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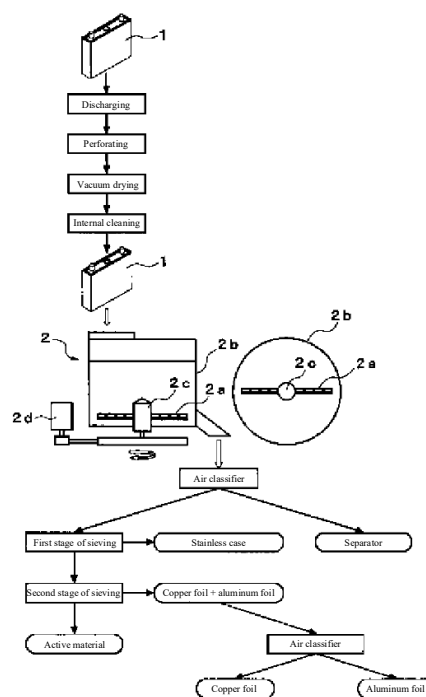
(54) [Title of the Invention] Lithium ion secondary battery recycling method

(57) [Abstract]

[Problem to be Solved] To propose a recycling method for a lithium-ion secondary battery, wherein the battery is crushed by a simple method to separate and recover various constituent materials.

[Means to Solve the Problem] A recycling method for a lithium-ion secondary battery 1, wherein a positive electrode 5, comprising an aluminum foil 5b coated with an active material 5a, and a negative electrode 6, comprising a copper foil 6b coated with an active material 6a, are laminated in multiple layers with a separator 4 made of plastic foil interposed therebetween, and a non-aqueous electrolyte is injected and sealed within a stainless steel container 3, the method comprising: introducing the lithium-ion secondary battery 1 into a cylindrical body 2b of a crusher 2, wherein a flexible wire 2a, having one end fixed to a rotating shaft 2c, rotates within the cylindrical body 2b around the rotating shaft 2c; crushing the battery by the rotating wire 2a; and separating and recovering at least the plastic, stainless steel, aluminum, copper, and active materials from the crushed material using a separation means.

[Selected Drawings] FIG. 1



10 [Scope of Claims]

[Claim 1]

A recycling method for a lithium-ion secondary battery, wherein a positive electrode comprising an aluminum foil coated with an active material and a negative electrode comprising a copper foil coated with an active material are laminated in multiple layers with a separator made of plastic foil interposed there between, and a non-aqueous electrolyte is injected and sealed within a stainless steel container, the method comprising: introducing the lithium-ion secondary battery into a cylindrical body of a crusher, wherein a flexible wire, having one end fixed to a rotating shaft, rotates within the cylindrical body around the rotating shaft; crushing the battery by the rotating wire; and separating and recovering at least the plastic, stainless steel, aluminum, copper, and active materials from the crushed material using a separation means.

20 [Claim 2]

The recycling method for a lithium-ion secondary battery according to Claim 1, wherein the lithium-ion secondary battery is sealed with a sealing plate having an explosion-proof valve, and prior to crushing the lithium-ion secondary battery, the method further comprises: discharging the lithium-ion secondary battery; opening the explosion-proof valve of the sealing plate of the discharged lithium-ion secondary battery; evaporating the non-aqueous electrolyte by vacuum drying; and washing the lithium-ion secondary battery from which the non-aqueous electrolyte has been evaporated.

[Detailed Description of the Invention]

[Technical Field]

[0001]

30 The present invention relates to a recycling method for a lithium-ion secondary battery, aimed at individually recovering various constituent materials of the lithium-ion secondary battery.

[Background Art]

[0002]

In recent years, with the widespread use of small electronic devices such as mobile phones and laptop computers, the demand for lithium-ion secondary batteries, which are compact, lightweight, and have high electromotive force, has been increasing. Additionally, from the perspectives of energy conservation and ecology, next-generation environmentally friendly vehicles such as hybrid and electric vehicles have gained significant attention, with rapidly growing demand. These hybrid and electric vehicles increasingly utilize lithium-ion secondary batteries due to their compact size, light weight, high electromotive force, and high energy density.

