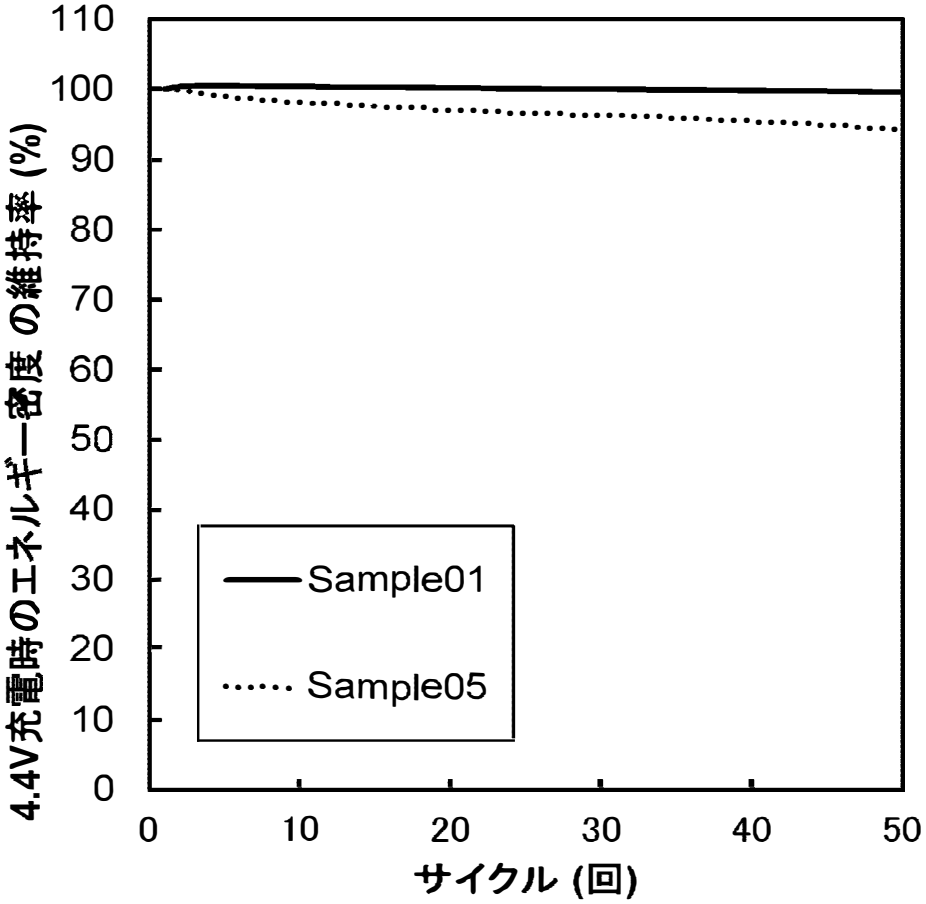
【図３３】


4.7
4.5
4.3
4.1
3.9
3.7
3.5
3.3
3.1
2.9
2.7
2.5
電圧 (V)
0 50 100 150 200 250 300
活物質重量当たりの容量 (mAh/g)

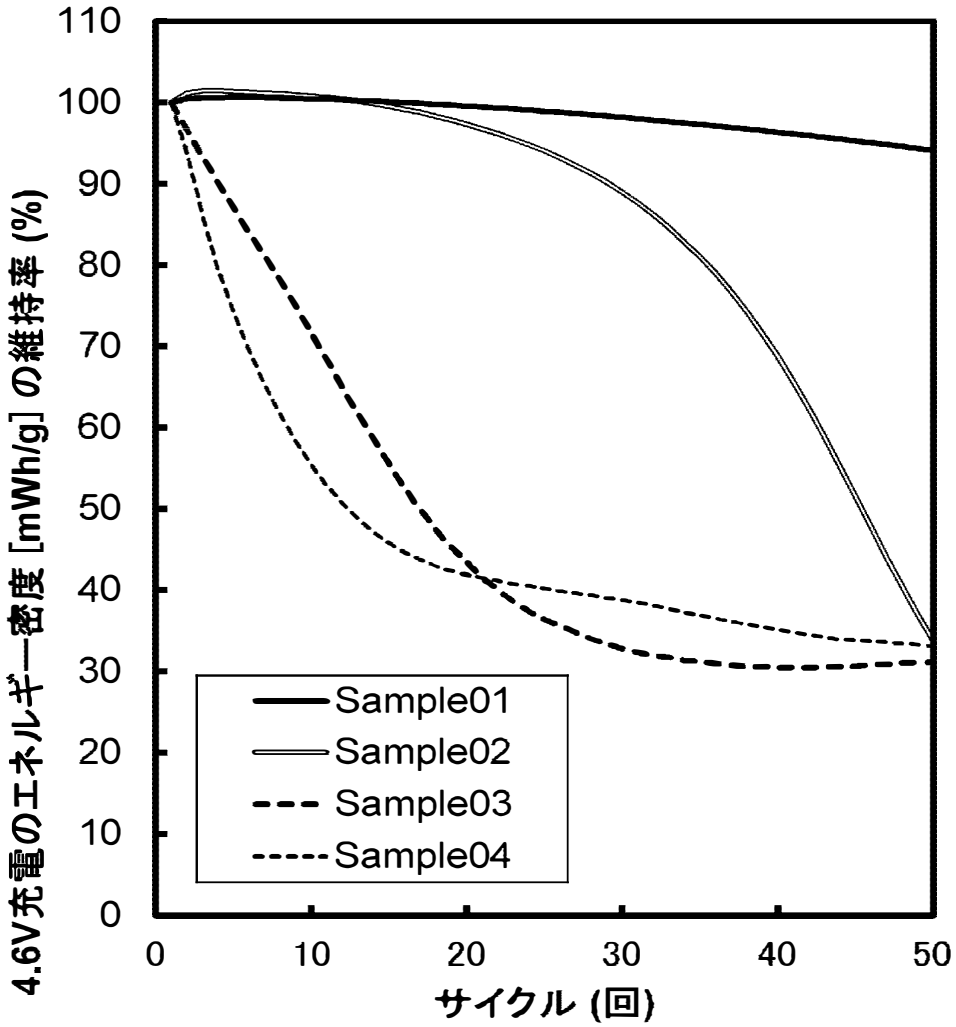
【図３４】


4.7
4.5
4.3
4.1
3.9
3.7
3.5
3.3
3.1
2.9
2.7
2.5
電圧 (V)
0
50
100
150
200
250
300
活物質重量当たりの容量 (mAh/g)

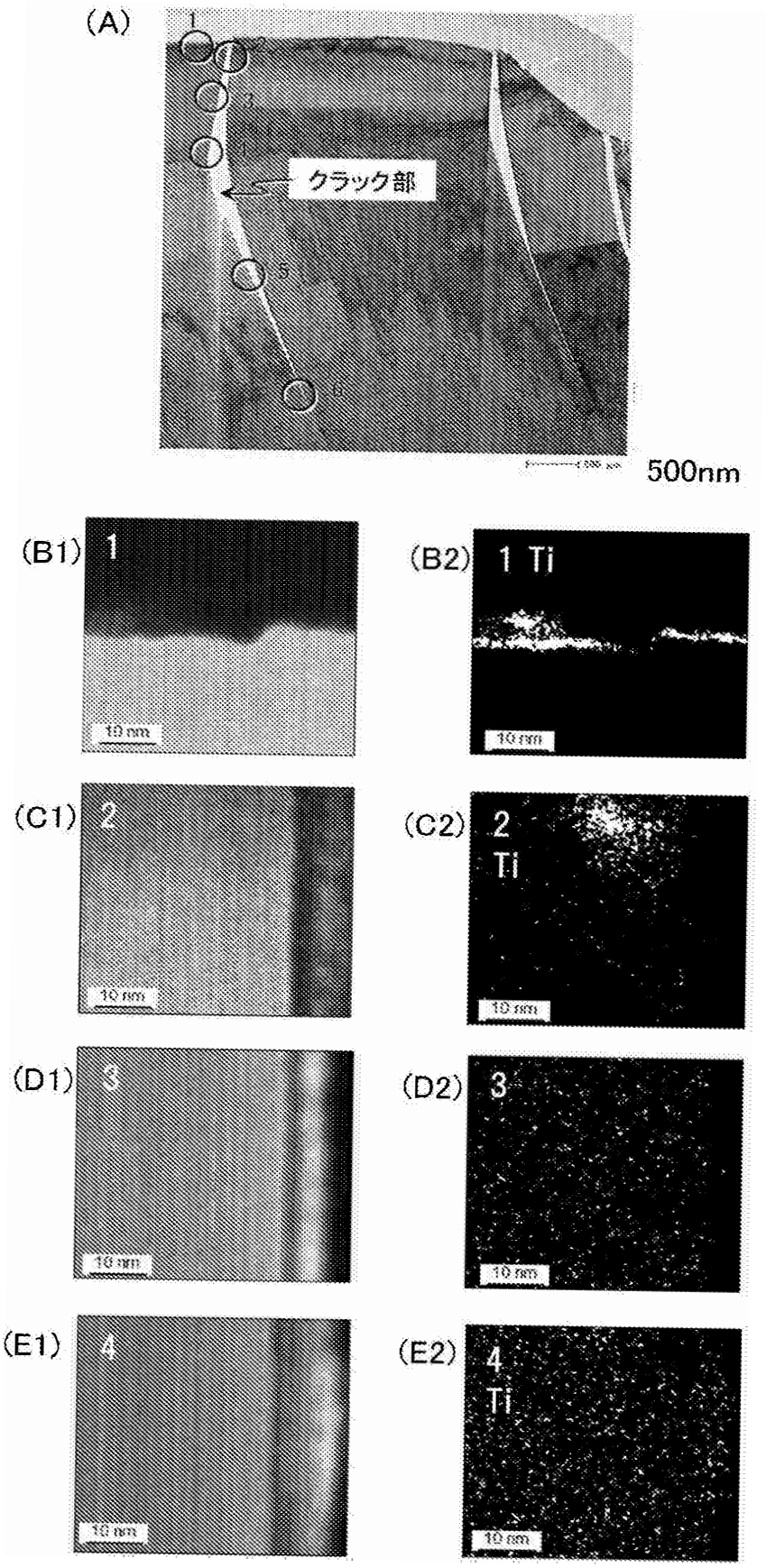
【図３５】


110
100
90
80
70
60
50
40
30
20
10
0
4.4V充電時のエネルギー密度の維持率 (%)
Sample01
Sample05
0
10
20
30
40
50
サイクル (回)

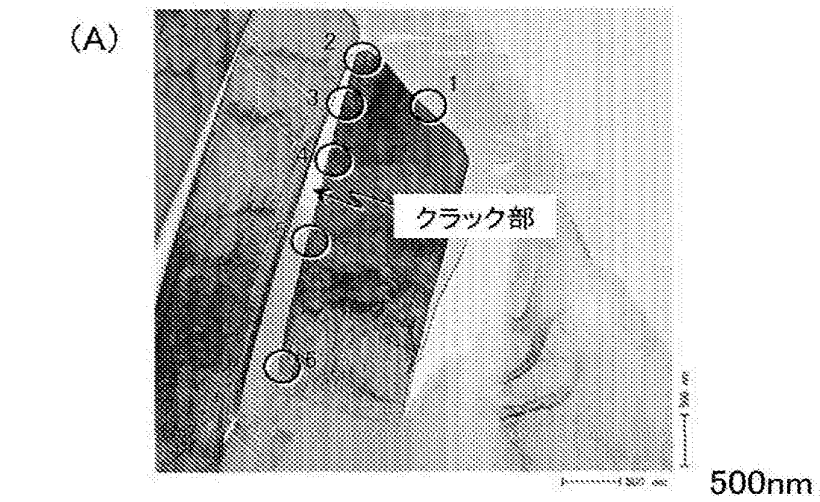
【図３６】


4.6V充電のエネルギー密度 [mWh/g] の維持率 (%)
サイクル (回)
Sample01
Sample02
Sample03
Sample04
0
10
20
30
40
50
60
70
80
90
100
110

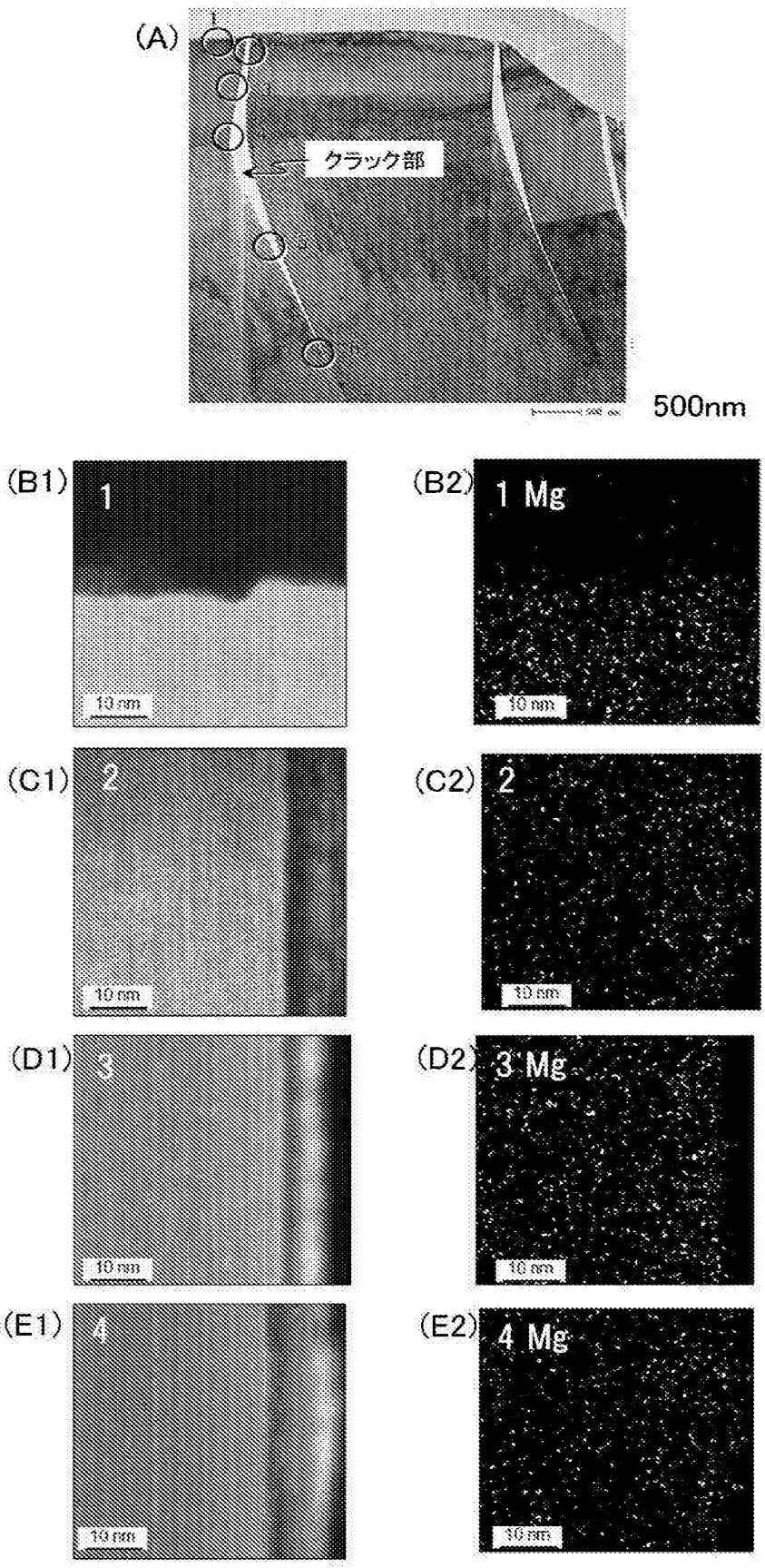
【図３７】


(A)
1
クラック部
500nm
(B1)
1
10 nm
(B2)
1 Ti
10 nm
(C1)
2
10 nm
(C2)
2
Ti
10 nm
(D1)
3
10 nm
(D2)
3
10 nm
(E1)
4
10 nm
(E2)
4
Ti
10 nm

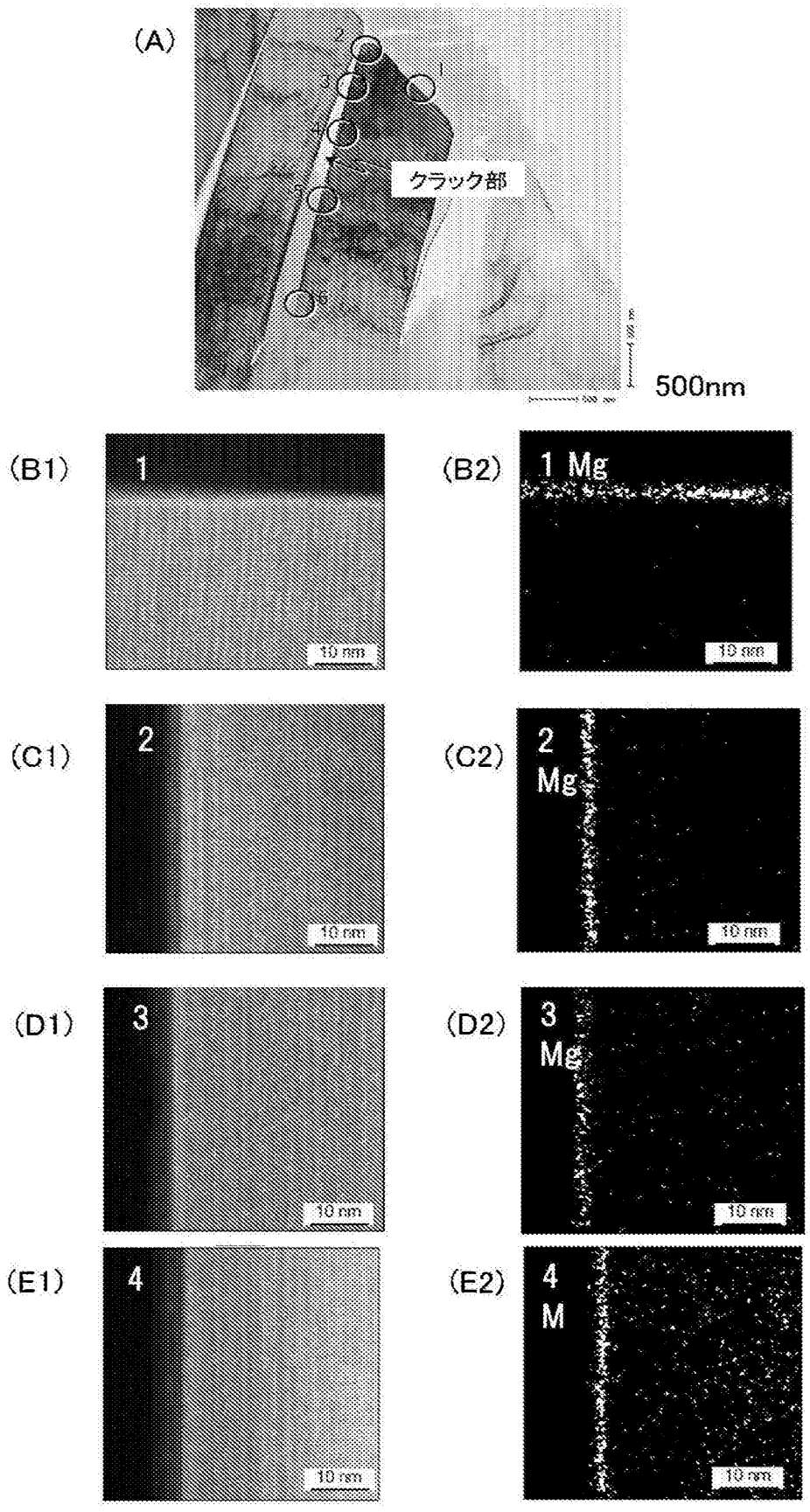
【図３８】


(A)
1
2
3
4
5
クラック部
500nm
(B1)
1
10 nm
(B2)
1 Ti
10 nm
(C1)
2
10 nm
(C2)
2
Ti
10 nm
(D1)
3
10 nm
(D2)
3
10 nm
(E1)
4
10 nm
(E2)
4
Ti
10 nm

【図３９】


(A)
クラック部
500nm
(B1)
1
10 nm
(B2)
1 Mg
10 nm
(C1)
2
10 nm
(C2)
2
10 nm
(D1)
3
10 nm
(D2)
3 Mg
10 nm
(E1)
4
10 nm
(E2)
4 Mg
10 nm

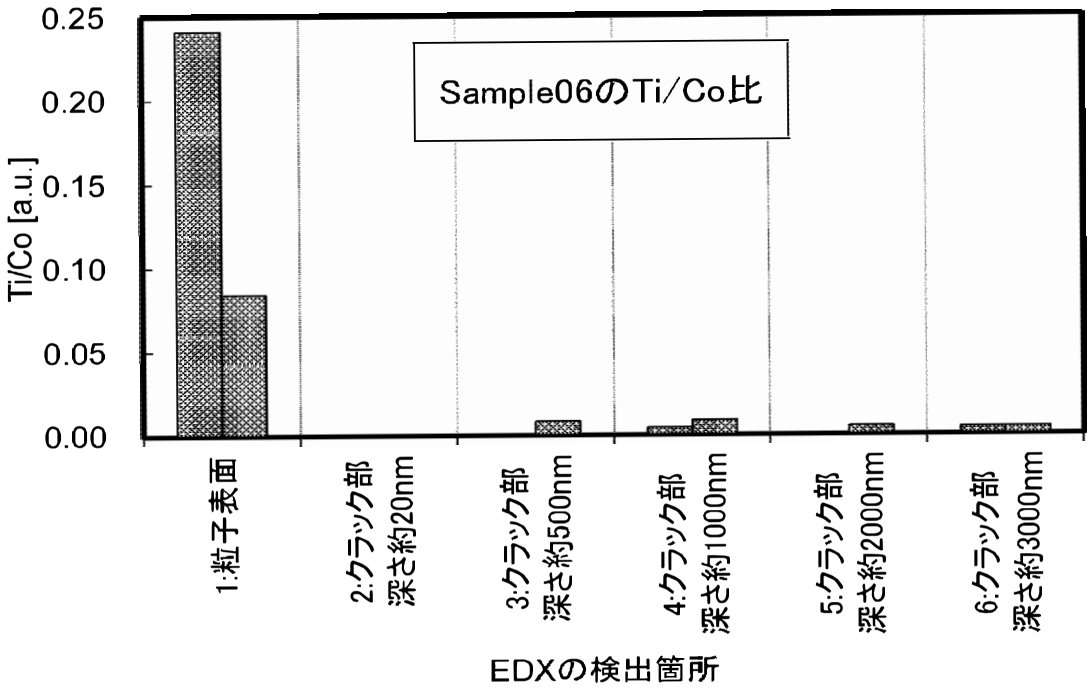
【図４０】


(A)
クラック部
500nm
(B1)
1
10 nm
(B2)
1 Mg
10 nm
(C1)
2
10 nm
(C2)
2
Mg
10 nm
(D1)
3
10 nm
(D2)
3
Mg
10 nm
(E1)
4
10 nm
(E2)
4
M
10 nm

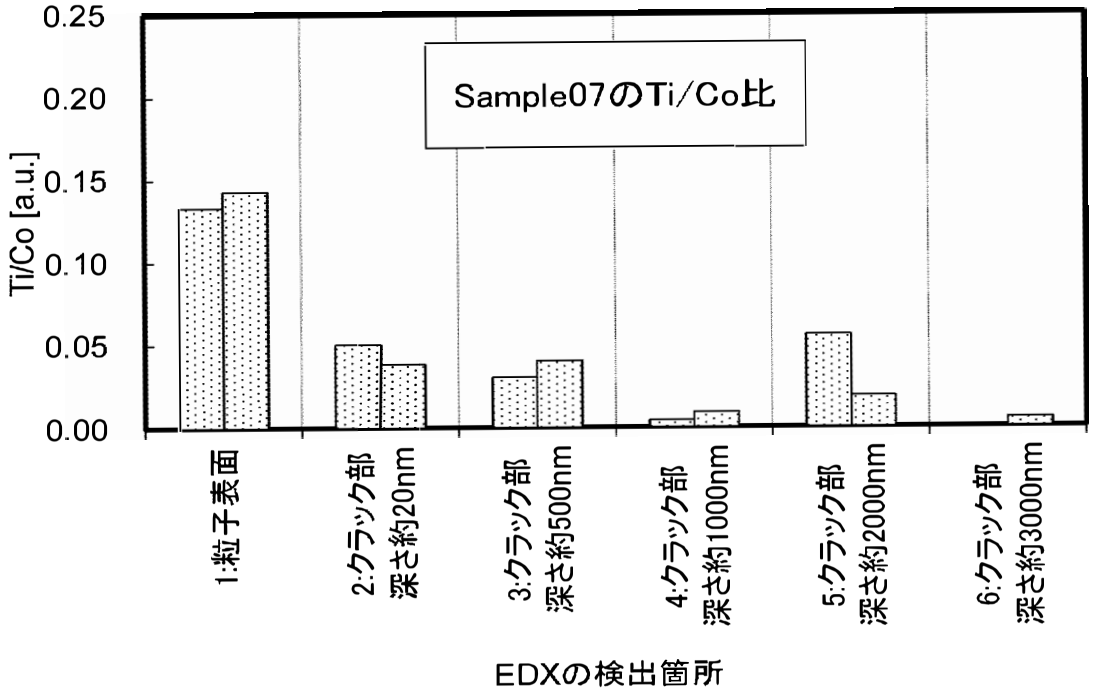
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【図４１】

(A)



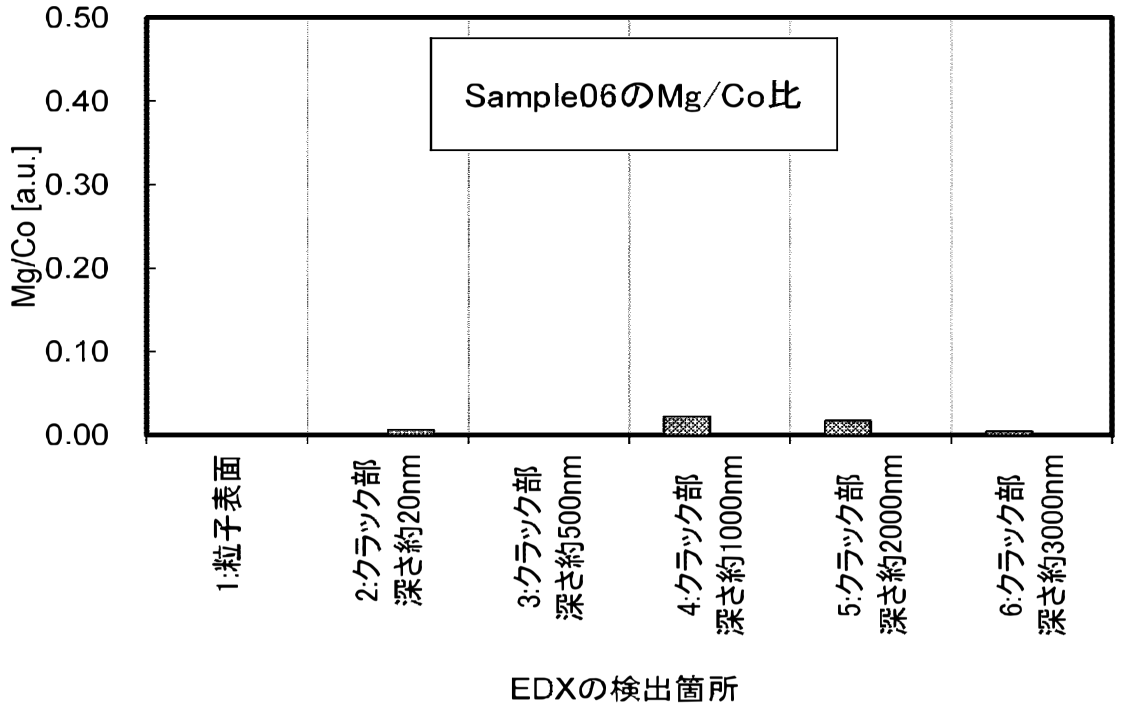
(B)



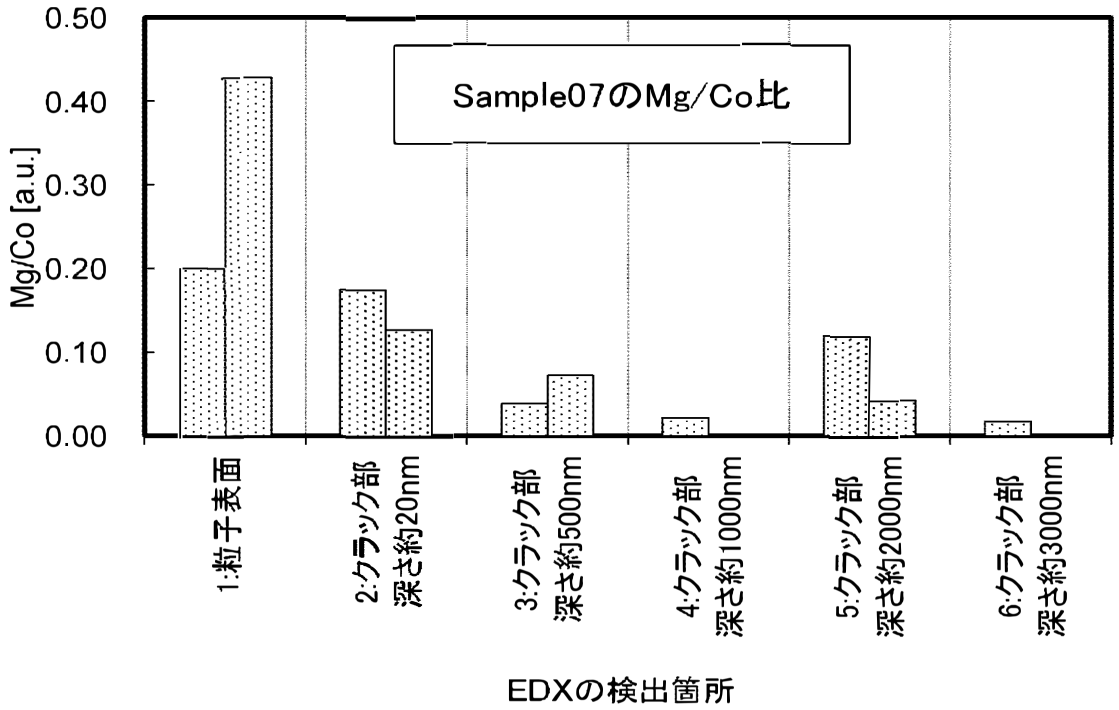
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【図４２】

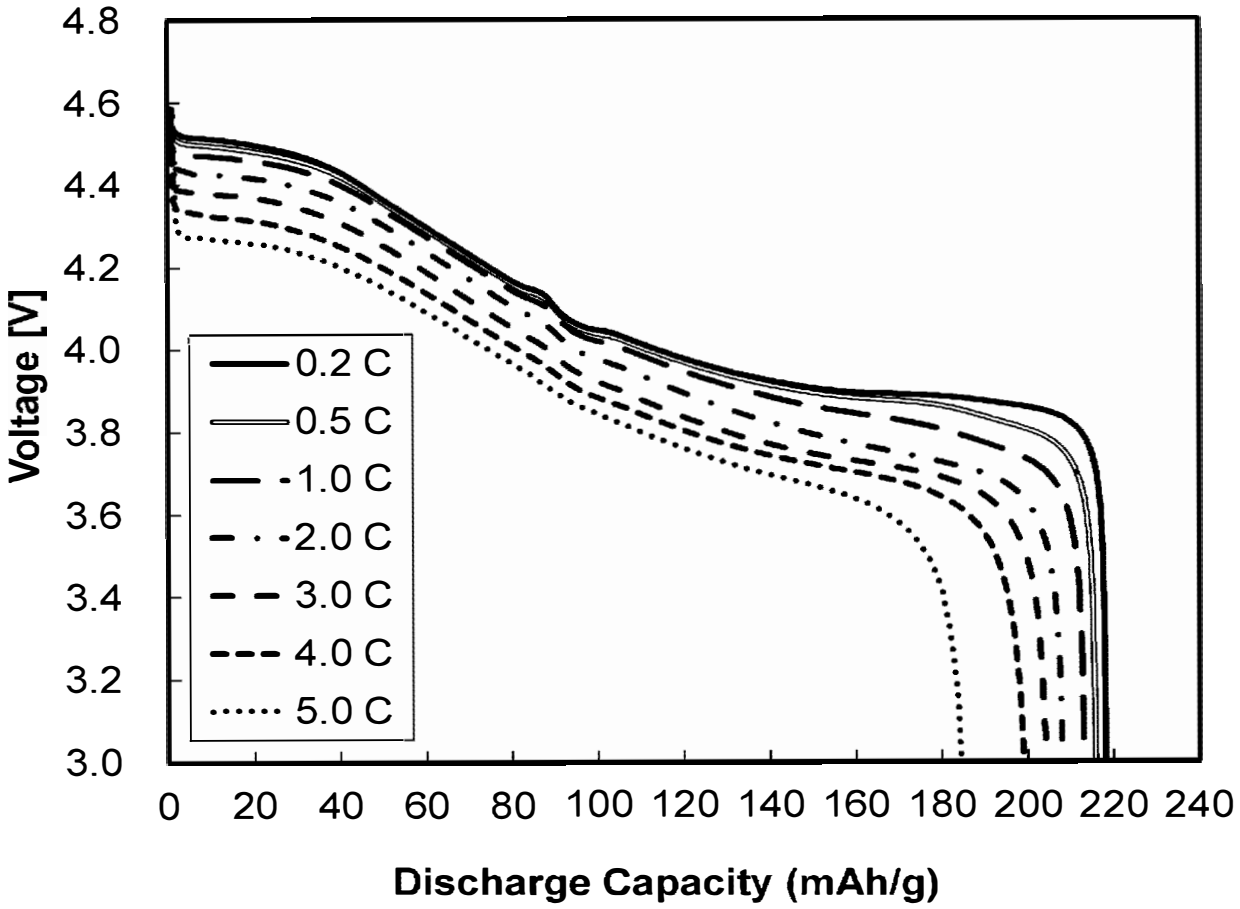
(A)



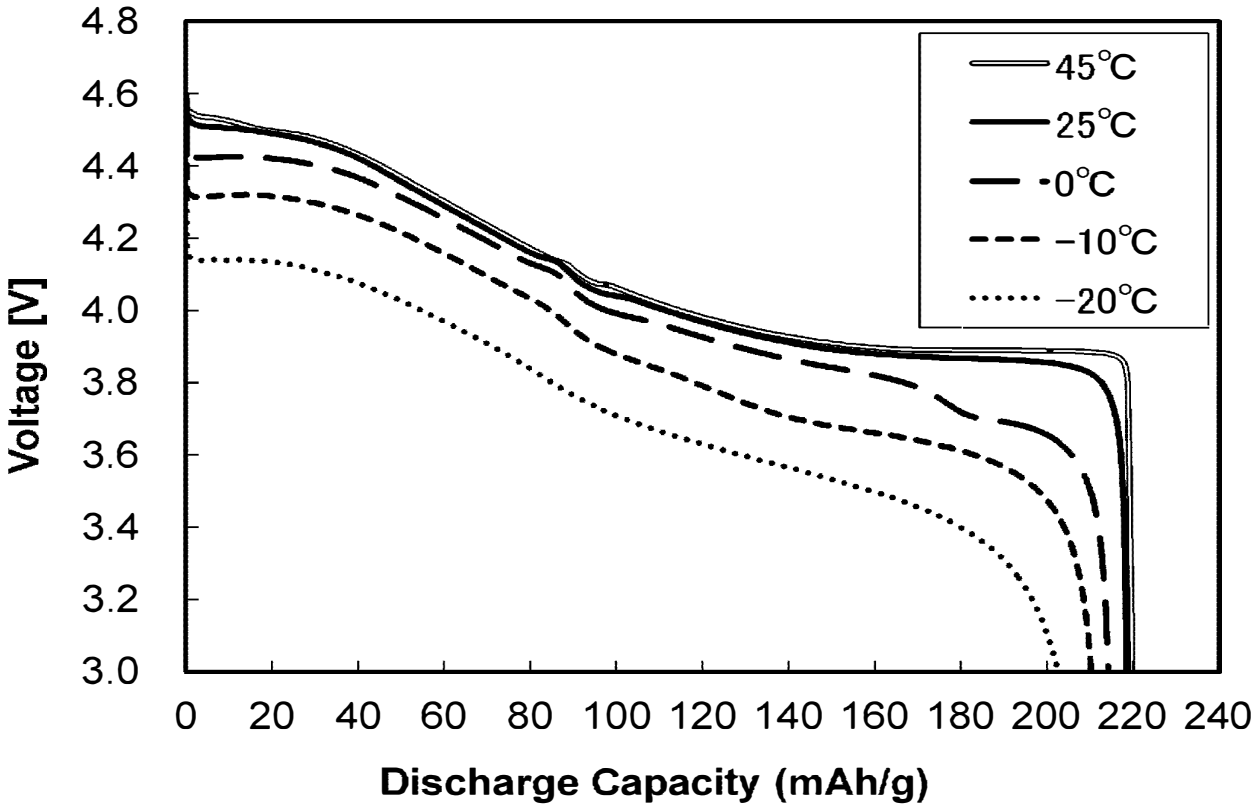
(B)



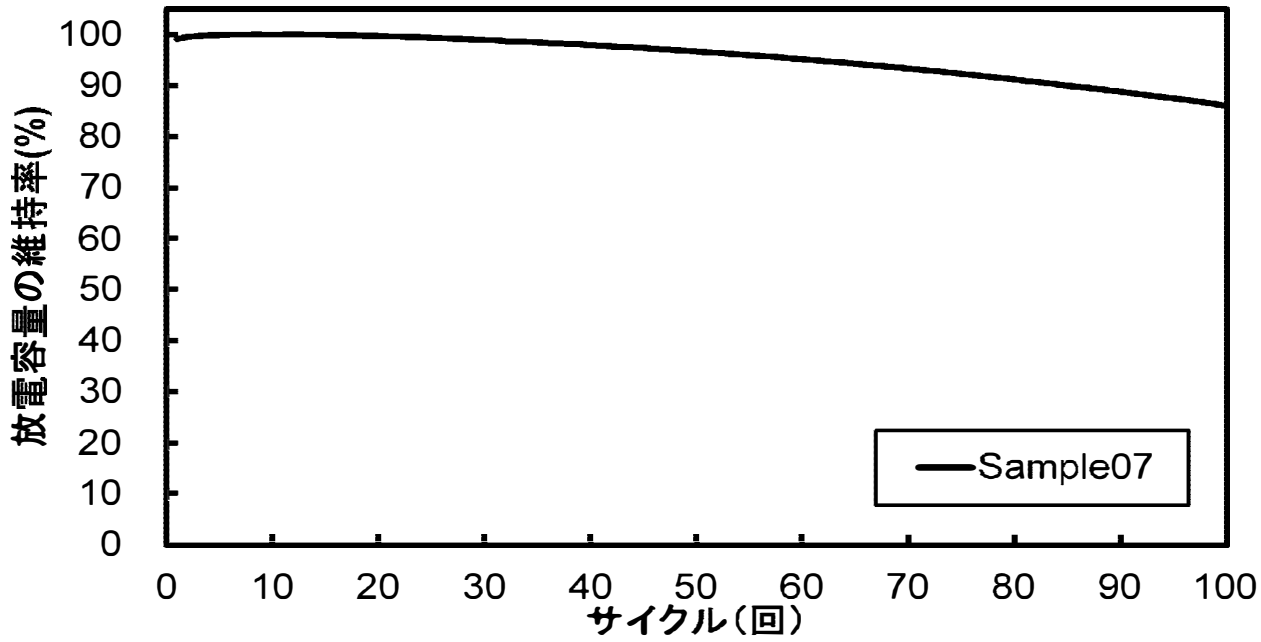
【図４３】


4.8
4.6
4.4
4.2
4.0
3.8
3.6
3.4
3.2
3.0
Voltage [V]
0.2 C
0.5 C
1.0 C
2.0 C
3.0 C
4.0 C
5.0 C
0
20
40
60
80
100
120
140
160
180
200
220
240
Discharge Capacity (mAh/g)

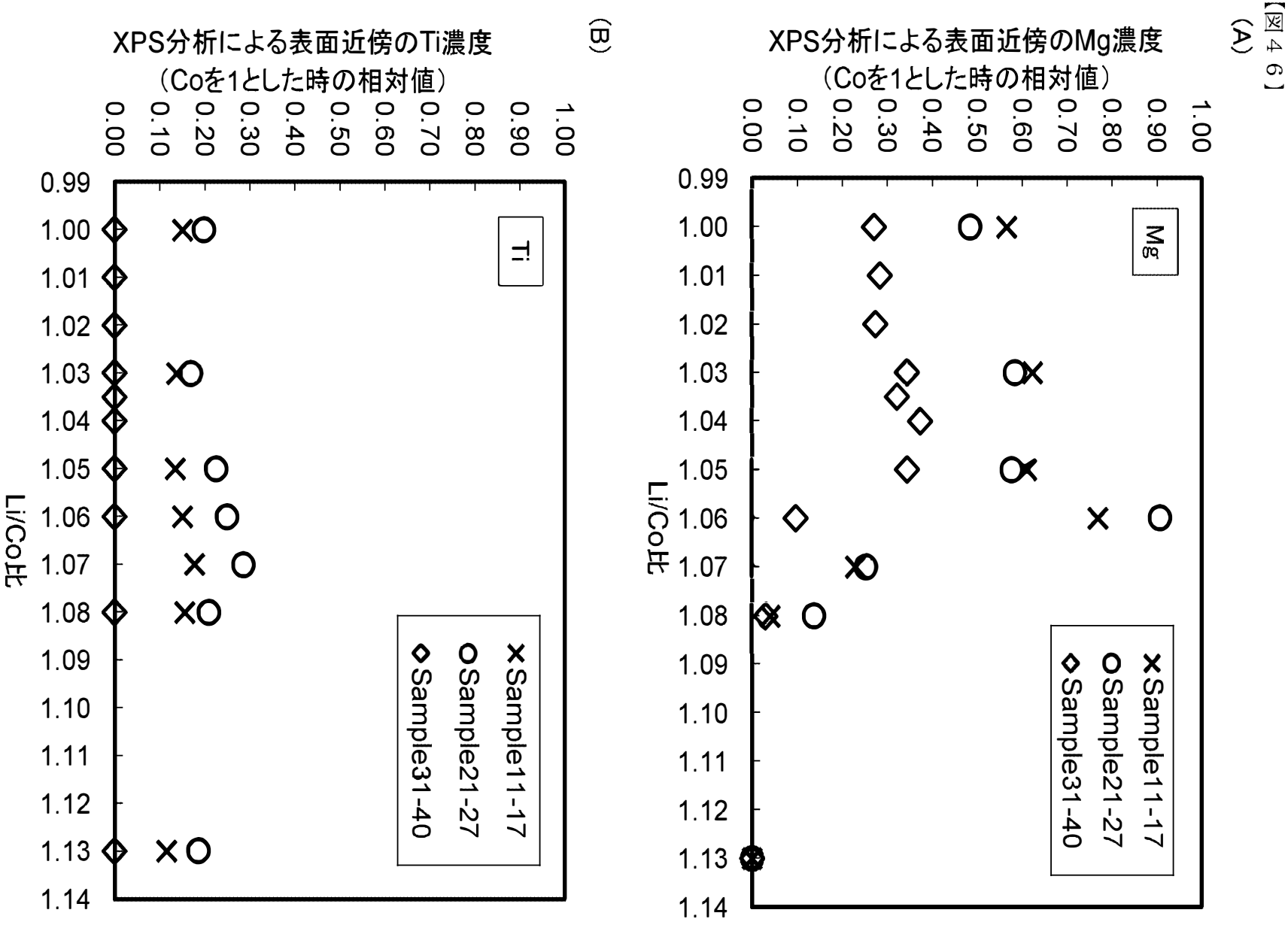
【図４４】


4.8
4.6
4.4
4.2
4.0
3.8
3.6
3.4
3.2
3.0
Voltage [V]
0 20 40 60 80 100 120 140 160 180 200 220 240
Discharge Capacity (mAh/g)
45℃
25℃
0℃
−10℃
−20℃

【図４５】

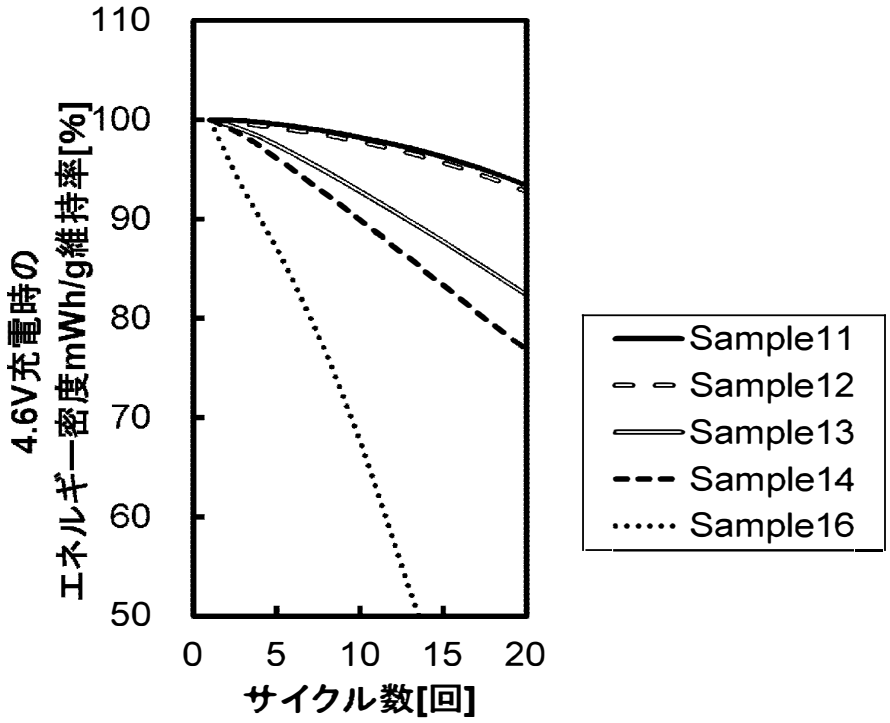

100
90
80
70
60
50
40
30
20
10
0
放電容量の維持率(%)
0 10 20 30 40 50 60 70 80 90 100
サイクル（回）
Sample07