40 [0003]

Lithium-ion secondary batteries were developed as an alternative to conventional aqueous electrolyte secondary batteries, such as nickel-cadmium and nickel-metal hydride batteries, which used aqueous electrolytes, in response to the high demand for compact and lightweight secondary batteries prior to the widespread adoption of mobile phones and laptop computers. These batteries have evolved to meet today's societal needs.

[0004]

50 In lithium-ion secondary batteries, carbon is used as the active material for the negative electrode's current collector, while the type of active material used for the positive electrode's current collector classifies the batteries into categories such as Co-based lithium-ion secondary batteries, Co-Ni-Mn-based lithium-ion secondary batteries, and Ni-Co-based lithium-ion secondary batteries. The active materials used in the positive electrodes of these lithium-ion secondary batteries are valuable rare metals such as cobalt, nickel, and manganese. With the increasing demand today, the prices of these materials are rising, and their recycling is required from the perspective of resource conservation.

[0005]

Consequently, methods have been proposed to recover lithium-ion secondary batteries used in small electronic devices such as mobile phones and laptop computers, and to extract and recycle various materials, including valuable rare metals. When recycling lithium-ion secondary batteries for small electronic devices, it is necessary to remove, stabilize, and detoxify the electrolyte and fluorine. Conventionally, these processes were performed through heat treatment or incineration, followed by separation and recovery of the materials.

[0006]

60 Since lithium-ion secondary batteries used in small electronic devices are mostly encased in plastic containers, heat treatment or incineration can burn the plastic containers, enabling the removal, stabilization, and detoxification of the electrolyte and fluorine, as well as facilitating the extraction of internal electrodes and other components.

[0007]

However, lithium-ion secondary batteries for automotive applications often use metal materials such as stainless steel for their containers. Unlike plastic containers, these metal containers do not burn during heat treatment or incineration and remain intact, making it difficult to extract internal electrodes and other components. This necessitates additional processes, such as cutting or crushing the stainless steel containers, which increases the number of processing steps and leads to the problem of larger-scale equipment.

10 [0008]

In response to this, Patent Document 1 below proposes a method for recovering valuable metals from a lithium-ion secondary battery, comprising: a disassembly step of dismantling the lithium-ion secondary battery; a washing step of cleaning the dismantled battery components with alcohol or water to remove the electrolyte and electrolytic substances; a positive electrode active material separation step of immersing the washed dismantled battery components in a sulfuric acid aqueous solution to separate the positive electrode active material from the positive electrode substrate; a leaching step of leaching the separated positive electrode active material in an acidic solution in the presence of a fixed carbon-containing substance; a neutralization step of separating and removing aluminum and copper from the obtained leachate by neutralization; a nickel-cobalt recovery step of separating and recovering nickel and cobalt from the leachate after the neutralization step; and a lithium recovery step of concentrating lithium in the remaining aqueous solution through solvent extraction and back-extraction, followed by separating and recovering lithium as solid lithium carbonate.

20

[0009]

In this conventional method, since the lithium-ion secondary battery is dismantled in the disassembly step and materials such as active materials, aluminum, and copper are subsequently separated and recovered, it is possible to recycle lithium-ion secondary batteries for automotive use, which are encased in stainless steel containers.

[0010]

However, in the conventional method described above, the processing steps performed after dismantling the lithium-ion secondary battery and washing the electrolyte and electrolytic substances with alcohol or water are complex. Additionally, the use of hazardous solutions complicates the equipment and necessitates the treatment of the used solutions, leading to increased equipment and maintenance costs.

30

[Prior Art Documents]

[Patent Documents]

[0011]

[Patent Document 1] Japanese Patent Laid-Open Publication No. 2007-122885

[Summary of the Invention]

[Problem to be Solved by the Invention]

[0012]

In view of these circumstances, the present invention aims to propose a recycling method for a lithium-ion secondary battery, wherein the battery is crushed by a simple method to separate and recover various constituent materials.