【図４６】

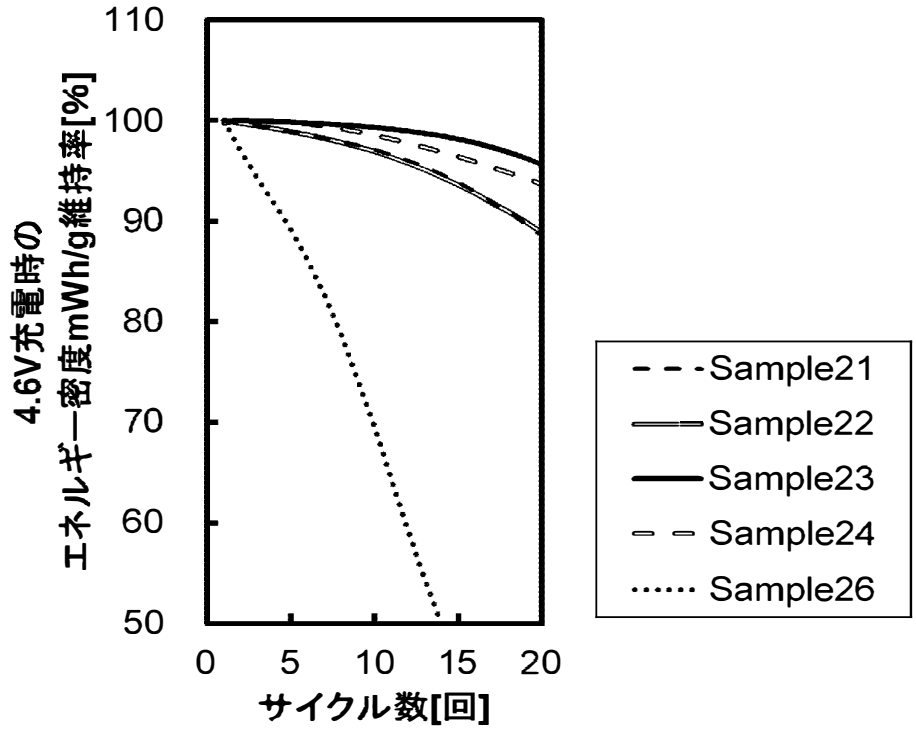

(A)
XPS分析による表面近傍のMg濃度
（Coを1とした時の相対値）
0.00
0.10
0.20
0.30
0.40
0.50
0.60
0.70
0.80
0.90
1.00
0.99
1.00
1.01
1.02
1.03
1.04
1.05
1.06
1.07
1.08
1.09
1.10
1.11
1.12
1.13
1.14
Li/Co比
Mg
×Sample11-17
○Sample21-27
◇Sample31-40
(B)
XPS分析による表面近傍のTi濃度
（Coを1とした時の相対値）
0.00
0.10
0.20
0.30
0.40
0.50
0.60
0.70
0.80
0.90
1.00
0.99
1.00
1.01
1.02
1.03
1.04
1.05
1.06
1.07
1.08
1.09
1.10
1.11
1.12
1.13
1.14
Li/Co比
Ti
×Sample11-17
○Sample21-27
◇Sample31-40

【図４７】

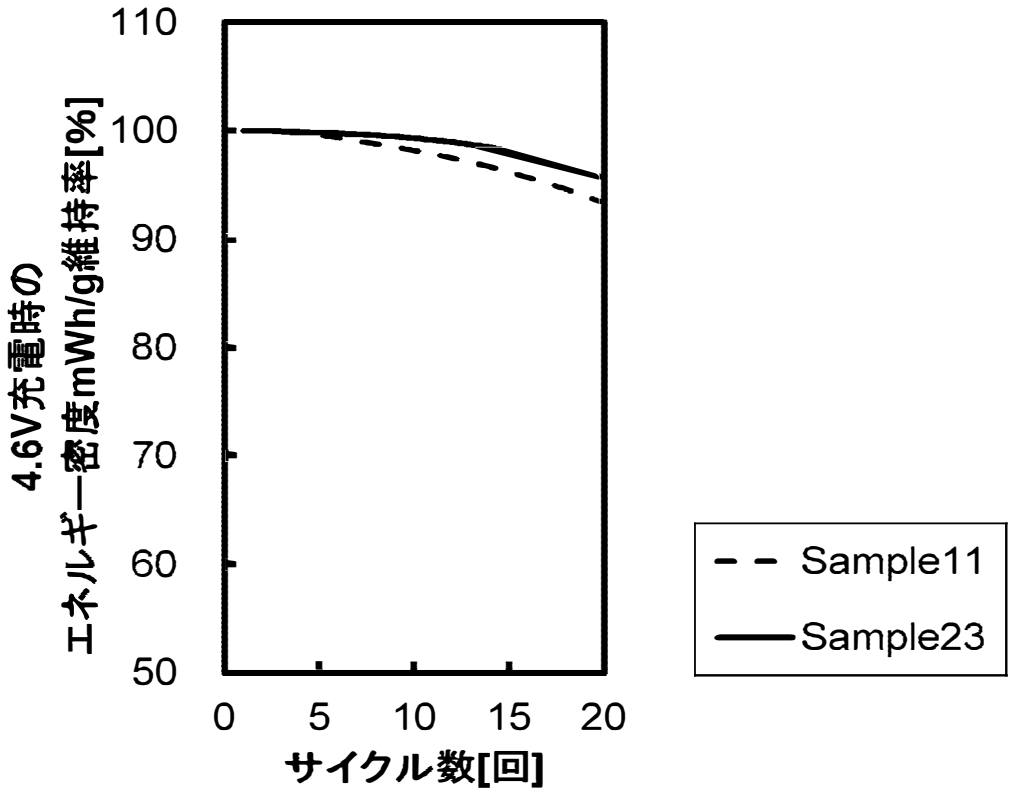
(A)


110
100
90
80
70
60
50
4.6V充電時の
エネルギー密度mWh/g維持率[%]
0
5
10
15
20
サイクル数[回]
Sample11
Sample12
Sample13
Sample14
Sample16

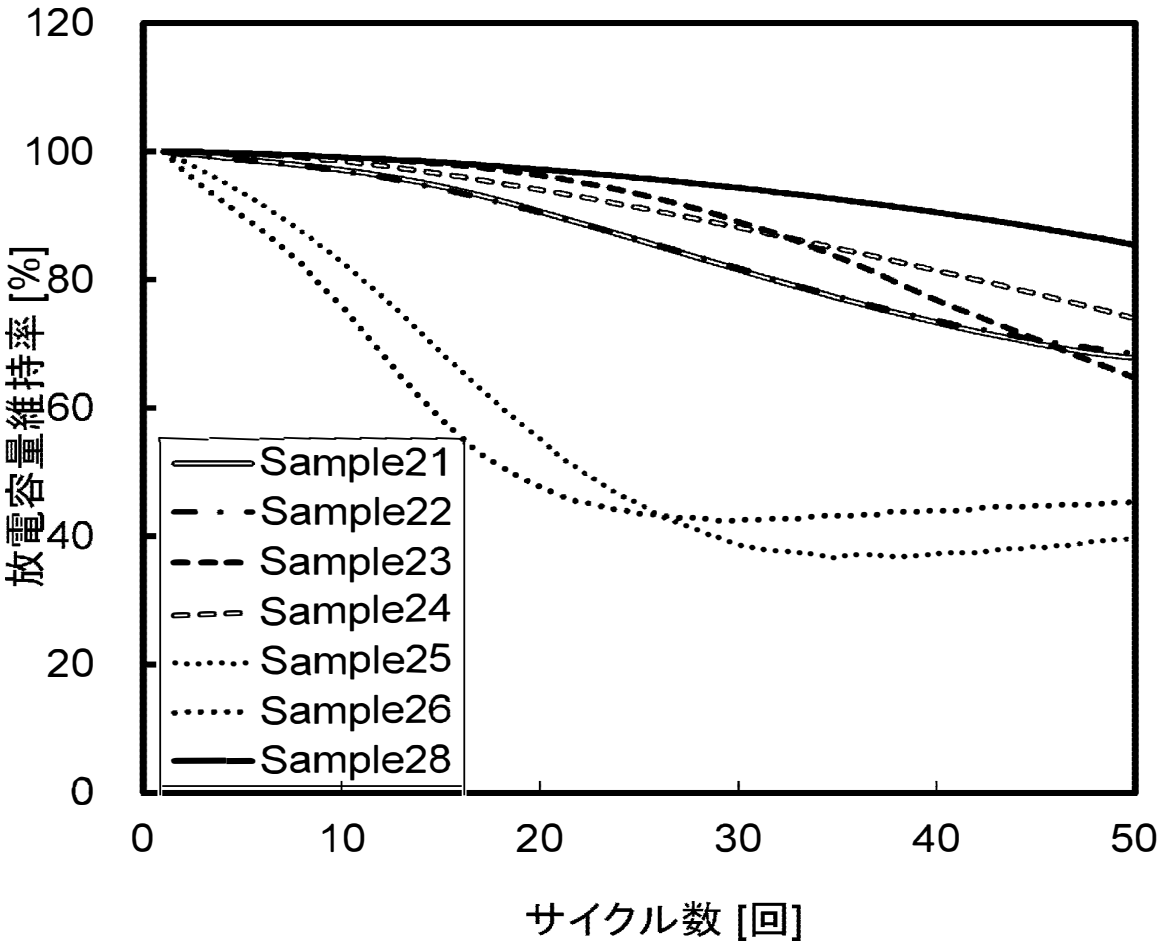
(B)


110
100
90
80
70
60
50
4.6V充電時の
エネルギー密度mWh/g維持率[%]
0
5
10
15
20
サイクル数[回]
Sample21
Sample22
Sample23
Sample24
Sample26

【図４８】

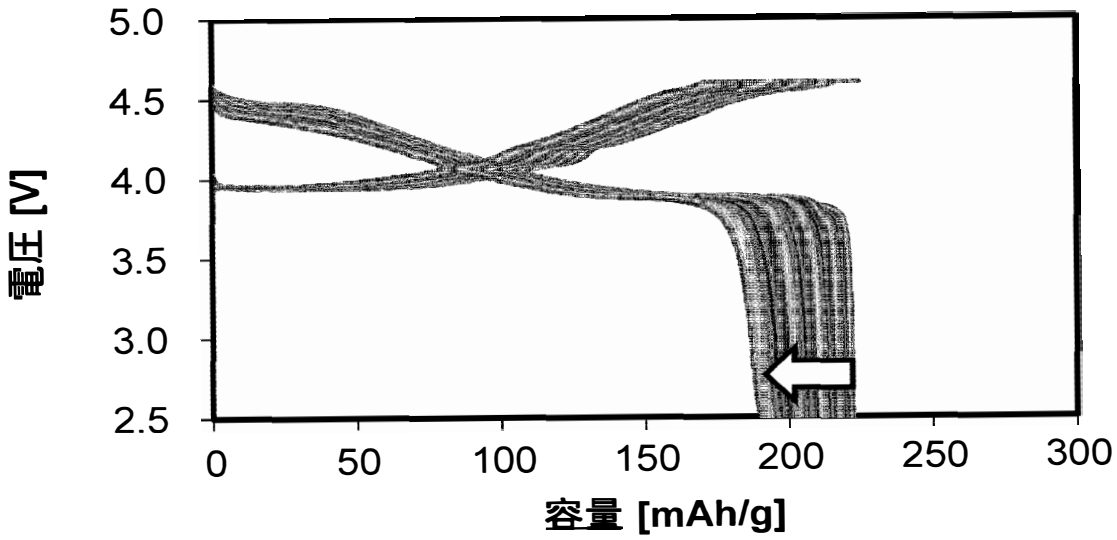

110
100
90
80
70
60
50
4.6V充電時の
エネルギー密度mWh/g維持率[%]
0
5
10
15
20
サイクル数[回]
Sample11
Sample23

【図４９】

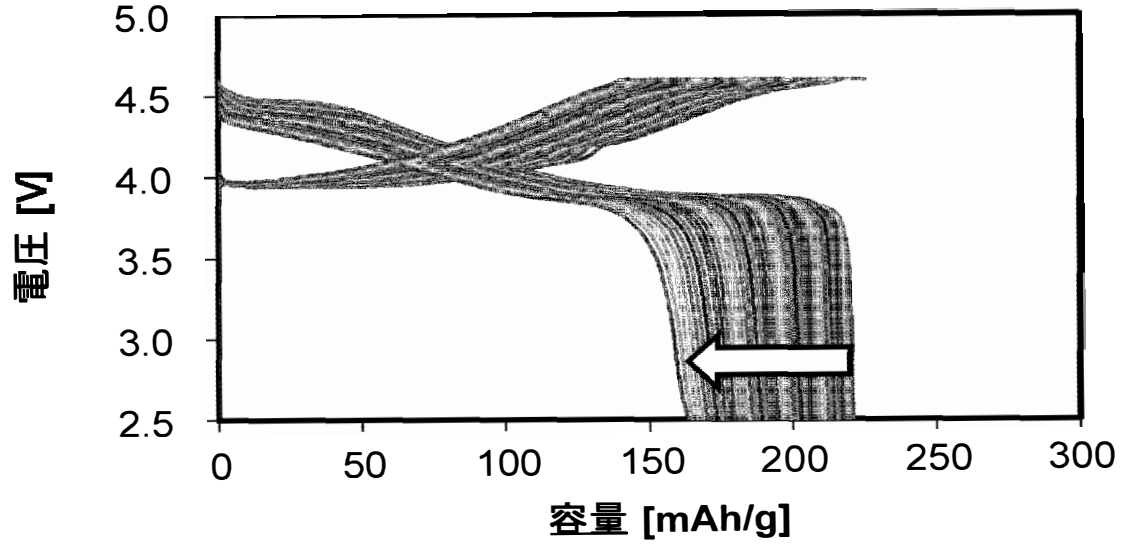

120
100
80
60
40
20
0
放電容量維持率 [%]
0
10
20
30
40
50
サイクル数 [回]
Sample21
Sample22
Sample23
Sample24
Sample25
Sample26
Sample28

【図５０】

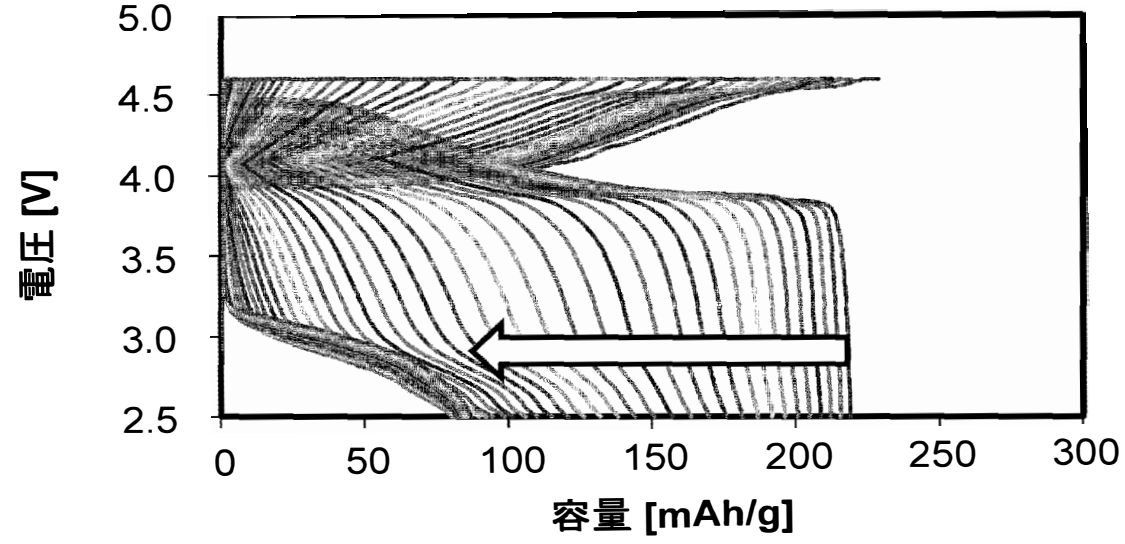
(A)


5.0
4.5
4.0
3.5
3.0
2.5
電圧 [V]
0
50
100
150
200
250
300
容量 [mAh/g]

(B)

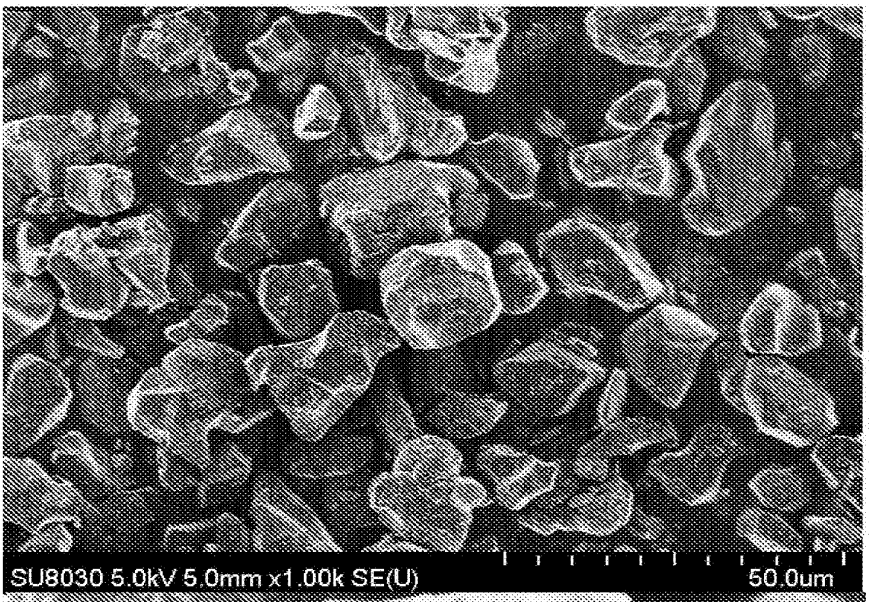

5.0
4.5
4.0
3.5
3.0
2.5
電圧 [V]
0
50
100
150
200
250
300
容量 [mAh/g]

(C)

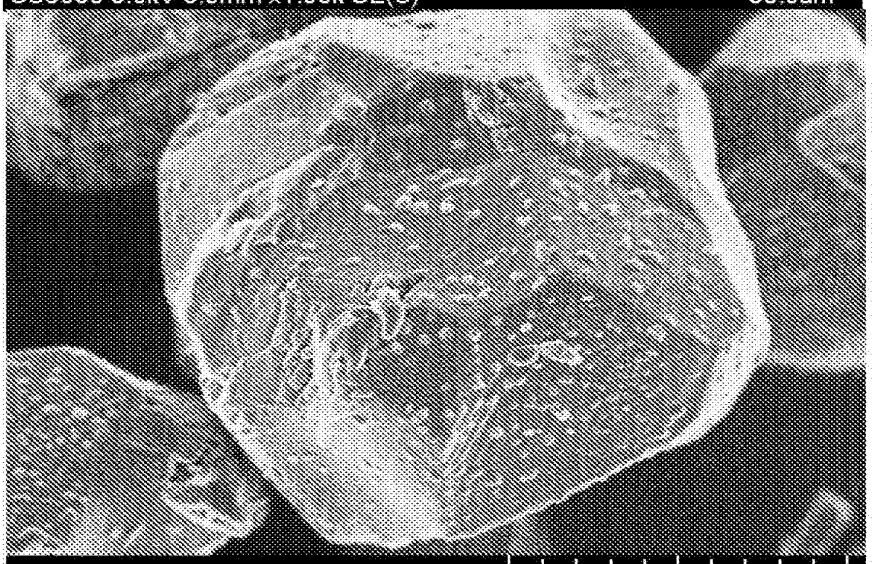

5.0
4.5
4.0
3.5
3.0
2.5
電圧 [V]
0
50
100
150
200
250
300
容量 [mAh/g]

【図５１】

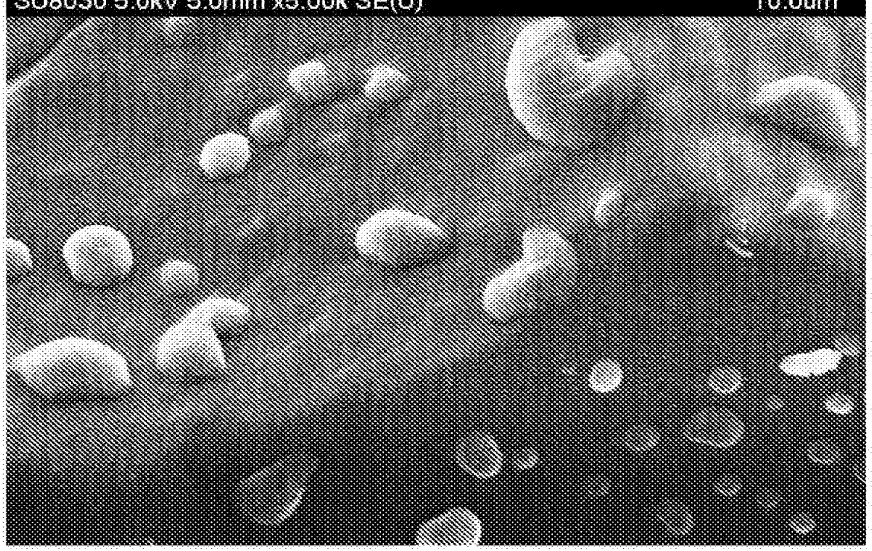
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50.0um

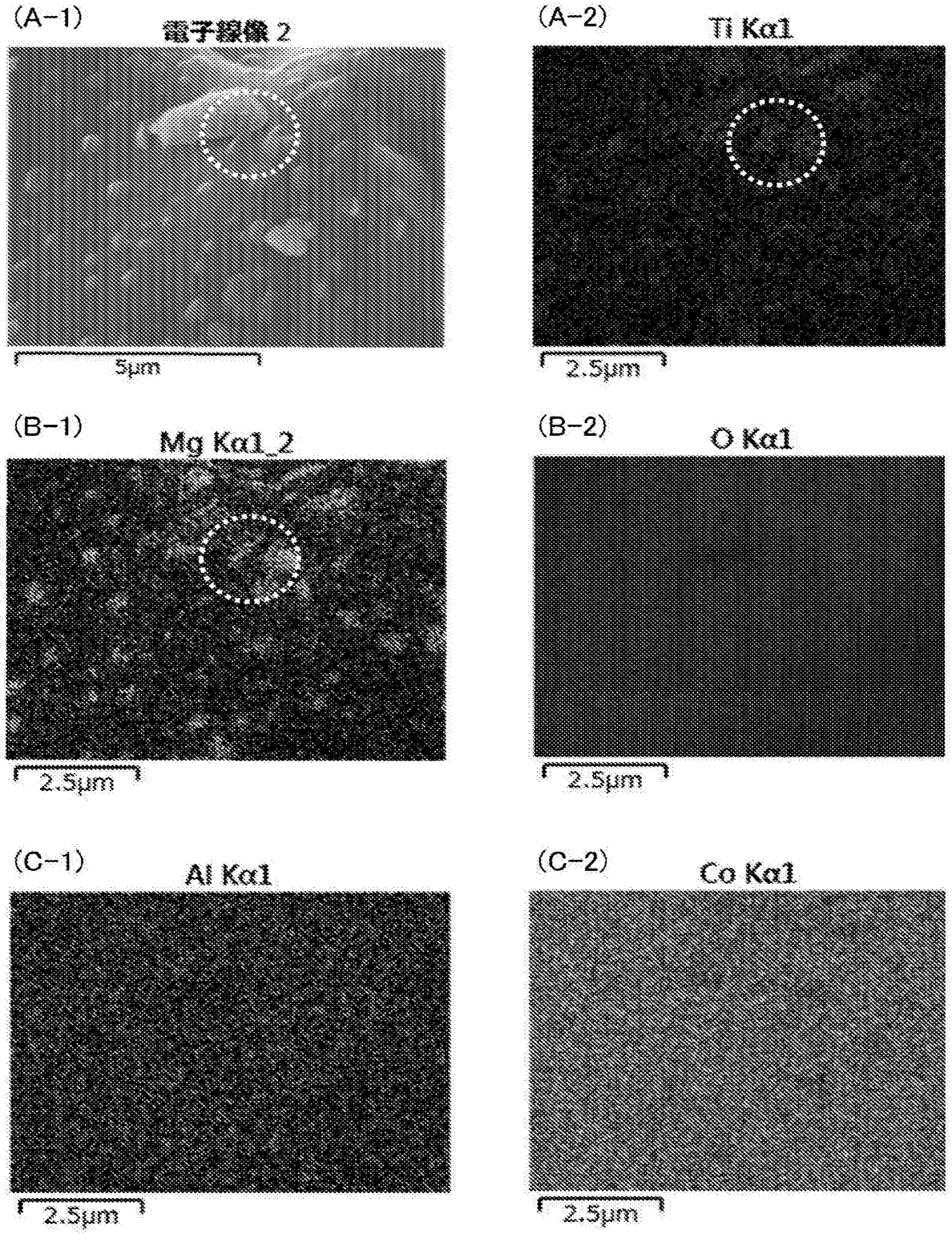
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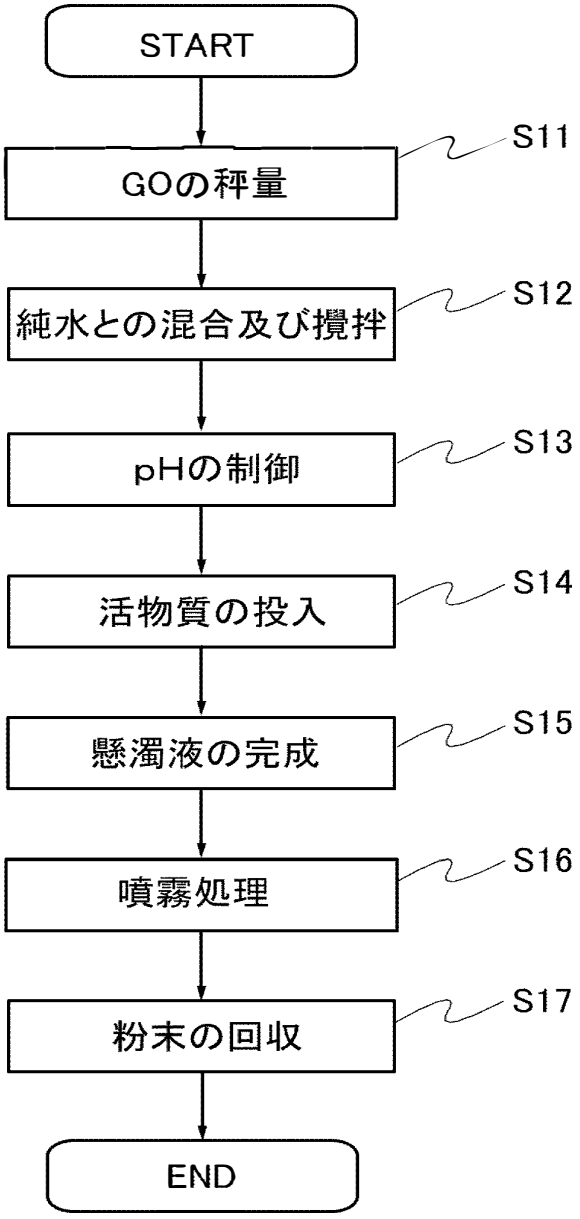
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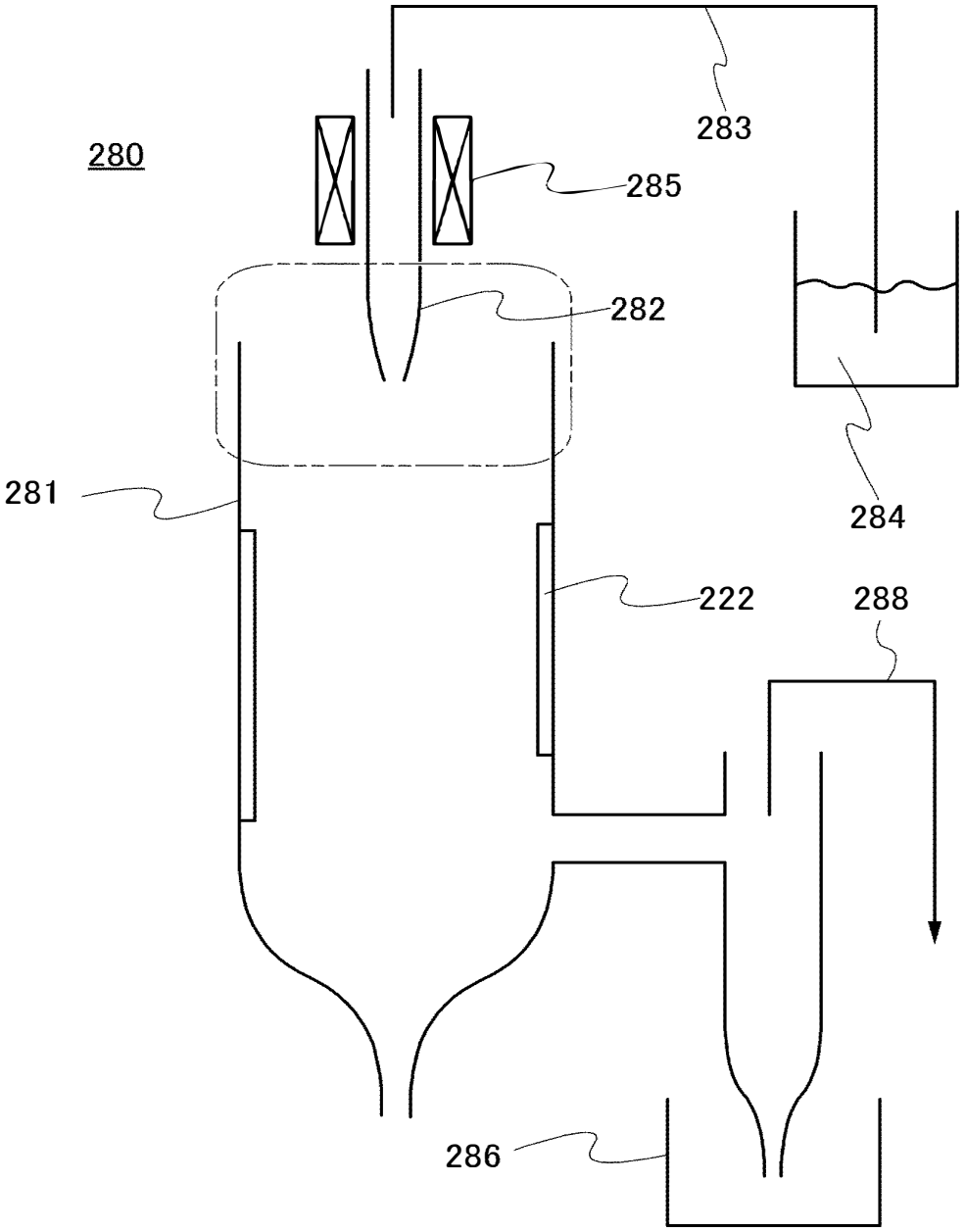
【図５２】


(A-1)
電子線像 2
5μm
(A-2)
Ti Kα1
2.5μm
(B-1)
Mg Kα1_2
2.5μm
(B-2)
O Kα1
2.5μm
(C-1)
Al Kα1
2.5μm
(C-2)
Co Kα1
2.5μm

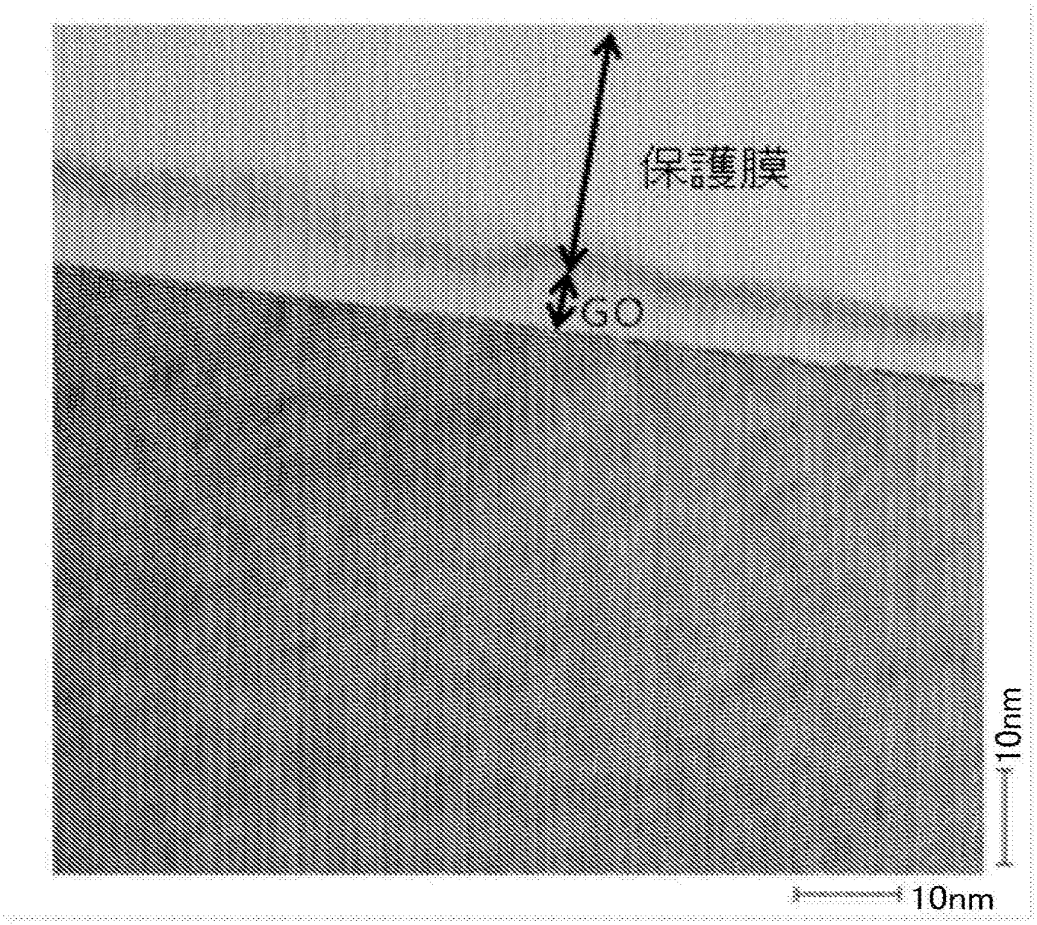
【図５３】


START
GOの秤量
S11
純水との混合及び攪拌
S12
pHの制御
S13
活物質の投入
S14
懸濁液の完成
S15
噴霧処理
S16
粉末の回収
S17
END

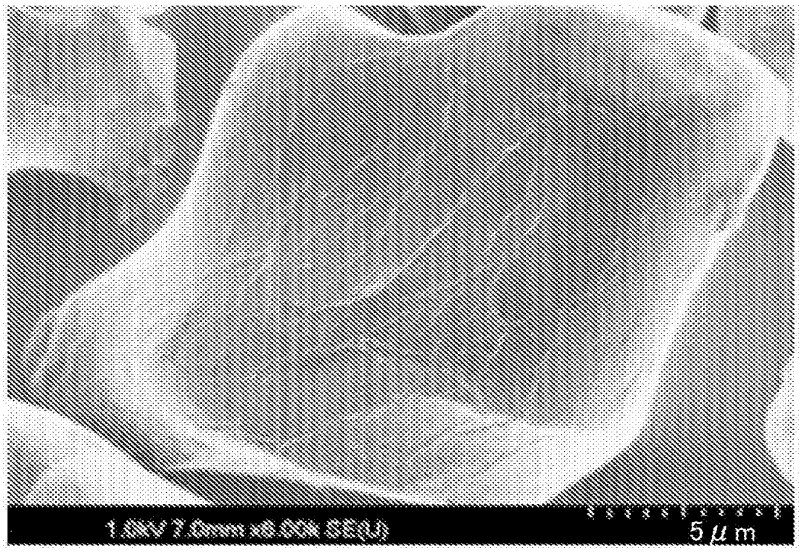
【図５４】



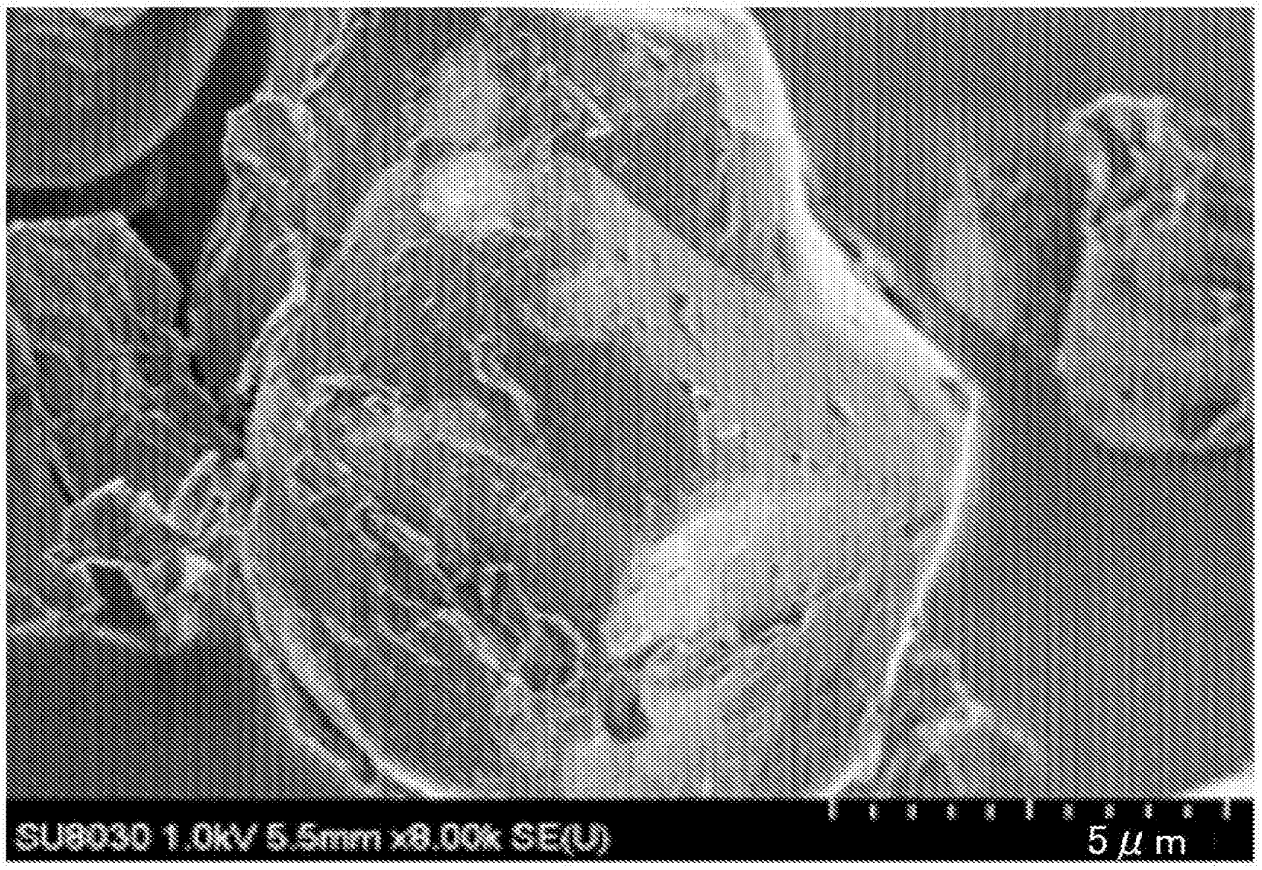
【図５５】



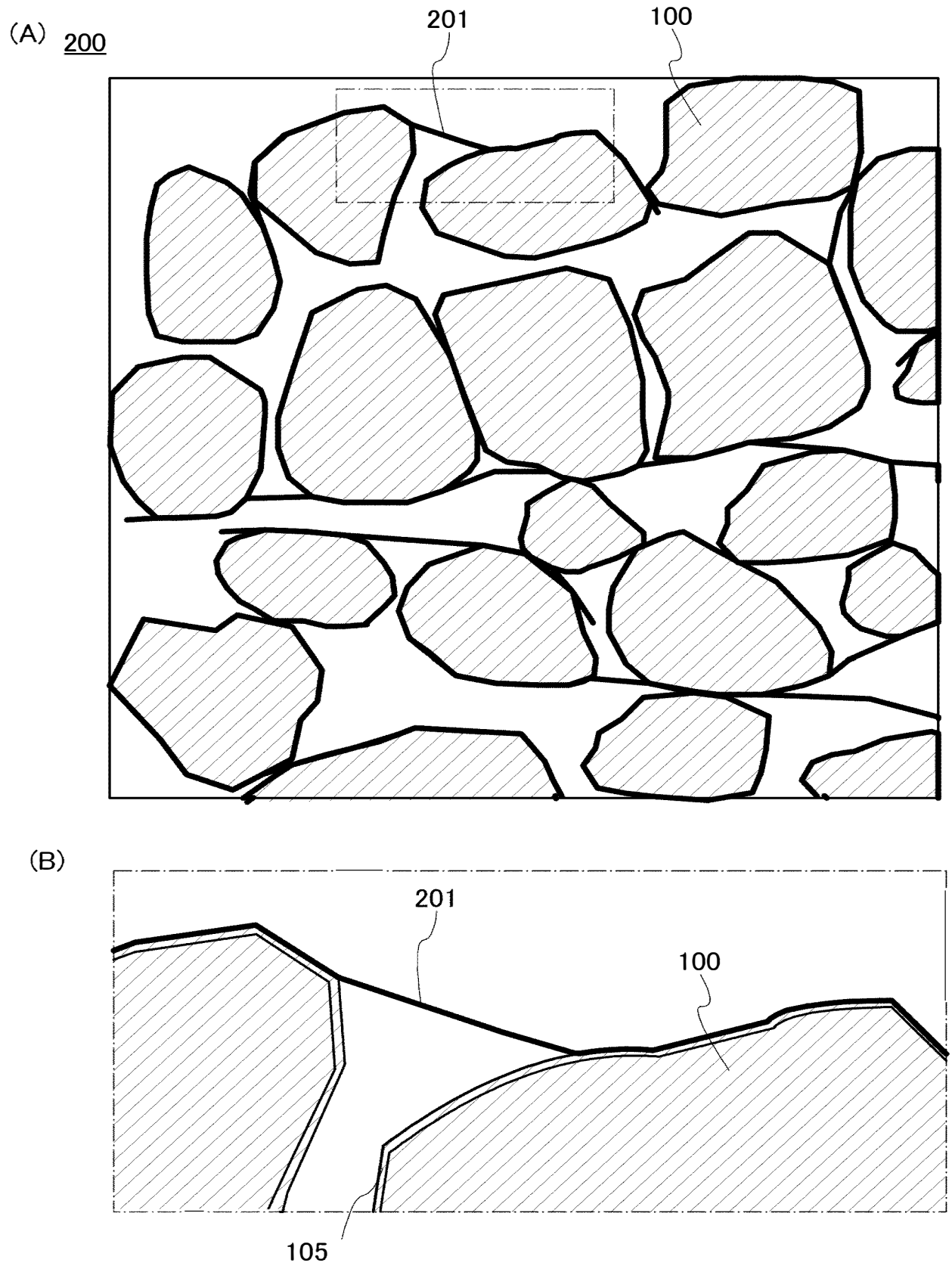
【図５６】



【図５７】



【図５８】



出願人履歴

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株式会社半導体エネルギー研究所

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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-8,927,148	01-06-2015	Kawakami.T	
		US-2010/0129714	05-27-2010	Toyama.T et al.	
		US-8,003,256	08-23-2011	Ohishi.Y	
		US-9,227,850	01-05-2016	Ooishi.Y	
		US-7,138,209	11-21-2006	Kweon.H et al.	
		US-6,846,592	01-25-2005	Kweon.H et al.	
		US-7,709,148	05-04-2010	Kawasato.T et al.	
		US-2016/0006032	01-07-2016	Paulsen.J et al.	
		US-5,834,139	11-10-1998	Shodai.T et al.	
		US-2010/0233542	09-16-2010	Endo.D et al.	
		US-2009/0220862	09-03-2009	Toyama.T et al.	
		US-7,393,476	07-01-2008	Shiozaki.R et al.	
		US-2004/0234857	11-25-2004	Shiozaki.R et al.	
		US-2007/0160906	07-12-2007	Tooyama.T et al.	
		US-2007/0212609	09-13-2007	Iwami.Y	
		US-2011/0229757	09-22-2011	Kawakami.T et al.	
		US-2009/0087731	04-02-2009	Fukui.A et al.	
		US-2007/0026315	02-01-2007	Lampe-Onnerud.C et al.	
		US-2006/0121352	06-08-2006	Kejha.J et al.	

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		JP-2003-331824A	11-21-2003			Abst.
		JP-2004-196604A	07-15-2004			Abst.
		JP-2012-074366A	04-12-2012			Abst.
		JP-2015-201432A	11-12-2015			Abst.
		JP-3172388	06-04-2001			Abst.
		JP-2005-225734A	08-25-2005			Abst.

Examiner Signature		Date Considered	

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Sheet 2 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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		US-2009/0123813	05-14-2009	Chiang.Y et al.	
		US-5,910,382	06-08-1999	Goodenough.J et al.	
		US-6,514,640	02-04-2003	Armand.M et al.	
		US-2002/0195591	12-26-2002	Ravet.N et al.	
		US-6,085,015	07-04-2000	Armand.M et al.	
		US-4,668,595	05-26-1987	Yoshino.A et al.	
		US-4,302,518	11-24-1981	Goodenough.J et al.	
		US-5,783,333	07-21-1998	Mayer.S	
		US-2001/0010807	08-02-2001	Matsubara.Y	
		US-2006/0051671	03-09-2006	Thackeray.M et al.	
		US-5,871,866	02-16-1999	Barker.J et al.	
		US-5,443,929	08-22-1995	Yamamoto.Y et al.	
		US-8,557,440	10-15-2013	Yu.D et al.	
		US-5,240,794	08-31-1993	Thackeray.M et al.	
		US-2006/0051673	03-09-2006	Johnson.C et al.	
		US-2009/0123842	05-14-2009	Thackeray.M et al.	
		US-2010/0143784	06-10-2010	Johnson.C et al.	
		US-7,303,840	12-04-2007	Thackeray.M et al.	
		US-7,635,536	12-22-2009	Johnson.C et al.	

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		JP-3959333	08-15-2007			Abst.
		JP-11-025983A	01-29-1999			Full
		JP-08-037007A	02-06-1996			Abst.
		JP-2008-166156A	07-17-2008			Abst.
		JP-2012-084257A	04-26-2012			Abst.
		CN-101148263A	03-26-2008			Abst.

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 3 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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		US-7,790,308	09-07-2010	Johnson.C et al.	
		US-8,080,340	12-20-2011	Thackeray.M et al.	
		US-2014/0332715	11-13-2014	Kawakami.T et al.	
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		US-2003/0134186	07-17-2003	Shizuki.T	
		US-2006/0286459	12-21-2006	Zhao.J et al.	
		US-2010/0035147	02-11-2010	Kotato.M et al.	
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		US-2013/0017435	01-17-2013	Sato.H et al.	
		US-2015/0014605	01-15-2015	Kawakami.T et al.	
		US-2015/0155556	06-04-2015	Kawakami.T et al.	
		US-2010/0216024	08-26-2010	Kanno.R et al.	
		US-6,680,143	01-20-2004	Thackeray.M et al.	
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		US-2013/0156683	06-20-2013	Holzapfel.M et al.	

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		JP-2013-062082A	04-04-2013			Full
		JP-2012-066944A	04-05-2012			Full
		JP-2013-012410A	01-17-2013			Abst.
		JP-2005-332629A	12-02-2005			Abst.
		JP-2015-082374A	04-27-2015			Abst.
		JP-2013-091581A	05-16-2013			Abst.