40

[Means for Solving the Problem]

[0013]

To solve the above problem, the invention according to Claim 1 provides a recycling method for a lithium-ion secondary battery, wherein a positive electrode comprising an aluminum foil coated with an active material and a negative electrode comprising a copper foil coated with an active material are laminated in multiple layers with a separator made of plastic foil interposed therebetween, and a non-aqueous electrolyte is injected and sealed within a stainless steel container. The method comprises: introducing the lithium-ion secondary battery into a cylindrical body of a crusher, wherein a flexible wire, having one end fixed to a rotating shaft, rotates within the cylindrical body around the rotating shaft; crushing the battery by the rotating wire; and separating and recovering at least the plastic, stainless steel, aluminum, copper, and active materials from the crushed material using a separation means.

50

[0014]

The invention according to Claim 2, in addition to the invention according to Claim 1, is characterized in that the lithium-ion secondary battery is sealed with a sealing plate having an explosion-proof valve, and prior to crushing the lithium-ion secondary battery, the method further comprises: discharging the lithium-ion secondary battery; opening the explosion-proof valve of the sealing plate of the discharged lithium-ion secondary battery; evaporating the non-aqueous electrolyte by vacuum drying; and washing the lithium-ion secondary battery from which the non-aqueous electrolyte has been evaporated.

[Effect of the Invention]

[0015]

60

According to the invention described in Claims 1 and 2, the lithium-ion secondary battery is introduced into a crusher wherein a flexible wire rotates around a rotating shaft within a cylindrical body, and the battery is crushed by the impact force of the wire's striking action. This allows the plastic, stainless steel, aluminum foil, copper foil, and active materials contained in the crushed material to be fragmented into distinct shapes and sizes. By utilizing the properties of each material, separation means such as an air classifier or sieving can be employed to individually separate and recover each material, thereby facilitating recycling.

[0016]

According to the invention described in Claim 2, as a pretreatment before crushing the lithium-ion secondary battery, the explosion-proof valve of the sealing plate is opened, the non-aqueous electrolyte is evaporated by vacuum drying, and the lithium-ion secondary battery, from which the non-aqueous electrolyte has been

10 evaporated, is washed. This enables the removal, stabilization, and detoxification of the electrolyte and fluorine without requiring heat treatment or incineration. As a result, large-scale equipment is not necessary, the number of processing steps can be reduced, and the pretreatment of the lithium-ion secondary battery can be performed safely and easily.

[Brief Description of the Drawings]

[0017]

[FIG. 1] A flowchart illustrating a series of steps in one embodiment of the recycling method for a lithium-ion secondary battery according to the present invention.

20 [FIG. 2] Illustrations of a lithium-ion secondary battery used in the recycling method of the present invention, wherein (a) is a schematic cross-sectional view, and (b) is a partial schematic cross-sectional view of the laminated structure.

[Embodiments for Carrying Out the Invention]

[0018]

30 As shown in Figure 1, one embodiment of the recycling method for a lithium-ion secondary battery according to the present invention involves discharging a used lithium-ion secondary battery for automotive applications, opening the explosion-proof valve 8 of the discharged lithium-ion secondary battery 1 by perforation, evaporating the non-aqueous electrolyte injected into the opened lithium-ion secondary battery 1 by vacuum drying, and, after evaporating the non-aqueous electrolyte, washing the interior of the lithium-ion secondary battery 1 with water for pretreatment. The washed lithium-ion secondary battery 1 is then introduced into a chain-type crusher (crusher) 2 for crushing. Utilizing the properties of each material in the crushed lithium-ion secondary battery, the materials are separated and recovered by separation means, including air classification and two-stage sieving.

[0019]

Here, the lithium-ion secondary battery 1 is a Co-Ni-Mn-based lithium-ion secondary battery. As shown in Figures 2(a) and 2(b), a laminated structure 11, comprising a positive electrode 5 with an active material 5a coated on an aluminum foil 5b and a negative electrode 6 with an active material 6a coated on a copper foil 6b, laminated in multiple layers with a separator 4 made of plastic foil interposed therebetween, is wound and disposed within a stainless steel container 3. A non-aqueous electrolyte is injected, and the container is sealed with a sealing plate 7.