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 4 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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		US-2010/0143799	06-10-2010	Park.K	
		US-2013/0183579	07-18-2013	Kim.S et al.	
		US-2013/0130103	05-23-2013	Kim.M et al.	
		US-2012/0118149	05-17-2012	Dabrowski.B et al.	
		US-6,582,814	06-24-2003	Swiler.D et al.	
		US-5,824,278	10-20-1998	Yao.T	
		US-2015/0099178	04-09-2015	Kawakami.T et al.	
		US-6,458,487	10-01-2002	Takeuchi.H et al.	
		US-2015/0099179	04-09-2015	Ikenuma.T et al.	
		US-2016/0087315	03-24-2016	Oyama.Y	
		US-2013/0316237	11-28-2013	Miki.N	
		US-2007/0122712	05-31-2007	Kang.Y et al.	
		US-2007/0099086	05-03-2007	Kang.Y et al.	
		US-2013/0189585	07-25-2013	Kang.Y et al.	
		US-2015/0325855	11-12-2015	Kawakami.T et al.	
		US-2010/0159330	06-24-2010	Sugiura.R et al.	
		US-2013/0177806	07-11-2013	Caldwell.M et al.	
		US-2002/0164156	11-07-2002	Bilbrey.B	
		US-2012/0328951	12-27-2012	Hirohashi.T et al.	

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		JP-2012-169217A	09-06-2012			Abst.
		JP-2006-318928A	11-24-2006			Abst.
		JP-2006-318929A	11-24-2006			Abst.
		JP-2009-179501A	08-13-2009			Abst.
		JP-2013-100197A	05-23-2013			Abst.
		JP-2013-246936A	12-09-2013			Abst.

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Sheet 5 of 10

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		US-2014/0023920	01-23-2014	Yamazaki.S et al.	
		US-8,470,477	06-25-2013	Miwa.T et al.	
		US-2013/0337320	12-19-2013	Yukawa.M	
		US-2012/0256337	10-11-2012	Yokoyama.S et al.	
		US-2012/0258358	10-11-2012	Yura.Y et al.	
		US-2012/0258365	10-11-2012	Yokoyama.S et al.	
		US-2012/0258369	10-11-2012	Yokoyama.S et al.	
		US-2016/0118658	04-28-2016	Kawakami.T 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]
		WO-2012/132387	10-04-2012			Abst.
		WO-2011/141486	11-17-2011			Abst.
		WO-2012/124242	09-20-2012			Abst.

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 6 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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-2016/0118646	04-28-2016	Ikenuma.T	
		US-2012/0328956	12-27-2012	Oguni.T et al.	
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		US-8,685,569	04-01-2014	Oguni.T et al.	
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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 7 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

U. S. PATENT DOCUMENTS

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Sheet 8 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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		US-2009/0011335	01-08-2009	Takeda.K et al.	
		US-8,945,772	02-03-2015	Kawakami.T et al.	
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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	Anna Basia Ridley
Sheet	9	of	10	Attorney Docket Number	740756-004563

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 10 of 10

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Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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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EFS ID:	30516909
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/Tanika LaFortune
Filer Authorized By:	Jeffrey Costellia
Attorney Docket Number:	740756-004563
Receipt Date:	02-OCT-2017
Filing Date:	30-JUN-2017
Time Stamp:	16:39:58
Application Type:	Utility under 35 USC 111(a)

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Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Foreign Reference	JP2003-331824.pdf	703891 a6c0fa17856d035e4f1ae0808f98ee65fc7a886c	no	13

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2	Foreign Reference	JP2004-196604.pdf	642669 1bf9aa1dbc1ba070b95bdf41f4583942493e8fbf	no	16
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3	Foreign Reference	JP2012-074366.pdf	3602488 6b057029d7f9690e24c4240fdf8708d31ba0 27c2	no	38
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4	Foreign Reference	JP2015-201432.pdf	2111030 342e225d7f9a4232c313c5e841df31dfc7887be1	no	28
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5	Foreign Reference	JP3172388.pdf	514317 0da74940b9db8452a8c72ba5a591459c709 a32ca	no	7
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14	Foreign Reference	JP2013-012410.pdf	758010 fae1f4136c6721526eb6a2ed337a8c9cab8903c1	no	20
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15	Foreign Reference	JP2005-332629.pdf	625325 9585b9c815be9ab2b093f037f5fc6b6753878d6a	no	13
Warnings:					
Information:					

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Warnings:					
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Warnings:					
Information:					

23	Foreign Reference	WO2011141486A1.pdf	1262949 51173c3b5e5223344d0f4cf41d95c4623542a54c	no	25
Warnings:					
Information:					
24	Foreign Reference	WO2012124242A1.pdf	1568870 bc23cec59a3aa4bd6dc1ecff9fff839b8ea36c65	no	39
Warnings:					
Information:					
25	Non Patent Literature	JCrystalGrowth_275_283.pdf	258702 401fc44e7bdc193cf001117940e04aad0bca04b1	no	6
Warnings:					
Information:					
26	Non Patent Literature	JPowerSources_328_161.pdf	588306 dd3f40678c1a192f7b83b6436ffe0920f735f8f4	no	6
Warnings:					
Information:					
27	Non Patent Literature	JMaterChem_17_3112.pdf	1919727 dd4b68e2521d08296b69622ee45db874bb3ec384	no	14
Warnings:					
Information:					
28	Non Patent Literature	JOURNAL_OF_ANALYTICAL_AND_APPLIED_PYROLYSIS_62_45.pdf	949574 db8fbdfb0434985267c851912c31baff588a063f	no	18
Warnings:					
Information:					
29	Non Patent Literature	ElectrochemistryCom_7ElectrochemistryCom_7_528.pdf	748121 41788b5a6f8cd612359a070ce002e8544b51f2fd	no	9
Warnings:					
Information:					

30	Non Patent Literature	JElectronicMaterials_42_6_1275.pdf	771533 d8f133d417698a0592e6c1777d716c7dc17ba997	no	7
Warnings:					
Information:					
31	Non Patent Literature	JPhysCondensMatter_24_456004.pdf	790663 0d36aa074ffef1530c3c2cefb3a7bfa24bef2c92	no	9
Warnings:					
Information:					
32	Non Patent Literature	JSolStaChem_184_1096.pdf	2529472 27c2b1f12ff5867871a16d19dace0c99db7b0a9a	no	9
Warnings:					
Information:					
33	Non Patent Literature	MICRON_31_571.pdf	1825015 1670808fcb84195625a789a98882bbd9245a51b9	no	10
Warnings:					
Information:					
34	Non Patent Literature	ULTRAMICROSCOPY_116_24.pdf	789998 e9e7d9f5149bfe315eef6e3c25aac091cf449e7b	no	10
Warnings:					
Information:					
35	Non Patent Literature	Solid_State_Ionics_242_1.pdf	1005565 05bef29f86b1b03f1a13405ec8038a3c50adba44	no	9
Warnings:					
Information:					
36	Non Patent Literature	ChemSocRev_39_228.pdf	1344097 aff67f8a38a5963ab31636cba8189887f6b23a6c	no	13
Warnings:					
Information:					

37	Transmittal Letter	740756-4563_IDS_Transmittal.pdf	69787 a8be6ae7c3a884e9db61d3a5bde83757ca0ae1bd	no	1
Warnings:					
Information:					
38	Foreign Reference	JP2009-179501.pdf	1264052 e17e94a5ca5a2ee32e5192a81f68d5eaeea6fca5	no	37
Warnings:					
Information:					
39	Foreign Reference	JPH08-037007.pdf	585594 8a5c09a52214a4d3c5711896e0c65f7e20ee3ea1	no	11
Warnings:					
Information:					
40	Foreign Reference	JP2006-318929.pdf	795706 d133fecf37b2015ae5fea15bd28786047 1f01060	no	17
Warnings:					
Information:					
41	Foreign Reference	JPH11-025983.pdf	4847086 0cdfbc7056b35017f691cf45233b77fbb886f7be	no	23
Warnings:					
Information:					
42	Information Disclosure Statement (IDS) Form (SB08)	740756-4563_Form1449.pdf	274117 628c83f880912f7c51e2efb990009b315bd7da5e	no	10
Warnings:					
Information:					
This is not an USPTO supplied IDS fillable form					
		Total Files Size (in bytes):	74229299		

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.

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: Anna Basia Ridley
Filed: June 30, 2017	)	Confirmation No.: 1074
For: POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY	)	

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.

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 within three months of filing, or before the mailing of a first Office action on the merits, whichever is later; 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

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

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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 4

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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-2012/0034516	02-09-2012	Koo.C et al.	
		US-2008/0131780	06-05-2008	Kawasato.T et al.	
		US-2011/0033749	02-10-2011	Uchida.M 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-2012-508444	04-05-2012			Abst.
		WO-2010/053328	05-14-2010			Abst.
		EP-2352190A	08-03-2011			Eng.
		KR-2010-0052419A	05-19-2010			Abst.
		CN-102210045A	10-05-2011			Abst.
		JP-2011-181527A	09-15-2011			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.*

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

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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 2 of 4

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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]
		WO-2007/052712	05-10-2007			Abst.
		KR-2008-0024105A	03-17-2008			Abst.
		CN-101208269A	06-25-2008			Abst.
		KR-10-0944137	02-24-2010			Abst.
		WO-2009/057722	05-07-2009			Abst.
		KR-2009-0086440A	08-12-2009			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.**

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PTO/SB/08A (07-06)
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U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 3 of 4

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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]
		CN-101675547A	03-17-2010			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.

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U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 4 of 4

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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]
		INTERNATIONAL SEARCH REPORT (APPLICATION NO. PCT/IB2017/053896) DATED October 3, 2017.	Eng.
		WRITTEN OPINION (APPLICATION NO. PCT/IB2017/053896) DATED October 3, 2017.	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 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, 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 (1-800-786-9199)** and select option 2.*

PATENT COOPERATION TREATY
PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference PCT24652-813	**FOR FURTHER ACTION**	see Form PCT/ISA/220 as well as, where applicable, item 5 below.
International application No. PCT/IB2017/053896	International filing date *(day/month/year)* 29.06.2017	(Earliest) Priority Date *(day/month/year)* 05.07.2016
Applicant SEMICONDUCTOR ENERGY LABORATORY CO., LTD.		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

a. With regard to the **language**, the international search was carried out on the basis of:

☑ the international application in the language in which it was filed.

☐ a translation of the international application into _____ which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b)).

b. ☐ This international search report has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43.6*bis*(a)).

c. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.

2. ☐ **Certain claims were found unsearchable** (see Box No. II).

3. ☐ **Unity of invention is lacking** (see Box No. III).

4. With regard to the **title**,

☑ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☑ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2, by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. With regard to the **drawings**,

a. the figure of the **drawings** to be published with the abstract is Figure No. 3

☑ as suggested by the applicant.

☐ as selected by this Authority, because the applicant failed to suggest a figure.

☐ as selected by this Authority, because this figure better characterizes the invention.

b. ☐ none of the figures is to be published with the abstract.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/IB2017/053896

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl. H01M4/525(2010.01)i, H01M4/36(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl. H01M4/525, H01M4/36

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Published examined utility model applications of Japan 1922-1996
Published unexamined utility model applications of Japan 1971-2017
Registered utility model specifications of Japan 1996-2017
Published registered utility model applications of Japan 1994-2017

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 2015-201432 A (TODA KOGYO CORP.) 2015.11.12, CLAIMS 1-10, EXAMPLES 1-10 (FAMILY: NONE)	1-22
A	US 2012/0034516 A1 (LG CHEM, LTD.) 2012.02.09, CLAIMS 1-18, EXAMPLES 1-16 & JP 2012-508444 A & WO 2010/053328 A2 & EP 2352190 A2 & KR 10-2010-52419 A & CN 102210045 A	1-22
A	US 2008/0131780 A1 (AGC SEIMI CHEMICAL CO., LTD.) 2008.06.05, CLAIMS 1-19, EXAMPLES 1-10 & JP 2011-181527 A & WO 2007/052712 A1 & KR 10-2008-24105 A & CN 101208269 A & KR 10-944137 B1	1-22

☑ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
21.09.2017	03.10.2017
Name and mailing address of the ISA/JP **Japan Patent Office** 3-4-3, Kasumigaseki, Chiyoda-ku, Tokyo 100-8915, Japan	Authorized officer SHO, Tomoaki Telephone No. +81-3-3581-1101 Ext. 3477

4X 3648

AMPEREX Ex. 1002
Page 747

INTERNATIONAL SEARCH REPORT

International application No.
PCT/IB2017/053896

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2011/0033749 A1 (AGC SEIMI CHEMICAL CO., LTD.) 2011.02.10, CLAIMS 1-12, EXAMPLES 1-12 & WO 2009/057722 A1 & KR 10-2009-86440 A & CN 101675547 A	1-22

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:
SEMICONDUCTOR ENERGY LABORATORY CO., LTD.

398, Hase Atsugi-shi, Kanagawa
2430036 JP

PCT

WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY
(PCT Rule 43*bis*.1)

Date of mailing *(day/month/year)*	03.10.2017

Applicant's or agent's file reference	**FOR FURTHER ACTION**
PCT24652-813	See paragraph 2 below

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/IB2017/053896	29.06.2017	05.07.2016

International Patent Classification (IPC) or both national classification and IPC
Int.Cl. H01M4/525(2010.01)i, H01M4/36(2006.01)i

Applicant
SEMICONDUCTOR ENERGY LABORATORY CO., LTD.

1. This opinion contains indications relating to the following items:

- [x] Box No. I Basis of the opinion
- [ ] Box No. II Priority
- [ ] Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- [ ] Box No. IV Lack of unity of invention
- [x] Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- [ ] Box No. VI Certain documents cited
- [ ] Box No. VII Certain defects in the international application
- [ ] Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Date of completion of this opinion	21.09.2017	
Name and mailing address of the ISA/JP	Authorized officer	4X 3648
Japan Patent Office 3-4-3, Kasumigaseki, Chiyoda-ku, Tokyo 100-8915, Japan	SHO, Tomoaki Telephone No. +81-3-3581-1101 Ext. 3477	

Form PCT/ISA/237 (cover sheet) (January 2015)

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/IB2017/053896

Box No. I Basis of this opinion

1. With regard to the **language**, this opinion has been established on the basis of:

   ☑ the international application in the language in which it was filed.

   ☐ a translation of the international application into _____ which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b)).

2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(b)).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing:

   a. ☐ forming part of the international application as filed:

      ☐ in the form of an Annex C/ST.25 text file.

      ☐ on paper or in the form of an image file.

   b. ☐ furnished together with the international application under PCT Rule 13ter.1(a) for the purposes of international search only in the form of an Annex C/ST.25 text file.

   c. ☐ furnished subsequent to the international filing date for the purposes of international search only:

      ☐ in the form of an Annex C/ST.25 text file (Rule 13ter.1(a)).

      ☐ on paper or in the form of an image file (Rule 13ter.1(b) and Administrative Instructions, Section 713).

4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that forming part of the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

5. Additional comments:

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/IB2017/053896

Box No. V **Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Claims	1-22	YES
	Claims		NO
Inventive step (IS)	Claims	1-22	YES
	Claims		NO
Industrial applicability (IA)	Claims	1-22	YES
	Claims		NO

2. Citations and explanations:

D1: JP 2015-201432 A (TODA KOGYO CORP.) 2015.11.12, CLAIMS 1-10, EXAMPLES 1-10 (FAMILY: NONE)

D2: US 2012/0034516 A1 (LG CHEM, LTD.) 2012.02.09, CLAIMS 1-18, EXAMPLES 1-16 & JP 2012-508444 A & WO 2010/053328 A2 & EP 2352190 A2 & KR 10-2010-52419 A & CN 102210045 A

D3: US 2008/0131780 A1 (AGC SEIMI CHEMICAL CO., LTD.) 2008.06.05, CLAIMS 1-19, EXAMPLES 1-10 & JP 2011-181527 A & WO 2007/052712 A1 & KR 10-2008-24105 A & CN 101208269 A & KR 10-944137 B1

D4: US 2011/0033749 A1 (AGC SEIMI CHEMICAL CO., LTD.) 2011.02.10, CLAIMS 1-12, EXAMPLES 1-12 & WO 2009/057722 A1 & KR 10-2009-86440 A & CN 101675547 A

The subject matter of claims 1 to 8 is neither disclosed in any of the documents cited in the International Search Report (ISR) nor obvious to a person skilled in the art.

D1 discloses a positive electrode active material particle powder includes: a core particle composed of cobaltic acid lithium having a layered structure containing element M (M is Mg and/or Al); and a surface layer composed of the cobaltic acid lithium having the layered structure in which at least Ti is incorporated into solid solution.

D2, D3 and D4 teach a cathode active material including a lithium transition metal oxide based on at least one transition metal selected from a group consisting of Ni, Mn and Co. The lithium transition metal oxide of D2 contains fluorine, and most of the fluorine is present on a surface of the lithium transition metal oxide. The lithium transition metal oxide of D3 contains a composite compound containing titanium and magnesium in the vicinity of the surface of particles of the lithium transition metal oxide. The lithium transition metal oxide of D4 contains a lithium titanium composite oxide in the surface layer of particles of the lithium transition metal oxide.

(Continue to Supplemental Box)

Supplemental Box

In case the space in any of the preceding boxes is not sufficient.

Continuation of: V

None of the above documents discloses, however, the feature in the subject matter of claims 1 to 8 that a positive electrode active material comprises a first region positioned inside the material, a second region, and a third region which is positioned closer to a surface than a second region, wherein the first region comprising an oxide of lithium and a first transition metal of a layered rock-salt crystal structure, the second region comprising an oxide of a second transition metal of rock-salt crystal structure, and the third region comprising representative elements of the rock-salt crystal structure. Having the said feature, the subject matter of claims 1 to 8 achieves the advantageous effect of "the positive electrode active material with excellent cycle characteristics".

The subject matter of claims 9 to 12 is neither disclosed in any of the documents cited in the International Search Report (ISR) nor obvious to a person skilled in the art.

None of the above documents discloses the feature in the subject matter of claims 9 to 12 that the first region comprises lithium cobaltate, the second region comprises lithium titanate, and the third region comprises magnesium oxide. Having the said feature, the subject matter of claims 9 to 12 achieves the advantageous effect of "the positive electrode active material with excellent cycle characteristics".

The subject matter of claims 13 to 16 is neither disclosed in any of the documents cited in the International Search Report (ISR) nor obvious to a person skilled in the art.

D1 discloses a positive electrode active material particle comprising lithium, titanium, cobalt, magnesium, and oxygen, while D2, D3 and D4 teach a positive electrode active material particle comprising lithium, titanium, cobalt, magnesium, oxygen, and fluorine.

None of the above documents discloses, however, concentrations of constituent elements in a superficial portion of a positive electrode active material particle measured by X-ray photoelectron spectroscopy. Having the said feature, the subject matter of claims 14 to 20 achieves the advantageous effect of "the positive electrode active material with excellent cycle characteristics".

The subject matter of claims 17 to 22 is neither disclosed in any of the documents cited in the International Search Report (ISR) nor obvious to a person skilled in the art.

The subject matter claimed, which is characterized by the combination of the technical features (i.e., a method for forming a positive electrode active material including the steps of: heating mixture of the source materials to obtain particles comprising lithium, cobalt, magnesium, oxygen, and fluorine; mixing the particles into an alcohol solution of alkoxide of the second transition metal.), is not rendered obvious by the available prior art. Those features could be combined only with the exercise of the applicant's inventive skill.

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: Anna Basia Ridley
Filed: June 30, 2017	)	Confirmation No.: 1074
For: POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY	)	

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 wishes to note that the *Patent Documents*: US-2012/0034516; US-2008/0131780; US-2011/0033749; JP-2012/508444; WO-2010/053328; EP-2 352 190; KR-2010-0052419; CN-102210045; JP-2011-181527; WO-2007/052712; KR-2008-0024105; CN-101208269; KR-10-0944137; WO-2009/057722; KR-2009-0086440; and CN-101675547 listed on the attached Form PTO-1449 were previously cited by International Searching Authority (JPO) in an International Search Report and a Written Opinion (copy of each enclosed) mailed **October 3, 2017** issued in a counterpart International Application No. PCT/IB2017/053896.

Applicant also wishes to bring to the Examiner's attention the following family information listed on the attached PTO-1449.

1. JP-2012/508444; WO-2010/053328; EP-2 352 190; KR-2010-0052419 and CN-102210045 are in the **same family** as US-2012/0034516, and were also **cited in** the foreign communication noted above.

2. JP-2011-181527; WO-2007/052712; KR-2008-0024105; CN-101208269 and KR-10-0944137 are in the **same family** as US-2008/0131780, and were also **cited in** the foreign communication noted above.

3. WO-2009/057722; KR-2009-0086440 and CN-101675547 are in the **same family** as US-2011/0033749, and were also **cited in** the foreign communication noted above.

The undersigned certifies that each item of information contained in this information disclosure statement 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 within three months of filing, or before the mailing of a first Office action on the merits, whichever is later; therefore, no fee or statement is believed to be necessary at this time. Applicant, nevertheless, provides the above statement as a courtesy.

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,

By: /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

Electronic Acknowledgement Receipt

EFS ID:	30725539
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
Filer Authorized By:	
Attorney Docket Number:	740756-004563
Receipt Date:	24-OCT-2017
Filing Date:	30-JUN-2017
Time Stamp:	15:06:13
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)	740756-004563_PTO1449.pdf	174411 7aa6a6fc249044354d63e3ac9ccf287c82d90d12	no	4

Warnings:

Information:					
This is not an USPTO supplied IDS fillable form					
2	Foreign Reference	JP2012-508444.pdf	4702770 c8257912461372e9b18c7f153c941d8911b875f5	no	23
Warnings:					
Information:					
3	Foreign Reference	WO2010053328A2.pdf	534478 83eb9c4e7e3ad37f52009b5cff39ff938516a877	no	23
Warnings:					
Information:					
4	Foreign Reference	EP2352190A2.pdf	472250 7e5bcd22131de1b45de8468246c04cff879be888	no	15
Warnings:					
Information:					
5	Foreign Reference	KR1020100052419A.pdf	9836311 78d653f12e21f3c91ba95c7b3714e2aed90c661a	no	16
Warnings:					
Information:					
6	Foreign Reference	CN102210045A.pdf	499970 0e35230675adca02c14a346ff40c0bd9eb922149	no	18
Warnings:					
Information:					
7	Foreign Reference	JP2011-181527.pdf	7011578 c1dc4202b49460b513e066a0b4bcffaf34ed68e0	no	23
Warnings:					
Information:					
8	Foreign Reference	WO2007052712A1.pdf	1157529 8286393e2a3747cf9b99699d899a3f92ef661393	no	43
Warnings:					
Information:					

9	Foreign Reference	KR1020080024105A.pdf	17137986 f78bfe21e8c0b3eb35dc4e8cc99161c8625a0f51	no	20
Warnings:					
Information:					
10	Foreign Reference	CN101208269A.pdf	889404 c8fa4a098233ef2ab533baef5a9b1ba3cc91b4a1	no	26
Warnings:					
Information:					
11	Foreign Reference	KR100944137B1.pdf	17210115 c7b3ee898b545f0f4893db66d77999bf6d813691	no	20
Warnings:					
Information:					
12	Foreign Reference	WO2009057722A1.pdf	1220054 fe8a980ece9adb3956a792bbf02523dd87244aa8	no	46
Warnings:					
Information:					
13	Foreign Reference	KR1020090086440A.pdf	19590396 c9b3a8db6325c456465256c315af59ad0f834e4d	no	22
Warnings:					
Information:					
14	Foreign Reference	CN101675547A.pdf	1033115 d6f6e16d174f7c061e11cceb0cfc24f79bc6fc4b	no	35
Warnings:					
Information:					
15	Non Patent Literature	171003_INTERNATIONALSEARCHREPORT_PCT24652-26813.pdf	69373 b7c16e2da3440224ba10ccd08dd9d09349eece4a	no	3
Warnings:					
Information:					

16	Non Patent Literature	171003_WRITTENOPINION_PCT 24652-26813.pdf	117767 88906843b9c5ef3f95d12591637a6799df089ac0	no	4
Warnings:					
Information:					
17	Transmittal Letter	740756-004563_IDS_Transmittal.pdf	140308 069f8102e953c9b417ad55bec3a1d715f852aadd	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	81797815		

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/AIA/80 (07-12)
Approved for use through 11/30/2014. OMB 0651-0035
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.

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: 22204

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 assigned only to the undersigned according to the USPTO assignment records or assignments documents 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: 22204

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		Date	09/21/2012
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.

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

PTO/AIA/96 (08-12)
Approved for use through 01/31/2013. 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 displays a valid OMB control number.

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 options 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 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 _____, Frame _____, 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.

PTO/AIA/96 (08-12)
Approved for use through 01/31/2013. 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 displays a valid OMB control number.

STATEMENT UNDER 37 CFR 3.73(c)

3. 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.

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.

/Jeffrey L. Costellia, Reg. No. 35,483/	November 20, 2017
Signature	Date
Jeffrey L. Costelllia, Attorney of Record	35,483
Printed or Typed Name	Title or Registration Number

NIXON PEABODYLLP
United States Patent Rights

Attorney Docket No. 740756-004563

ASSIGNMENT

Serial No. 15/638,449 Filed June 30, 2017

WHEREAS, Teruaki OCHIAI, Takahiro KAWAKAMI, Mayumi MIKAMI, Yohei MOMMA, Masahiro TAKAHASHI and Ayae TSURUTA (hereinafter designated as the undersigned) has (have) invented certain new and useful improvements in POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY for which an application for Letters Patent of the United States of America has been executed by the undersigned on _____, and;

WHEREAS, Semiconductor Energy Laboratory Co., Ltd. of 398, Hase, Atsugi-shi, Kanagawa-ken, 243-0036 Japan and its heirs, successors, legal representatives and assigns (hereinafter designated as the Assignee) is desirous of acquiring the entire right, title and interest in and to said invention and in and to any Letters Patent(s) that may be granted therefor in the United States of America;

NOW, THEREFORE, in consideration of the sum of One Dollar ($1.00) to the undersigned in hand paid, the receipt of which is hereby acknowledged, and other good and valuable consideration, the undersigned has (have) sold, assigned and transferred, and by these presents do sell, assign and transfer unto said Assignee the full and exclusive right to the said invention in the United States of America and its territories and for all foreign countries, dependencies and possessions and the entire right, title and interest in and to any and all Letters Patent(s) which may be granted therefor in the United States of America and its territories, dependencies and possessions, and in and to any and all divisions, reissues, continuations and extensions thereof for the full term or terms for which the same may be granted.

The undersigned agree(s) to execute all papers necessary in connection with this application and any continuing, divisional or reissue applications thereof and also to execute separate assignments in connection with such applications as the Assignee may deem necessary or expedient.

The undersigned agree(s) to execute all papers necessary in connection with any interference which may be declared concerning this application or any continuation, division or reissue thereof or Letters Patent(s) or reissue patent issued thereon and to cooperate with the Assignee in every way possible in obtaining and producing evidence and proceeding with such interference.

The undersigned agree(s) to execute all papers and documents and to perform any act which may be necessary in connection with claims under or provisions of the International Convention for the Protection of Industrial Property or similar agreements.

The undersigned agree(s) to perform all affirmative acts which may be necessary to obtain a grant of a valid United States patent(s) to the Assignee and to vest all rights therein hereby conveyed to said Assignee as fully and entirely as the same would have been held by

the undersigned if this Assignment and sale had not been made.

The undersigned hereby authorize(s) and request(s) the Commissioner of Patents and Trademarks to issue any and all Letters Patents of the United States of America resulting from said application or any division or divisions or continuing or reissue applications thereof to the said Assignee, as Assignee of the entire interest, and hereby convenants that he has (they have) the full right to convey the entire interest herein assigned, and that he has (they have) not executed, and will not execute, any agreement in conflict herewith.

The undersigned hereby grant(s) the law firm of Nixon Peabody LLP the power to insert on this Assignment any further identification which may be necessary or desirable in order to comply with the rules of the U.S. Patent and Trademark Office for recordation of this document.

In witness thereof, this Assignment has been executed by the undersigned on the date(s) opposite the undersigned name(s).

Date 07/19/2017, Name of Inventor Teruaki Ochiai (SEAL)
Teruaki OCHIAI

Date _____, Name of Inventor _____ (SEAL)
Takahiro KAWAKAMI

Date 07/19/2017, Name of Inventor Mayumi Mikami (SEAL)
Mayumi MIKAMI

Date 07/20/2017, Name of Inventor Yohei Momma (SEAL)
Yohei MOMMA

Date 07/19/2017, Name of Inventor Masahiro Takahashi (SEAL)
Masahiro TAKAHASHI

Date 07/14/2017, Name of Inventor Ayae TSURUTA (SEAL)
Ayae TSURUTA

(This assignment should preferably be acknowledged before a United States Consul or Notary Public. If not, then the execution by the Inventor(s) should be witnessed has (have) invented certain new and useful improvements in by at least two other persons who should

sign here.)

Witness
(name) (signature)

Witness
(name) (signature)

Witness
(name) (signature)

NIXON PEABODYLLP
United States Patent Rights

Attorney Docket No. 740756-004563

ASSIGNMENT

Serial No. 15/638,449 Filed June 30, 2017

WHEREAS, Teruaki OCHIAI, Takahiro KAWAKAMI, Mayumi MIKAMI, Yohei MOMMA, Masahiro TAKAHASHI and Ayae TSURUTA
(hereinafter designated as the undersigned) has (have) invented certain new and useful improvements in POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY for which an application for Letters Patent of the United States of America has been executed by the undersigned on _____, and;

WHEREAS, Semiconductor Energy Laboratory Co., Ltd. of 398, Hase, Atsugi-shi, Kanagawa-ken, 243-0036 Japan and its heirs, successors, legal representatives and assigns (hereinafter designated as the Assignee) is desirous of acquiring the entire right, title and interest in and to said invention and in and to any Letters Patent(s) that may be granted therefor in the United States of America;

NOW, THEREFORE, in consideration of the sum of One Dollar ($1.00) to the undersigned in hand paid, the receipt of which is hereby acknowledged, and other good and valuable consideration, the undersigned has (have) sold, assigned and transferred, and by these presents do sell, assign and transfer unto said Assignee the full and exclusive right to the said invention in the United States of America and its territories and for all foreign countries, dependencies and possessions and the entire right, title and interest in and to any and all Letters Patent(s) which may be granted therefor in the United States of America and its territories, dependencies and possessions, and in and to any and all divisions, reissues, continuations and extensions thereof for the full term or terms for which the same may be granted.

The undersigned agree(s) to execute all papers necessary in connection with this application and any continuing, divisional or reissue applications thereof and also to execute separate assignments in connection with such applications as the Assignee may deem necessary or expedient.

The undersigned agree(s) to execute all papers necessary in connection with any interference which may be declared concerning this application or any continuation, division or reissue thereof or Letters Patent(s) or reissue patent issued thereon and to cooperate with the Assignee in every way possible in obtaining and producing evidence and proceeding with such interference.

The undersigned agree(s) to execute all papers and documents and to perform any act which may be necessary in connection with claims under or provisions of the International Convention for the Protection of Industrial Property or similar agreements.

The undersigned agree(s) to perform all affirmative acts which may be necessary to obtain a grant of a valid United States patent(s) to the Assignee and to vest all rights therein hereby conveyed to said Assignee as fully and entirely as the same would have been held by

the undersigned if this Assignment and sale had not been made.

The undersigned hereby authorize(s) and request(s) the Commissioner of Patents and Trademarks to issue any and all Letters Patents of the United States of America resulting from said application or any division or divisions or continuing or reissue applications thereof to the said Assignee, as Assignee of the entire interest, and hereby convenants that he has (they have) the full right to convey the entire interest herein assigned, and that he has (they have) not executed, and will not execute, any agreement in conflict herewith.

The undersigned hereby grant(s) the law firm of Nixon Peabody LLP the power to insert on this Assignment any further identification which may be necessary or desirable in order to comply with the rules of the U.S. Patent and Trademark Office for recordation of this document.

In witness thereof, this Assignment has been executed by the undersigned on the date(s) opposite the undersigned name(s).

Date_____, Name of Inventor_____ (SEAL)
Teruaki OCHIAI

Date 09/08/2017 Name of Inventor Takahiro Kawakami (SEAL)
Takahiro KAWAKAMI

Date_____, Name of Inventor_____ (SEAL)
Mayumi MIKAMI

Date_____, Name of Inventor_____ (SEAL)
Yohei MOMMA

Date_____, Name of Inventor_____ (SEAL)
Masahiro TAKAHASHI

Date_____, Name of Inventor_____ (SEAL)
Ayae TSURUTA

(This assignment should preferably be acknowledged before a United States Consul or Notary Public. If not, then the execution by the Inventor(s) should be witnessed has (have) invented certain new and useful improvements in by at least two other persons who should

sign here.)

Witness

(name) (signature)

Witness

(name) (signature)

Witness

(name) (signature)

Electronic Acknowledgement Receipt

EFS ID:	30995932
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/KEYONNA PITTMAN
Filer Authorized By:	Jeffrey Costellia
Attorney Docket Number:	740756-004563
Receipt Date:	20-NOV-2017
Filing Date:	30-JUN-2017
Time Stamp:	15:47:12
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	Oath or Declaration filed	740756-4563_Declaration.pdf	1542370 5cb588300d1939ddca4afa06b271fedd9fbe f132	no	12

Warnings:

Information:					
2		740756-4563_Power_of_Attorney.pdf	1031103 de8080ea8fb6725e024eb869c7eab78b7c0fbdcd	yes	9

Multipart Description/PDF files in .zip description		
Document Description	**Start**	**End**
Power of Attorney	1	1
Assignee showing of ownership per 37 CFR 3.73	2	9

Warnings:	
Information:	
Total Files Size (in bytes):	2573473

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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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DECLARATION (37 CFR 1.63) FOR UTILITY OR DESIGN APPLICATION USING AN APPLICATION DATA SHEET (37 CFR 1.76)

発明の名称 Title of Invention	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY

下記発明者である私は、つぎのことがらを宣誓します。
As the below named inventor, I hereby declare that:

本宣誓は
This declaration is directed to:

☐ 添付されている、あるいは
The attached application, or

■ 米国出願、あるいは_____に出願されたPCT国際願番号_____として出願されているものに宛てられています。
United States application or PCT international application number 15/638,449 filed on June 30, 2017.

上記の出願は私自身、あるいは私が権限を授与したものによって行われたものです。
The above-identified application was made or authorized to be made by me.

私は本出願書中にあらわれるもともとの発明者、あるいはもともとの共同発明者です。
I believe that I am the original inventor or an original joint inventor of a claimed invention in the application.

本宣誓書において故意に虚偽の申し立てを行った場合は 18 U.S.C. 1001 により、罰金あるいは最高五(5)年の禁固刑、あるいはその両方による罰則の対象となることを認めます。
I hereby acknowledge that any willful false statement made in this declaration is punishable under 18 U.S.C. 1001 by fine or imprisonment of not more than five (5) years, or both.

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発明者の正式氏名
LEGAL NAME OF INVENTOR

発明者:
Inventor: Teruaki OCHIAI

日付(任意):
Date (Optional): 07/19/2017

署名:
Signature: Teruaki Ochiai

備考: 出願データシート(PTO/AIA/14 あるいはその同等用紙) は、発明の自主独立体全体の命名を含め、本用紙に添付すること。なお残余の発明者ごとにPTO/SB/AIA01 用紙を使用する。
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本用紙に記載された情報は、下記の通常使用目的に従います。

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発明の名称 Title of Invention	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY

下記発明者である私は、つぎのことがらを宣誓します。
As the below named inventor, I hereby declare that:

本宣誓は
This declaration is directed to:

☐ 添付されている、あるいは
The attached application, or

■ 米国出願、あるいは_____に出願されたPCT国際願番号_____として出願されているものに宛てられています。
United States application or PCT international application number 15/638,449 filed on June 30, 2017.

上記の出願は私自身、あるいは私が権限を授与したものによって行われたものです。
The above-identified application was made or authorized to be made by me.

私は本出願書中にあらわれるもともとの発明者、あるいはもともとの共同発明者です。
I believe that I am the original inventor or an original joint inventor of a claimed invention in the application.

本宣誓書において故意に虚偽の申し立てを行った場合は 18 U.S.C. 1001 により、罰金あるいは最高五(5)年の禁固刑、あるいはその両方による罰則の対象となることを認めます。
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発明者の正式氏名
LEGAL NAME OF INVENTOR

発明者:
Inventor: Takahiro KAWAKAMI

日付(任意):
Date (Optional): 09/18/2017

署名:
Signature: Takahiro Kawakami

備考: 出願データシート(PTO/AIA/14 あるいはその同等用紙) は、発明の自主独立体全体の命名を含め、本用紙に添付すること。なお残余の発明者ごとに PTO/SB/AIA01 用紙を使用する。
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申請データシート(37 CFR 1.76)を使った実用及び意匠登録出願宣誓書(37 CFR 1.63)
DECLARATION (37 CFR 1.63) FOR UTILITY OR DESIGN APPLICATION USING AN APPLICATION DATA SHEET (37 CFR 1.76)

発明の名称 Title of Invention	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY

下記発明者である私は、つぎのことがらを宣誓します。
As the below named inventor, I hereby declare that:

本宣誓は
This declaration is directed to:

☐ 添付されている、あるいは
The attached application, or

■ 米国出願、あるいは_____に出願されたPCT国際願番号_____として出願されているものに宛てられています。
United States application or PCT international application number 15/638,449 filed on June 30, 2017.

上記の出願は私自身、あるいは私が権限を授与したものによって行われたものです。
The above-identified application was made or authorized to be made by me.

私は本出願書中にあらわれるもともとの発明者、あるいはもともとの共同発明者です。
I believe that I am the original inventor or an original joint inventor of a claimed invention in the application.

本宣誓書において故意に虚偽の申し立てを行った場合は 18 U.S.C. 1001 により、罰金あるいは最高五(5)年の禁固刑、あるいはその両方による罰則の対象となることを認めます。
I hereby acknowledge that any willful false statement made in this declaration is punishable under 18 U.S.C. 1001 by fine or imprisonment of not more than five (5) years, or both.

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発明者の正式氏名
LEGAL NAME OF INVENTOR

発明者:
Inventor: Mayumi MIKAMI

日付(任意):
Date (Optional): 7/19/2017

署名:
Signature: Mayumi Mikami

備考: 出願データシート(PTO/AIA/14 あるいはその同等用紙) は、発明の自主独立体全体の命名を含め、本用紙に添付すること。なお残余の発明者ごとにPTO/SB/AIA01 用紙を使用する。
Note: An application data sheet (PTO/AIA/14 or equivalent), including naming the entire inventive entity, must accompany this form. Use an additional PTO/SB/AIA01 form for each additional inventor.

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1. 本用紙に記載されている情報は、情報公開法 (5 U.S.C. 552) およびプライバシー保護法 (5 U.S.C 552a) が許容する範囲において極秘扱いとなります。本記録システムの記録は、本記録の開示が情報公開法で要求されているか否かを判断するために、司法省に開示される場合があります。
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DECLARATION (37 CFR 1.63) FOR UTILITY OR DESIGN APPLICATION USING AN APPLICATION DATA SHEET (37 CFR 1.76)

発明の名称 Title of Invention	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY

下記発明者である私は、つぎのことがらを宣誓します。
As the below named inventor, I hereby declare that:

本宣誓は
This declaration is directed to:

☐ 添付されている、あるいは
The attached application, or

■ 米国出願、あるいは_____に出願されたPCT国際願番号_____として出願されているものに宛てられています。
United States application or PCT international application number 15/638,449 filed on June 30, 2017.

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The above-identified application was made or authorized to be made by me.

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I believe that I am the original inventor or an original joint inventor of a claimed invention in the application.

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発明者の正式氏名
LEGAL NAME OF INVENTOR

発明者:
Inventor: Yohei MOMMA

日付(任意):
Date (Optional): 07/20/2017

署名:
Signature: Yohei Momma

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申請データシート(37 CFR 1.76)を使った実用及び意匠登録出願宣誓書(37 CFR 1.63)
DECLARATION (37 CFR 1.63) FOR UTILITY OR DESIGN APPLICATION USING AN APPLICATION DATA SHEET (37 CFR 1.76)

発明の名称 Title of Invention	POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY

下記発明者である私は、つぎのことがらを宣誓します。
As the below named inventor, I hereby declare that:

本宣誓は
This declaration is directed to:

☐ 添付されている、あるいは
The attached application, or

■ 米国出願、あるいは_____に出願されたPCT国際願番号_____として出願されているものに宛てられています。
United States application or PCT international application number 15/638,449 filed on June 30, 2017.

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The above-identified application was made or authorized to be made by me.

私は本出願書中にあらわれるもともとの発明者、あるいはもともとの共同発明者です。
I believe that I am the original inventor or an original joint inventor of a claimed invention in the application.

本宣誓書において故意に虚偽の申し立てを行った場合は 18 U.S.C. 1001 により、罰金あるいは最高五(5)年の禁固刑、あるいはその両方による罰則の対象となることを認めます。
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発明者の正式氏名
LEGAL NAME OF INVENTOR

発明者:
Inventor: Masahiro TAKAHASHI

日付(任意):
Date (Optional): 07/19/2017

署名:
Signature: Masahiro Takahashi

備考: 出願データシート(PTO/AIA/14 あるいはその同等用紙) は、発明の自主独立体全体の命名を含め、本用紙に添付すること。なお残余の発明者ごとにPTO/SB/AIA01 用紙を使用する。
Note: An application data sheet (PTO/AIA/14 or equivalent), including naming the entire inventive entity, must accompany this form. Use an additional PTO/SB/AIA01 form for each additional inventor.

This collection of information is required by 35 U.S.C. 115 and 37 CFR 1.63. 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 1 minute 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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本用紙に記載された情報は、下記の通常使用目的に従います。

1. 本用紙に記載されている情報は、情報公開法 (5 U.S.C. 552) およびプライバシー保護法 (5 U.S.C 552a) が許容する範囲において極秘扱いとなります。本記録システムの記録は、本記録の開示が情報公開法で要求されているか否かを判断するために、司法省に開示される場合があります。
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5. 特許協力条約のもとで出願された国際出願に関する本記録システム内の記録は、通常使用目的として、特許協力条約に基づき、世界知的所有権機関に開示される場合があります。
6. 本記録システムの記録は通常使用目的として、国家安全保障 (35 U.S.C. 181) による再審理 、および原子力法 (42 U.S.C. 218(c) にもとづく再審理の目的において、他の連邦政府機関に開示される場合があります。
7. 本記録システムの記録は通常使用目的として、44 U.S.C. 2904 及び 2906 に基づく記録管理慣行及びプログラムの改善を推奨するために、米一般調達局長官(GSA)により、当機関の責任の一部として行われる記録の検査機関中に、GSA 、またはその被指名人に開示される場合があります。上記の開示は、本目的のための記録検査を規定する GSA 規定、及び関連 (GSA あるいは商務省) の指令に準拠して行われます。かかる開示は、個人を特定する目的のもとに使用されてはなりません。
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下記発明者である私は、つぎのことがらを宣誓します。
As the below named inventor, I hereby declare that:

本宣誓は
This declaration is directed to:

☐ 添付されている、あるいは
The attached application, or

■ 米国出願、あるいは_____に出願されたPCT国際願番号_____として出願されているものに宛てられています。
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発明者の正式氏名
LEGAL NAME OF INVENTOR

発明者:
Inventor: Ayae TSURUTA

日付(任意):
Date (Optional): 07 / 14 / 2017

署名:
Signature: Ayae TSURUTA

備考: 出願データシート(PTO/AIA/14 あるいはその同等用紙) は、発明の自主独立体全体の命名を含め、本用紙に添付すること。なお残余の発明者ごとにPTO/SB/AIA01 用紙を使用する。
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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

22204
NIXON PEABODY, LLP
799 Ninth Street, NW
SUITE 500
WASHINGTON, DC 20001

POA ACCEPTANCE LETTER

OC000000095622972

Date Mailed: 11/22/2017

NOTICE OF ACCEPTANCE OF POWER OF ATTORNEY

This is in response to the Power of Attorney filed 11/20/2017.

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**.

/rnegga/

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15/638,449	06/30/2017	Teruaki OCHIAI	740756-004563

CONFIRMATION NO. 1074

PUBLICATION NOTICE

22204
NIXON PEABODY, LLP
799 Ninth Street, NW
SUITE 500
WASHINGTON, DC 20001

OC000000096624165

Title:POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY

Publication No.US-2018-0013130-A1
Publication Date:01/11/2018

NOTICE OF PUBLICATION OF APPLICATION

The above-identified application will be electronically published as a patent application publication pursuant to 37 CFR 1.211, et seq. The patent application publication number and publication date are set forth above.

The publication may be accessed through the USPTO's publically available Searchable Databases via the Internet at www.uspto.gov. The direct link to access the publication is currently http://www.uspto.gov/patft/.

The publication process established by the Office does not provide for mailing a copy of the publication to applicant. A copy of the publication may be obtained from the Office upon payment of the appropriate fee set forth in 37 CFR 1.19(a)(1). Orders for copies of patent application publications are handled by the USPTO's Public Records Division. The Public Records Division can be reached by telephone at (571) 272-3150 or (800) 972-6382, by facsimile at (571) 273-3250, by mail addressed to the United States Patent and Trademark Office, Public Records Division, Alexandria, VA 22313-1450 or via the Internet.

In addition, information on the status of the application, including the mailing date of Office actions and the dates of receipt of correspondence filed in the Office, may also be accessed via the Internet through the Patent Electronic Business Center at www.uspto.gov using the public side of the Patent Application Information and Retrieval (PAIR) system. The direct link to access this status information is currently https://portal.uspto.gov/pair/PublicPair. Prior to publication, such status information is confidential and may only be obtained by applicant using the private side of PAIR.

Further assistance in electronically accessing the publication, or about PAIR, is available by calling the Patent Electronic Business Center at 1-866-217-9197.

Office of Data Managment, Application Assistance Unit (571) 272-4000, or (571) 272-4200, or 1-888-786-0101

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
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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 01/07/2019

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
01/07/2019	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 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 2 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 30 June 2017.

☐ 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) 1-22 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) _____ is/are rejected.

8) ☐ Claim(s) _____ is/are objected to.

9) ☑ Claim(s) 1-22 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

Election/Restriction

1. Restriction to one of the following inventions is required under 35 U.S.C. 121:

I. **Claims 1-16**, drawn to a positive electrode active material, classified in H01M4/366.

II. **Claims 17-22**, drawn to a method for forming a positive electrode active material, classified in C23C22/83.

The inventions are independent or distinct, each from the other because:

2. Inventions II and I are related as process of making and product made. The inventions are distinct if either or both of the following can be shown: (1) that the process as claimed can be used to make another and materially different product or (2) that the product as claimed can be made by another and materially different process (MPEP § 806.05(f)). In the instant case, the product as claimed can be made by another and materially different process, such as, a method that omits the first mixing and the first heating steps.

3. Restriction for examination purposes as indicated is proper because all the inventions listed in this action are independent or distinct for the reasons given above and there would be a serious search and/or examination burden if restriction were not required because one or more of the following reasons apply:

- the inventions have acquired a separate status in the art in view of their different classification; and
- the inventions require a different field of search (e.g., searching different classes/subclasses or electronic resources, or employing different search strategies or search queries).

Applicant is advised that the reply to this requirement to be complete must include (i) an election of a invention to be examined even though the requirement may be traversed (37 CFR 1.143) **and (ii) identification of the claims encompassing the elected invention**.

The election of an invention may be made with or without traverse. To reserve a right to petition, the election must be made with traverse. If the reply does not distinctly and specifically point out supposed errors in the restriction requirement, the election shall be treated as an election without traverse. Traversal must be presented at the time of election in order to be considered timely. Failure to timely traverse the requirement will result in the loss of right to petition under 37 CFR 1.144. If claims are added after the election, applicant must indicate which of these claims are readable upon the elected invention.

Should applicant traverse on the ground that the inventions are not patentably distinct, applicant should submit evidence or identify such evidence now of record showing the inventions to be obvious variants or clearly admit on the record that this is the case. In either instance, if the examiner finds one of the inventions unpatentable over the prior art, the evidence or admission may be used in a rejection under 35 U.S.C. 103 or pre-AIA 35 U.S.C. 103(a) of the other invention.

4. Applicant is reminded that upon the cancellation of claims to a non-elected invention, the inventorship must be corrected in compliance with 37 CFR 1.48(a) if one or more of the currently named inventors is no longer an inventor of at least one claim remaining in the application. A request to correct inventorship under 37 CFR 1.48(a) must be accompanied by an application data sheet in accordance with 37 CFR 1.76 that identifies each inventor by his or her legal name and by the processing fee required under 37 CFR 1.17(i).

5. The examiner has required restriction between product or apparatus claims and process claims. Where applicant elects claims directed to the product/apparatus, and all product/apparatus claims are subsequently found allowable, withdrawn process claims that include all the limitations of the allowable product/apparatus claims should be considered for rejoinder. All claims directed to a nonelected process invention must include all the limitations of an allowable product/apparatus claim for that process invention to be rejoined.

In the event of rejoinder, the requirement for restriction between the product/apparatus claims and the rejoined process claims will be withdrawn, and the rejoined process claims will be fully examined for patentability in accordance with 37 CFR 1.104. Thus, to be allowable, the rejoined claims must meet all criteria for patentability including the requirements of 35 U.S.C. 101, 102, 103 and 112. Until all claims to the elected product/apparatus are found allowable, an otherwise proper restriction requirement between product/apparatus claims and process claims may be maintained. Withdrawn process claims that are not commensurate in scope with an allowable product/apparatus claim will not be rejoined. See MPEP § 821.04. Additionally, in order for rejoinder to occur, applicant is advised that the process claims should be amended during prosecution to require the limitations of the product/apparatus claims. **Failure to do so may result in no rejoinder.** Further, note that the prohibition against double patenting rejections of 35 U.S.C. 121 does not apply where the restriction requirement is withdrawn by the examiner before the patent issues. See MPEP § 804.01.

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

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								
	1	÷								
	2	÷								
	3	÷								
	4	÷								
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	22	÷								

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	)	

RESPONSE TO RESTRICTION REQUIREMENT AND AMENDMENT

Mail Stop Amendment - Via EFS
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the Office Action mailed **January 7, 2019**, please amend the above-identified application and consider Applicants' remarks as follows.

Amendments to the Claims begins on **page 2** of this paper.

Remarks begins on **page 6** of this paper

IN THE CLAIMS:

1. (Original) A positive electrode active material comprising:

a first region positioned inside the positive electrode active material; and

a second region and a third region positioned in a superficial portion of the positive electrode active material,

wherein the third region is positioned closer to a surface of the positive electrode active material than the second region is,

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 representative elements, 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. (Original) The positive electrode active material according to claim 1,

wherein the third compound essentially consists of the representative elements.

7. (Original) The positive electrode active material according to claim 1,

wherein the third compound comprises at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride.

8. (Original) A secondary battery comprising:

a positive electrode comprising the positive electrode active material according to claim 1; and

a negative electrode.

9. (Original) A positive electrode active material comprising:

a first region positioned inside the positive electrode active material; and

a second region and a third region positioned in a superficial portion of the positive electrode active material,

wherein the third region is positioned closer to a surface of the positive electrode active material than the second region is,

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. (Original) 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 measured by X-ray photoelectron spectroscopy is 1, a concentration of titanium is greater than or equal to 0.05 and less than or equal to 0.4, a concentration of magnesium is greater than or equal to 0.4 and less than or equal to 1.5, and a concentration of fluorine is greater than or equal to 0.05 and less than or equal to 1.5.

14. (Original) The positive electrode active material particle according to claim 13, further comprising:

a first region positioned inside the positive electrode active material particle; and

a second region and a third region positioned in the superficial portion of the positive electrode active material particle,

wherein the third region is positioned closer to a surface of the positive electrode active material particle than the second region is,

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)

REMARKS

The Restriction Requirement of **January 7, 2019** was received and carefully reviewed. In response to this requirement, Applicant hereby elects **Invention I**, claims 1-16, drawn to a positive electrode active material, without traverse.

Non-elected claims 17-22 are canceled at this time to file a divisional application directed thereto. Accordingly, claims 1-16 are pending for consideration.

Favorable consideration and prompt allowance of the instant application are respectfully requested.

Should the Examiner believe anything further is desirable in order to place the application in better condition for allowance, the Examiner is encouraged to contact Applicant's undersigned representative at the telephone number listed below.

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
Tel: 202-585-8000

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. Cullien
Filed: June 30, 2017	)	Confirmation No.: 1074
For: POSITIVE ELECTRODE ACTIVE MATERIAL, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY	)	

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-2004-103566; WO-2012-005180 and JP-2004-342554 were previously cited by Japan Patent Office in a Japanese Office Action mailed **January 8, 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.

The undersigned certifies that each item of information contained in this information disclosure statement 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

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

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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

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-2004-103566A	04-02-2004			Abst.
		WO-2012/005180	01-12-2012			Abst.
		JP-2004-342554A	12-02-2004			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.*

Electronic Acknowledgement Receipt

EFS ID:	35304092
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:	01-MAR-2019
Filing Date:	30-JUN-2017
Time Stamp:	17:26:10
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_Response_to_RR_OA.pdf	130806 b89840d4714f641ddf5f3324acc7df54276742ef	yes	6

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Response to Election / Restriction Filed	1	1
	Claims	2	5
	Applicant Arguments/Remarks Made in an Amendment	6	6

Warnings:					
Information:					
2	Transmittal Letter	740756-004563_IDS_Transmittal.pdf	106473 b4a8861ac2ee5fecb661da2a970fa351fae0922d	no	2
Warnings:					
Information:					
3	Information Disclosure Statement (IDS) Form (SB08)	740756-004563_IDS_1449.pdf	151843 f924915b6d376e7e9cb7a72d8aa28c03913c7512	no	1
Warnings:					
Information:					
This is not an USPTO supplied IDS fillable form					
4	Foreign Reference	JP2004-103566.pdf	3593276 566444a61e973ca02c240b821cbc47a9d718dcdb	no	19
Warnings:					
Information:					
5	Foreign Reference	WO2012005180A1.pdf	3016290 cd5de9beb87390785b7e811f0478a2a2d3b3c9ca	no	69
Warnings:					
Information:					
6	Foreign Reference	JP2004-342554.pdf	2980857 827359b4857d5bb159bcd5e263e4cfb3f4619445	no	19
Warnings:					
Information:					
		Total Files Size (in bytes):	9979545		

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
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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	03/01/2019	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 16	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".	/KAREN T WASHINGTON/
*** 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.

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		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.

[1] Applicant's unique citation designation number (optional). [2] Applicant is to place a check mark here if English language Translation is attached.

Electronic Acknowledgement Receipt

EFS ID:	35369559
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:	08-MAR-2019
Filing Date:	30-JUN-2017
Time Stamp:	16:08:33
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	Transmittal Letter	740756-4563_Supplemental_IDS.pdf	169100 e1d949c5420153199777ae069687b6d5765 40a17	no	2

Warnings:

Information:					
2	Information Disclosure Statement (IDS) Form (SB08)	740756-004563_PTO-1449.pdf	164368 3bf39e0c02327ea7cb8c688d45895504f4d61418	no	1
Warnings:					
Information:					
This is not an USPTO supplied IDS fillable form					
3	Foreign Reference	WO2012132387.pdf	22750313 49787c6043b3bf6338e1d3429b29c1bbe4aae59c	no	188
Warnings:					
Information:					
		Total Files Size (in bytes):	23083781		

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.

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, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY	)	

SUPPLEMENTAL INFORMATION DISCLOSURE STATEMENT

Mail Stop Amendment – VIA EFS
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

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.

Applicant wishes to direct the attention of the Examiner to the *Foreign Patent Document* **WO-2012-132387** cited on **Sheet 5** of the Form PTO-1449 filed in the IDS of October 2, 2017 in the instant application, which was filed with an *English-language Abstract*.

The instant Supplemental Information Disclosure Statement is being filed to cite a **Full *English-language translation*** of the **WO-2012-132387** for the Examiner's consideration at this time.

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 documents are being submitted within three (3) months of the filing of this application or entry into the national stage of this application, or before the first Office Action on the merits, whichever is later, therefore no fee or certification is required under 37 C.F.R. § 1.97(b).

It is believed that no fee is associated with this Information Disclosure Statement. Nevertheless, the Commissioner is hereby authorized to charge any fees required in connection with this filing, or credit any overpayment, to Deposit Account No. 19-2380.

Respectfully submitted,

By: /Jeffrey L. Costellia, Reg. No. 35,483/
Jeffrey L. Costellia
Registration No. 35,483

NIXON PEABODY LLP
CUSTOMER No. 22204
799 9th Street NW., Suite 500
Washington, D.C. 20001-4501
(202) 585-8000

UNITED STATES PATENT AND TRADEMARK OFFICE

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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 03/14/2019

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
03/14/2019	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 01 March 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) 1-16 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) 1-16 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 01 March 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

2. **Claims 1-16** are pending.

3. **Claims 17-22** are canceled.

Election/Restrictions

4. Applicant's election without traverse of Group I in the reply filed on 01 March 2019 is acknowledged.

Priority

5. Receipt is acknowledged of certified copies of papers required by 37 CFR 1.55.

Information Disclosure Statement

6. The information disclosure statements (IDS) submitted on 02 October 2017, 24 October 2017, and 01 March 2019 were filed before the mailing of a first Office Action on the merits. The submissions comply with the provisions of 37 CFR 1.97. Accordingly, the information disclosure statements are being considered by the examiner.

Drawings

7. 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.

8. The drawings are objected to because:

Reference character **9634** does not have a lead line or are not underlined. Lead lines are required for each reference character except for those which indicate the surface or cross section on which they are placed. Such a reference character must be underlined to make it clear that a lead line has not been left out by mistake. Lead lines must be executed in the same way as lines in the drawing. See 37 CFR 1.84 (q).

Figs. 37A-40E2 do not have satisfactory reproduction characteristics. See U.S. 2018/0013130 A1. All drawings must be made by a process, which will give them satisfactory reproduction characteristics. Every line, number, and letter must be durable, clean, black (except for color drawings), sufficiently dense and dark, and uniformly thick and well defined. The weight of all lines and letters must be heavy enough to permit adequate reproduction. This requirement applies to all lines however fine, to shading, and to lines representing cut surfaces in sectional views. Lines and strokes of different thicknesses may be used in the same drawing where different thicknesses have a different meaning. See MPEP 37 CFR 1.84 (l).

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. The figure or figure number of an amended drawing should not be labeled as "amended." If a drawing figure is to be canceled, the appropriate figure must be removed from the replacement sheet, and where necessary, the remaining figures must be renumbered and appropriate changes made to the brief description of the several views of the drawings for consistency. Additional replacement sheets may be necessary to show the renumbering of the remaining figures. 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.

9. 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., [0369] and [0670]),

11a (e.g., [0414]-[0419], [0421]-[0428], [0431], [0434]-[0436], and [0670]),

11b (e.g., [0414]-[0419], [0421]-[0428], [0431], [0434]-[0436], and [0670]),

12a (e.g., [0414], [0415], [0421], [0431], and [0670]),

12b (e.g., [0414]-[0419], [0421]-[0428], [0431], [0434]-[0436], and [0670]),

14 (e.g., [0415], [0416], [0418], and [0670]),

15a (e.g., [0419] and [0670]),

15b (e.g., [0419] and [0670])

17 (e.g., [0434] and [0670]),

50 (e.g., [0414]-[0416], [0423], [0424], [0427], [0431]-[0436], and [0670]),

51 (e.g., [0414], [0420]-[0422], [0424], [0427], [0431], [0433], [0435], and [0670]),

61 (e.g., [0421], [0431], [0434], and [0670]),

62 (e.g., [0421], [0422], [0424], [0425], [0427], [0428], and [0670]),

71 (e.g., [0422], [0423], and [0670]),

72 (e.g., [0422], [0423], and [0670]),

73 (e.g., [0431], [0435], and [0670]),

9631a (e.g., [0458], [0460], [0461], [0464], [0466], and [0670]),

9631b (e.g., [0458], [0460], [0461], [0464], [0466], and [0670]),

9638 (e.g., [0460] and [0670]),

9639 (e.g., [0461] and [0670]), and

8401 (e.g., [0487] and [0670]).

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 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),

8025 (e.g., Fig. 27B), 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

11. The disclosure is objected to because of the following informalities:

Reference character **300** is referred to as a secondary battery in paragraph [0363]. Reference character **300** is used to designate a coin-type secondary battery. Reference character **300** should be referred to as a coin-type secondary battery in paragraph [0363].Reference character **600** is referred to as a coin-type secondary battery in paragraph [0368].

Reference character **600** is used to designate a cylindrical secondary battery. Reference

character **600** should be referred to as a cylindrical secondary battery in paragraph [0368].

Reference character **600** is referred to as a secondary battery in paragraph [0369]. Reference character **600** is used to designate a cylindrical secondary battery. Reference character **600** should be referred to as a cylindrical secondary battery in paragraph [0369].

Reference character **617** is described in paragraph [0369]. However, reference character **617** is not present in the drawings.

Reference character **980** is referred to as a secondary battery in paragraph [0395]. Reference character **980** is used to designate a laminated secondary battery. Reference character **980** should be referred to as a laminated secondary battery in paragraph [0395].

Reference character **980** is referred to as a secondary battery in paragraph [0398]. Reference character **980** is used to designate a laminated secondary battery. Reference character **980** should be referred to as a laminated secondary battery in paragraph [0398].

Reference character **980** is referred to as a secondary battery in paragraph [0399]. Reference character **980** is used to designate a laminated secondary battery. Reference character **980** should be referred to as a laminated secondary battery in paragraph [0399].

Reference character **500** is referred to as a secondary battery in paragraph [0412]. Reference character **500** is used to designate a laminated secondary battery. Reference character **500** should be referred to as a laminated secondary battery in paragraph [0412].

Reference character **50** is described in paragraphs [0414]-[0416], [0423], [0424], [0427], and [0431]-[0436]. However, reference character **50** is not present in the drawings.

Reference character **11a** is described in paragraphs [0414]-[0419], [0421]-[0428], [0431], and [0434]-[0436]. However, reference character **11a** is not present in the drawings.

Reference character **11b** is described in paragraphs [0414]-[0419], [0421]-[0428], [0431],

and [0434]-[0436]. However, reference character **11b** is not present in the drawings.

Reference character **12a** is described in paragraphs [0414], [0415], [0421], and [0431]. However, reference character **12a** is not present in the drawings.

Reference character **12b** is described in paragraphs [0414]-[0419], [0421]-[0428], [0431], and [0434]-[0436]. However, reference character **12b** is not present in the drawings.

Reference character **51** is described in paragraphs [0414], [0420]-[0422], [0424], [0427], [0431], [0433] and [0435]. However, reference character **51** is not present in the drawings.

Reference character **14** is described in paragraphs [0415], [0416], and [0418]. However, reference character **14** is not present in the drawings.

Reference character **61** is described in paragraphs [0421], [0431], and [0434]. However, reference character **61** is not present in the drawings.

Reference character **62** is described in paragraphs [0421], [0422], [0424], [0425], [0427], and [0428]. However, reference character **62** is not present in the drawings.

Reference character **71** is described in paragraphs [0422] and [0423]. However, reference character **71** is not present in the drawings.

Reference character **72** is described in paragraphs [0422] and [0423]. However, reference character **72** is not present in the drawings.

Reference character **15a** is described in paragraph [0419]. However, reference character **15a** is not present in the drawings.

Reference character **15b** is described in paragraph [0419]. However, reference character **15b** is not present in the drawings.

Reference character **214** is described in paragraph [0426] as "not illustrated." However, reference character **214** is illustrated in Fig. 23A.

Reference character **73** is described in paragraphs [0431] and [0435]. However, reference character **73** is not present in the drawings.

Reference character **17** is described in paragraph [0434]. However, reference character **17** is not present in the drawings.

Reference character **9631a** is described in paragraphs [0458], [0460], [0461], [0464], and [0466]. However, reference character **9631a** is not present in the drawings.

Reference character **9631b** is described in paragraphs [0458], [0460], [0461], [0464], and [0466]. However, reference character **9631b** is not present in the drawings.

Reference character **9638** is described in paragraph [0460]. However, reference character **9638** is not present in the drawings.

Reference character **9639** is described in paragraph [0461]. However, reference character **9639** is not present in the drawings.

Reference character **8401** is described in paragraph [0487]. However, reference character **8401** is not present in the drawings.

Reference character **101** is referred to as a region in paragraph [0670]. Reference character **101** is used to designate a first region. Reference character **101** should be referred to as a first region in paragraph [0670].

Reference character **102** is referred to as a region in paragraph [0670]. Reference character **101** is used to designate a second region. Reference character **102** should be referred to as a second region in paragraph [0670].

Reference character **103** is referred to as a region in paragraph [0670]. Reference character **103** is used to designate a third region. Reference character **103** should be referred to as a third region in paragraph [0670].

Reference character **104** is referred to as a region in paragraph [0670]. Reference character **104** is used to designate a fourth region. Reference character **104** should be referred to as a fourth region in paragraph [0670].

Reference character **112** is referred to as a layer in paragraph [0670]. Reference character **112** is used to designate a layer containing titanium. Reference character **112** should be referred to as a layer containing titanium in paragraph [0670].

Reference character **122** is referred to as a layer in paragraph [0670]. Reference character **122** is used to designate a layer containing titanium. Reference character **122** should be referred to as a layer containing titanium in paragraph [0670].

Reference character **125** is referred to as a layer in paragraph [0670]. Reference character **125** is used to designate a layer containing lithium titanate. Reference character **125** should be referred to as a layer containing lithium titanate in paragraph [0670].

Reference character **300** is referred to as a secondary battery in paragraph [0670]. Reference character **300** is used to designate a coin-type secondary battery. Reference character **300** should be referred to as a coin-type secondary battery in paragraph [0670].

Reference character **500** is referred to as a secondary battery in paragraph [0670]. Reference character **500** is used to designate a laminated secondary battery. Reference character **500** should be referred to as a laminated secondary battery in paragraph [0670].

Reference character **600** is referred to as a secondary battery in paragraph [0670]. Reference character **600** is used to designate a cylindrical secondary battery. Reference character **600** should be referred to as a cylindrical secondary battery in paragraph [0670].

Reference character **617** is described in paragraph [0670]. However, reference character **617** is not present in the drawings.

Reference character **980** is referred to as a secondary battery in paragraph [0670]. Reference character **980** is used to designate a laminated secondary battery. Reference character **980** should be referred to as a laminated secondary battery in paragraph [0670].

Reference character **11a** is described in paragraph [0670]. However, reference character **11a** is not present in the drawings.

Reference character **11b** is described in paragraph [0670]. However, reference character **11b** is not present in the drawings.

Reference character **12a** is described in paragraph [0670]. However, reference character **12a** is not present in the drawings.

Reference character **12b** is described in paragraph [0670]. However, reference character **12b** is not present in the drawings.

Reference character **14** is described in paragraph [0670]. However, reference character **14** is not present in the drawings.

Reference character **15a** is described in paragraph [0670]. However, reference character **15a** is not present in the drawings.

Reference character **15b** is described in paragraph [0670]. However, reference character **15b** is not present in the drawings.

Reference character **17** is described in paragraph [0670]. However, reference character **17** is not present in the drawings.

Reference character **50** is described in paragraph [0670]. However, reference character **50** is not present in the drawings.

Reference character **51** is described in paragraph [0670]. However, reference character **51** is not present in the drawings.

Reference character **61** is described in paragraph [0670]. However, reference character **61** is not present in the drawings.

Reference character **62** is described in paragraph [0670]. However, reference character **62** is not present in the drawings.

Reference character **71** is described in paragraph [0670]. However, reference character **71** is not present in the drawings.

Reference character **72** is described in paragraph [0670]. However, reference character **72** is not present in the drawings.

Reference character **73** is described in paragraph [0670]. However, reference character **73** is not present in the drawings.

Reference character **8021** is referred to as a charging apparatus in paragraph [0670]. Reference character **8021** is used to designate a ground-based charging apparatus. Reference character **8021** should be referred to as a ground-based charging apparatus in paragraph [0670].

Reference character **8401** is described in paragraph [0670]. However, reference character **8401** is not present in the drawings.

Reference character **9625** is referred to as a switch in paragraph [0670]. Reference character **9625** is used to designate a power saving mode changing switch. Reference character **9625** should be referred to as a power saving mode changing switch in paragraph [0670].

Reference character **9626** is referred to as a switch in paragraph [0670]. Reference character **9626** is used to designate a display mode switch. Reference character **9626** should be referred to as a display mode changing switch in paragraph [0670].

Reference character **9631a** is described in paragraph [0670]. However, reference character **9631a** is not present in the drawings.

Reference character **9631b** is described in paragraph [0670]. However, reference character **9631b** is not present in the drawings.

Reference character **9638** is described in paragraph [0670]. However, reference character **9638** is not present in the drawings.

Reference character **9639** is described in paragraph [0670]. However, reference character **9639** is not present in the drawings.

Appropriate correction is required.

12. The title of the invention is not descriptive. A new title is required that is clearly indicative of the invention to which the claims are directed.

The following title is suggested: POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY.

Claim Rejections - 35 USC § 112

13. The following is a quotation of 35 U.S.C. 112(b):

> (b) CONCLUSION.—The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the inventor or a joint inventor regards as the invention.

The following is a quotation of 35 U.S.C. 112 (pre-AIA), second paragraph:

> The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

14. **Claims 1-16** 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 1 recites the limitation "a first region positioned inside the positive electrode active material." Claim 1 also recites "[a] positive electrode active material comprising: a first region." It is unclear how a component (i.e., a first region) of an element (i.e., a positive electrode active material) can be inside the element. Therefore, the configuration of the positive electrode active material is unclear.

The term "representative" in **claim 1** 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 unclear.

Claims 2-7 are directly dependent from claim 1 and include all the limitations of claim 1. Therefore, claims 2-7 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 8 recites the limitation "the positive electrode active material according to claim 1" and includes all the limitations of claim 1. Therefore, claim 8 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 9 recites the limitation "a first region positioned inside the positive electrode active material." Claim 9 also recites "[a] positive electrode active material comprising: a first region." It is unclear how a component (i.e., a first region) of an element (i.e., a positive electrode active material) can be inside the element. Therefore, the configuration of the positive electrode active material is unclear.

Claim 10-12 are directly dependent from claim 9 and include all the limitations of claim 1. Therefore, claims 10-12 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 13 recites the limitation "a concentration of titanium is greater than or equal to 0.05 and less than or equal to 0.4." Claim 13 has previously recited the limitation "a concentration of cobalt in a superficial portion of the positive electrode active material measured by X-ray photoelectron spectroscopy." It is unclear if "a concentration of titanium" is also "in a superficial portion of the positive electrode active material" and "measured by X-ray photoelectron spectroscopy."

Claim 13 recites the limitation "a concentration of magnesium is greater than or equal to 0.4 and less than or equal to 1.5." Claim 13 has previously recited the limitation "a concentration of cobalt in a superficial portion of the positive electrode active material measured by X-ray photoelectron spectroscopy." It is unclear if "a concentration of magnesium" is also "in a superficial portion of the positive electrode active material" and "measured by X-ray photoelectron spectroscopy."

Claim 13 recites the limitation "a concentration of fluorine is greater than or equal to 0.05 and less than or equal to 1.5." Claim 13 has previously recited the limitation "a concentration of cobalt in a superficial portion of the positive electrode active material measured by X-ray photoelectron spectroscopy." It is unclear if "a concentration of fluorine" is also "in a superficial portion of the positive electrode active material" and "measured by X-ray photoelectron spectroscopy."

Claim 14 recites the limitation "a first region positioned inside the positive electrode active material particle." Claim 14 also recites "[t]he positive electrode active material particle … comprising: a first region." It is unclear how a component (i.e., a first region) of an element (i.e., a positive electrode active material particle) can be inside the element. Therefore, the configuration of the positive electrode active material is unclear.

Claim 15 is directly dependent from claim 14, indirectly dependent from claim 13, and includes all the limitations of claims 13 and 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 16 recites the limitation "the positive electrode active material particle according to claim 13" and includes all the limitations of claim 13. Therefore, claim 16 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. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless –
>
> (a)(1) the claimed invention was patented, described in a printed publication, or in public use, on sale or otherwise available to the public before the effective filing date of the claimed invention.

16. **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 positioned inside the positive electrode active material (see complex oxide particles, [0022]); and
- a second region (see coating layer, [0021]) and a third region (see surface layer, [0030]) positioned in a superficial portion of the positive electrode active material (see cathode active material, [0032]),
- wherein the third region (see surface layer, [0030]) is positioned closer to a surface of the positive electrode active material than the second region is (see coating layer, [0030]),
- 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 representative elements (see surface layer, [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 7**, Ooyama discloses all claim limitations set forth above and further discloses a positive electrode active material:

- wherein the third compound comprises at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride (see magnesium, [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 positioned inside the positive electrode active material (see complex oxide particles, [0022]); and
- a second region (see coating layer, [0021]) and a third region (see surface layer, [0030]) positioned in a superficial portion of the positive electrode active material (see cathode active material, [0032]),
- wherein the third region (see surface layer, [0030]) is positioned closer to a surface of the positive electrode active material than the second region is (see coating layer, [0030]),

- 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 representative elements (see surface layer, [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 measured by X-ray photoelectron spectroscopy is 1 (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),

- a concentration of titanium is greater than or equal to 0.05 and less than or equal to 0.4 (see surface layer, [0030]),
- a concentration of magnesium 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 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 positioned inside the positive electrode active material particle (see complex oxide particles, [0022]); and
- a second region (see coating layer, [0021]) and a third region (see surface layer, [0030]) positioned in a superficial portion of the positive electrode active material particle (see cathode active material, [0032]),
- 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 measured by X-ray photoelectron spectroscopy is 1 (see $Li_{(1+x)}Co_{(1-y)}M_yO_{(2-z)}$, [0022]),
- a concentration of titanium is greater than or equal to 0.05 and less than or equal to 0.4 (see surface layer, [0030]),
- a concentration of magnesium 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 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

17. The following is a quotation of 35 U.S.C. 103 which forms the basis for all obviousness rejections set forth in this Office action:

> A patent for a claimed invention may not be obtained, notwithstanding that the claimed invention is not identically disclosed as set forth in section 102, if the differences between the claimed invention and the prior art are such that the claimed invention as a whole would have been obvious before the effective filing date of the claimed invention to a person having ordinary skill in the art to which the claimed invention pertains. Patentability shall not be negated by the manner in which the invention was made.

18. The factual inquiries set forth in *Graham v. John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103 are summarized as follows:

1. Determining the scope and contents of the prior art.

2. Ascertaining the differences between the prior art and the claims at issue.

3. Resolving the level of ordinary skill in the pertinent art.

4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

19. This application currently names joint inventors. In considering patentability of the claims the examiner presumes that the subject matter of the various claims was commonly owned as of the effective filing date of the claimed invention(s) absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and effective filing dates of each claim that was not commonly owned as of the effective filing date of the later invention in order for the examiner to consider the applicability of 35 U.S.C. 102(b)(2)(C) for any potential 35 U.S.C. 102(a)(2) prior art against the later invention.

20. **Claims 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 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.

21. **Claim 5** is/are rejected under 35 U.S.C. 103 as being unpatentable over Ooyama (US 2007/0224506 A1) as applied to **claim 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.

22. **Claim 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 1**, Ooyama discloses a positive electrode active material comprising:

- a first region positioned inside the positive electrode active material (see complex oxide particles, [0022]); and
- a second region (see coating layer, [0021]) and a third region (see surface layer, [0030]) positioned in a superficial portion of the positive electrode active material (see cathode active material, [0032]),
- wherein the third region (see surface layer, [0030]) is positioned closer to a surface of the positive electrode active material than the second region is (see coating layer, [0030]),
- 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.

23. **Claims 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 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.

Conclusion

24. 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-20070224506-A1	09-2007	Ooyama; Tomoyo	C01G23/04	429/231.3
*	B	US-20040229123-A1	11-2004	Takahashi, Takeshi	H01M4/131	429/231.1
*	C	US-20120258358-A1	10-2012	YURA; Yukinobu	H01M4/02	429/209
*	D	US-20100233542-A1	09-2010	Endo; Daisuke	H01M4/0445	429/223
	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
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NON-PATENT DOCUMENTS

*		Include as applicable: Author, Title Date, Publisher, Edition or Volume, Pertinent Pages)
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	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						
	1	÷	✓						
	2	÷	✓						
	3	÷	✓						
	4	÷	✓						
	5	÷	✓						
	6	÷	✓						
	7	÷	✓						
	8	÷	✓						
	9	÷	✓						
	10	÷	✓						
	11	÷	✓						
	12	÷	✓						
	13	÷	✓						
	14	÷	✓						
	15	÷	✓						
	16	÷	✓						
	17	÷	-						
	18	÷	-						
	19	÷	-						
	20	÷	-						
	21	÷	-						
	22	÷	-						

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

CPC Combination Sets - Searched*		
Symbol	Date	Examiner

US Classification - Searched*			
Class	Subclass	Date	Examiner
429	231.3	03/07/2019	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

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-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/06 10:02
S3	55	(CN-101148263-$ | JP-08037007-$ | JP-11025983-$ | JP-2003331824-$ | JP-2004196604-$ | JP-2005225734-$ | JP-2005332629-$ | JP-2006318928-$ | JP-	US-PGPUB; USPAT; USOCR; FPRS;	OR	ON	2019/03/06 10:03

		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-$).DID.	EPO; JPO; DERWENT; IBM_TDB			
S4	35	(US-0033749-$ | 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-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/06 10:25
S6	261	S2 S3 S4 S5	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/06 10:28
S5	6	(JP-2004103566-$ | JP-2004342554-$ | VA-223131450-$ | WO-2012005180-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/06 10:28
S12	3	S11 and mismatch	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S11	261	S7 S8 S9 S10	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S10	6	(JP-2004103566-$ | JP-2004342554-$ | VA-223131450-$ | WO-2012005180-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S9	35	(US-0033749-$ | US-0034516-$ | US-0131780-$ | US-0944137-$ | US-20080131780-$ | US-20110033749-$ | US-20120034516-$).DID. (CN-101208269-$ | CN-101675547-$ | CN-102210045-$ | EP-2352190-$ | IB-2017053896-$ | JP-	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT;	OR	ON	2019/03/07 09:44

		2011181527-$ | JP-2012508444-$ | KR-100944137-$ | KR-20080024105-$ | KR-20090086440-$ | KR-20100052419-$ | VA-223131450-$ | WO-2007052712-$ | WO-2009057722-$ | WO-2010053328-$).DID.	IBM_TDB			
S8	55	(CN-101148263-$ | 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-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:44
S7	166	(US-3172388-$ | 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-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	2019/03/07 09:44
S15	4	S12 S14	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$4	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:45
S13	0	S11 and mis-match	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:45
S17	1	S16 and mismatch	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 09:49
S16	2	US-20110300441-$.did.	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-$).did.	USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB			10:12
S23	18	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) with (lithium adj titanate "LiTiO.sub.2") near2 (coating layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:21
S24	35	H01M.cpcl. and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide LiCoO2) with (lithium adj titanate "LiTiO.sub.2" LiTiO2) near2 (coating coat coated layer film)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:22
S26	8	(WO-2014032405-$\|JP-2004103566-$|JP-2003007299-$\|JP-2003331846-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:26
S25	6	(WO-20014032405-$\|JP-2004103566-$|JP-2003007299-$\|JP-2003331846-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:26
S27	4	(US-20030211391-$\|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
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
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			
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
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
S32	87	S31 not S29	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
S36	117	S35 not S29	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:37
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
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-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 10:59
S43	64	S41 not (S35 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 11:00
S42	1	15/638449.app.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 11:00
S44	7	(KR-20060085085-$\| 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
S47	10	S46 not (S41 S35 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS;	OR	ON	2019/03/07 12:11

			EPO; JPO; DERWENT; IBM_TDB			
S46	10	(US-20070202411-$ | US-20120021284-$ | US-20120156565-$ | US-20160149215-$).DID. (KR-20070082892-$ | KR-20120069398-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:11
S48	4	(US-20140057173-$\|US-20150017550-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:20
S50	15	S49 not (S46 l36l30 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:24
S49	17	(US-20030087155-$ | US-20160149215-$ | US-9343745-$).DID. (EP-3086389-$ | JP-2017216243-$ | KR-20080045855-$ | KR-20180064196-$ | WO-2015093725-$ | WO-2018038501-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:24
S51	2	US-20160372739-$.did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:30
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
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-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 12:44

		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-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-$).DID.			
S54	169	(US-20020071991-$ | 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-2018511911-$ | KR-100914406-$ | KR-20160127887-$ | TW-201208183-$ | TW-	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:12

		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-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:39
S58	88	S57 not (S54 S52 S49 S46 S41 S35 S25 S37 S39 S40 S16 S11 S42)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:40
S57	108	(US-20020071991-$ | 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-$).DID.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 13:40
S59	6	(WO-2011016553-$\|JP-2011028976-$|JP-2008016243-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT;	OR	ON	2019/03/07 14:01

			IBM_TDB			
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-$).did.	US-PGPUB; USPAT; FPRS; JPO	OR	ON	2019/03/07 14:02
S60	4	(US-20120135315-$\|US-20110020708-$).did.	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	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
S64	1	S62 and S63	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:06
S63	1	S61 and Fm3m	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; IBM_TDB	OR	ON	2019/03/07 14:10
S66	2	S61 and (lithium adj cobaltate "LiCo.2" lithium adj cobalt adj oxide) and (lithium adj	US-PGPUB; USPAT;	OR	ON	2019/03/07 14:27

		titanate "LiTiO.sub.2") and (magnesium adj oxide MgO)	USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB			
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
S70	8	S69 not S68	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/07 14:34
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
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
L2	38	L1 and (F fluoride fluorine)	US-PGPUB; USPAT; USOCR; FPRS; EPO; JPO; DERWENT; IBM_TDB	OR	ON	2019/03/08 12:02

L1	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-$) .d id . or (US-20120135315-$ or US-20110020708-$).d id . or (US-8927148-$) d. idor.(WO-2012005180-$ or WO-2014032405-$ or KR-20060085085-$ or WO-2011016553-$) .d id . or (JP-2004342554-$ or JP-2004103566-$ or JP-2003331846-$ or JP-2002151078-$ or JP-2012113823-$ or JP-2010118312-$ or JP-2009218198-$ or JP-2017033641-$ or JP-2008016243-$) .d id .	US-PGPUB; USPAT; FPRS; JPO	OR	ON	2019/03/08 12:02

EAST Search History (Interference)

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3/8/2019 12:26:52 PM
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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 1 of 4

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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-2012/0034516	02-09-2012	Koo.C et al.	
		US-2008/0131780	06-05-2008	Kawasato.T et al.	
		US-2011/0033749	02-10-2011	Uchida.M et al.	

FOREIGN PATENT DOCUMENTS

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		JP-2012-508444	04-05-2012			Abst.
		WO-2010/053328	05-14-2010			Abst.
		EP-2352190A	08-03-2011			Eng.
		KR-2010-0052419A	05-19-2010			Abst.
		CN-102210045A	10-05-2011			Abst.
		JP-2011-181527A	09-15-2011			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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Sheet 2 of 4

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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		WO-2007/052712	05-10-2007			Abst.
		KR-2008-0024105A	03-17-2008			Abst.
		CN-101208269A	06-25-2008			Abst.
		KR-10-0944137	02-24-2010			Abst.
		WO-2009/057722	05-07-2009			Abst.
		KR-2009-0086440A	08-12-2009			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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Sheet 3 of 4

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First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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		CN-101675547A	03-17-2010			Abst.

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				Filing Date	June 30, 2017
				First Named Inventor	Teruaki OCHIAI
(Use as many sheets as necessary)				Art Unit	1725
				Examiner Name	Anna Basia Ridley
Sheet	4	of	4	Attorney Docket Number	740756-004563

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		INTERNATIONAL SEARCH REPORT (APPLICATION NO. PCT/IB2017/053896) DATED October 3, 2017.	Eng.
		WRITTEN OPINION (APPLICATION NO. PCT/IB2017/053896) DATED October 3, 2017.	Eng.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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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

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Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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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, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY

FILING or 371(c) DATE	CLASS	GROUP ART UNIT	ATTORNEY DOCKET NO.
06/30/2017	429	1725	740756-004563
RULE			

APPLICANTS

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Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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Sheet 2 of 10

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Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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Sheet 3 of 10

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Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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Sheet 4 of 10

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		WO-2012/132387	10-04-2012			Abst.
		WO-2011/141486	11-17-2011			Abst.
		WO-2012/124242	09-20-2012			Abst.

Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 6 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

U. S. PATENT DOCUMENTS

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		US-2016/0118646	04-28-2016	Ikenuma.T	
		US-2012/0328956	12-27-2012	Oguni.T et al.	
		US-2013/0045418	02-21-2013	Oguni.T et al.	
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FOREIGN PATENT DOCUMENTS

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Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

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Sheet 7 of 10

Complete if Known	
Application Number	15/638,449
Filing Date	June 30, 2017
First Named Inventor	Teruaki OCHIAI
Art Unit	1725
Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

U. S. PATENT DOCUMENTS

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		US-2014/0295068	10-02-2014	Nanba.K et al.	
		US-8,883,351	11-11-2014	Todoriki.H et al.	
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Sheet	8	of	10

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Attorney Docket Number	740756-004563

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		US-2009/0011335	01-08-2009	Takeda.K et al.	
		US-8,945,772	02-03-2015	Kawakami.T et al.	
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		US-2012/0064409	03-15-2012	Zhamu.A et al.	

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Art Unit	1725
Examiner Name	Anna Basia Ridley
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		Katsuno.H et al., "Growth modes in two-dimensional heteroepitaxy on an elasticsubstrate", J. CRYST. GROWTH (JOURNAL OF CRYSTAL GROWTH), February 15, 2005, Vol. 275, No. 1-2, pp. e283-e288, ELSEVIER.	Eng.
		Taguchi.N et al., "Characterization of MgO-coated-LiCoO2 particles by analytical transmission electron microscopy", JOURNAL OF POWER SOURCES , 2016, Vol. 328, pp. 161-166, ELSEVIER.	Eng.
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Examiner Signature	/Sean P Cullen, Ph.D./	Date Considered	03/06/2019

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Sheet 10 of 10

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Examiner Name	Anna Basia Ridley
Attorney Docket Number	740756-004563

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		TAN.H et al., "Oxidation state and chemical shift investigation in transition metal oxides by EELS", ULTRAMICROSCOPY, May 1, 2012, Vol. 116, pp. 24-33, ELSEVIER.	Eng.
		McCalla.E et al., "The spinel and cubic rocksalt solid-solutions in the Li-Mn-Ni oxide pseudo-ternary system", Solid State Ionics, May 2, 2013, Vol. 242, pp. 1-9, ELSEVIER.	Eng.
		DREYER.D et al., "THE CHEMISTRY OF GRAPHENE OXIDE", CHEMICAL SOCIETY REVIEWS, November 3, 2009, Vol. 39, No. 1, pp. 228-240.	Eng.

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JUN 14 2019
PATENT & TRADEMARK OFFICE

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 **(As Currently Amended)**	)	

AMENDMENT

Mail Stop Amendment – Via EFS
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the **Non-Final Office Action** mailed **March 14, 2019**, please amend the above-identified application and consider Applicant's remarks as follows.

Amendments to the Specification begin on **page 2** of this paper.

Amendments to the Drawings begin on **page 3** of this paper and include attached replacement sheets.

Amendments to the Claims begins on **page 4** of this paper.

Remarks begin on **page 8** of this paper.

Amendment to Specification

Please amend the title of the application as follows:

--POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE ~~, METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY~~--

Amendments to the Specification:

The amendments to the specification are shown in the attached *Substitute Specification* showing the changes in the specification relative to the previous version as per 37 C.F.R. § 1.125 and a clean version of the substitute specification.

Substitute Specification shows the amendments to paragraphs [0336], [0339], [0340], [0353], [0367], [0402], [430], and [0615].

No new matter is introduced by the present amendment.

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. In addition, original FIGs. 37A-40E accompanies this replacement sheet to replace the corresponding drawings on record, to show the cleaner and legible drawings.

No new matter is introduced by the present amendment.

Attachment: Replacement Sheets

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 ~~positioned inside~~ in the positive electrode active material; ~~and~~

a second region ~~and a third region positioned~~ in ~~a superficial portion of~~ 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 third region is positioned closer to a surface of the positive electrode active material than the second region is,~~

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 ~~representative elements~~ 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. (Currently Amended) The positive electrode active material according to claim 1,

wherein the third compound essentially consists of ~~the representative elements~~ the one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride.

7. (Cancelled)

8. (Original) A secondary battery comprising:

a positive electrode comprising the positive electrode active material according to claim 1; and

a negative electrode.

9. (Currently Amended) A positive electrode active material comprising:

a first region ~~positioned inside~~ in the positive electrode active material; ~~and~~

a second region ~~and a third region positioned~~ in ~~a superficial portion of~~ 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 third region is positioned closer to a surface of the positive electrode active material than the second region is,~~

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 ~~measured by X-ray photoelectron spectroscopy~~ is 1, a concentration of titanium in a 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. (Currently Amended) The positive electrode active material particle according to claim 13, further comprising:

a first region ~~positioned inside~~ in the positive electrode active material particle; ~~and~~

a second region ~~and a third region positioned~~ in ~~the superficial portion of~~ 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 third region is positioned closer to a surface of the positive electrode active material particle than the second region is,~~

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)

REMARKS

The Non-Final Office Action mailed **March 14, 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 1-16 were pending in the present application, of which claims 1, 9 and 13 are independent.

By this amendment, claims 1, 6, 9, 13 and 14 are amended and claim 7 is canceled.

For the reasons set forth in detail below, all claims are believed to be in condition for allowance. Favorable reconsideration is requested.

Claim Amendments

Claim 1 and 9 are amended to recite that the positive electrode active material as "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." Claim 14 is similarly amended. These are clarifying amends which are supported in the as filed specification, for example in paragraph [0054] and FIG 1A.

Claim 1 and 6 are amended to recite "wherein the third region comprises a third compound comprising at least one of magnesium oxide, calcium oxide, beryllium oxide, lithium fluoride, and sodium fluoride." Support for this amendment can be found in the as filed specification, for example in paragraph [0083].

Claim 13 is amended to clarify "a concentration of titanium in a 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." Claim 13 is also amended by deletion of the recitation of "measured by X-ray photoelectron spectroscopy."

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 **9634** does not have a lead line or are not underlined. Lead lines are required for each reference character except for those which indicate the surface or cross section on which they are placed. Examiner proposes such a reference character must be underlined to make it clear that a lead line has not been left out by mistake.

D. Figs. 37A-40E2 do not have satisfactory reproduction characteristics.

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: **617**, **11a**, **11b**, **12a**, **12b**, **14**, **15a**, **15b**, **17**, **50**, **51**, **61**, **62**, **71**, **72**, **73**, **9631b**, **9638**, **9639**, and **8401**.

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 D above, the Figs. 37A-40E2 as filed on June 30, 2017 are being re-printed and hand delivered to the Office. These figures are of high resolution and sharpness but appear to lose some resolution during uploading at filing via EFS. Applicant submits that, although the as filed figures are of sufficient quality for understanding of the application, the hand delivered figures are of higher quality and suggests the Office may be able to scan these into their system with a higher resolution than the Applicant representative can.

Regarding the §1.84(p)(5) objection, Applicant amends the figures as reflected in the Replacement Sheets to address this objection.

For these reasons Applicant submits the Objection to the drawings should be withdrawn.

Objection to the Specification

The Examiner objects to the specification for various reasons enumerated on pages 7-14 of the Office Action. The Examiner also objected to the title.

In response, Applicant made amendments to the claims and specification consistent with the Examiner's request, shown in the instant response and in the Substitute specification,

and provided also as a clean replacement specification. The title has also been amended using the wording suggested by the Examiner.

For these reasons Applicant submits the Objection to the specification should be withdrawn.

Claim Rejections – 35 USC § 112

Claims 1-16 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.

Regarding claim 1, 9 the Examiner on page 15 states that "the configuration of the positive electrode active material is unclear." Regarding claim 9 the Examiner also believes "the limitation "representative elements" is unclear." With respect to claim 13 the Examiner views the recitations of a concentration of titanium, magnesium, and fluorine as unclear, where it is not clear if these are in a superficial portion of the positive electrode active material and measured by X-ray photoelectron spectroscopy. Regarding claim 14 the Examiner believes the configuration of the positive electrode active material is unclear.

Without acquiescing to this rejection and desirous to expedite the prosecution of this application, the base claims 1, 9 and 14 are amended. The amendment to claim 1 and 9 clarifies the configuration of the positive electrode active material as "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." Similar clarifying language is used to amend claim 14. Claim 1 is also amended by deletion of the term "representative elements." Claim 13 is amended to specify "a concentration of titanium in a 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." Claim 13 is also amended by deletion of the recitation of "measured by X-ray photoelectron spectroscopy."

For at least these reasons, the § 112 rejection of the base claims 1, 9 and 13 should be withdrawn, and the rejection of claim 14 should be withdrawn. The rejection of dependent

claims 2–8, 10–12 and 14–16 by virtue of their dependence on one of the base claims should also be withdrawn.

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.

As stated in the MPEP. § 2131 "A claim is anticipated only if each and every element as set forth in the claim is found, either expressly or inherently described, in a single prior art reference." Ooyama does not disclose each and every element of the base claims and therefore cannot support this § 102 rejection. For example, 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. Applicant submits that 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 other elements as recited in claim 1. Specifically, [0022] of Ooyama does not teach "a type of a crystal structure of the first compound"; [0035] and [0036] of Ooyama does not teach "a type of a crystal structure of the second compound"; and [0030] of Ooyama 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".

Nor does Ooyama teach other elements as recited in claim 13. Claim 13 clearly recites the feature of "a superficial portion of the positive electrode active material particle", which Applicant notes may correspond to "a surface layer" in Ooyama. However, [0022], [0028], [0035], or [0029] is not related to "the surface layer". Ooyama [0022] also does not teach the element "polycrystal" as recited in the instant claim 4.

For at least these reasons, this §102 rejection of base claims 1 and 13 should be withdrawn. The rejection of claim 4 should be withdrawn for the reason stated above. 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,

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OPAP AKINAS JUN 14 2019 INT & TRADEMARK OFFICE

DESCRIPTION

POSITIVE ELECTRODE ACTIVE MATERIAL INCLUDING LITHIUM COBALTATE COATED WITH LITHIUM TITANATE AND MAGNESIUM OXIDE, ~~METHOD FOR MANUFACTURING POSITIVE ELECTRODE ACTIVE MATERIAL, AND SECONDARY BATTERY~~

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