[0020]

40 The positive electrode 5 comprises an aluminum foil 5b serving as the positive electrode current collector, with the active material 5a coated on both surfaces of the aluminum foil 5b. The active material 5a is $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$, formed into a paste. The negative electrode 6 comprises a copper foil 6b serving as the negative electrode current collector, with the active material 6a coated on both surfaces of the copper foil 6b. The active material 6a is carbon (C), formed into a paste.

[0021]

The sealing plate 7, which seals the stainless steel container 3, is provided with an explosion-proof valve 8 at its center. The explosion-proof valve 8 is sealed with an aluminum sheet 8a covering a through-hole. Additionally, the sealing plate 7 is provided with a positive electrode terminal 9 connected to the positive electrode 5 and a negative electrode terminal 10 connected to the negative electrode 6, both protruding outward.

[0022]

50 On the other hand, as shown in Figure 1, the chain-type crusher 2 comprises a bottomed cylindrical body 2b, a cylindrical rotating shaft 2c that stands upright at the center of the bottom of the cylindrical body 2b and rotates by the drive of a motor 2d, and chains (wires) 2a, each having one end connected to the outer peripheral surface of the rotating shaft 2c and the other end as a free end. The chains 2a are arranged such that another chain is connected at a position 180° apart in the circumferential direction of the rotating shaft 2c, with its other end also being a free end. Additionally, two chains 2a are connected to one side of the outer peripheral surface of the rotating shaft 2c.

[0023]

60 The two-stage sieving, which serves as the separation means, consists of a mesh sieve in the first stage that retains crushed material with a shape dimension of 20 mm or larger on the sieve while allowing crushed material with a shape dimension smaller than 20 mm to fall to the second-stage sieve. In the second stage, a mesh sieve retains crushed material with a shape dimension smaller than 20 mm but 1 mm or larger on the sieve, while allowing crushed material with a shape dimension smaller than 1 mm to fall below.

[0024]

By using the lithium-ion secondary battery 1, the chain-type crusher 2, and the two-stage sieving configured as described above, the various materials constituting the lithium-ion secondary battery 1 for automotive applications can be separated and recovered as follows. First, as shown in Figure 1, a used lithium-ion secondary battery 1 for automotive applications is discharged using a commonly available discharger to release any remaining electrical charge.

[0025]

10 Next, a round rod with a slightly smaller diameter than the through-hole of the explosion-proof valve 8 provided on the sealing plate 7 is pressed against the through-hole, breaking through the aluminum sheet 8a sealing the through-hole of the explosion-proof valve 8 to create an opening. The lithium-ion secondary battery 1 is then placed in a vacuum dryer to evaporate the electrolyte injected inside. During this process, the vacuum pressure of the vacuum dryer is set to 0.1 MPa or lower, preferably 0.01 MPa or lower, the drying temperature is set to 80 to 120°C, and drying is performed for 6 to 8 hours.

[0026]

Subsequently, water is injected through the through-hole of the explosion-proof valve 8 provided on the sealing plate 7 of the dried lithium-ion secondary battery 1 to wash the interior. During this process, the interior is washed multiple times with water.

20 [0027]

Next, the dried lithium-ion secondary battery 1 is introduced into the chain-type crusher 2. In this chain-type crusher 2, the rotating shaft 2c rotates by the drive of the motor 2d, causing the chains 2a connected to the rotating shaft 2c to rotate within the cylindrical body 2b.

[0028]

30 The lithium-ion secondary battery 1 introduced into the chain-type crusher 2 is struck by the chains 2a rotating within the cylindrical body 2b for approximately 1 to 2 minutes, thereby crushing the stainless steel container 3. The aluminum foil 5b forming the positive electrode 5 and the copper foil 6b forming the negative electrode 6, both disposed inside the container 3, are crushed, along with the active material 5a coated on the aluminum foil 5b and the active material 6a coated on the copper foil 6b. Additionally, the separator 4 formed of plastic foil is crushed. To prevent ignition during crushing by the chain-type crusher 2, an inert gas may be introduced through an inert gas introduction pipe (not shown) to maintain an inert gas atmosphere within the cylindrical body 2b during crushing.

[0029]

40 During this process, the impact of the striking action of the flexible chain 2a causes the plastic foil to be cut into large pieces with minimal deformation, the stainless steel to be crushed into medium-sized pieces, and the aluminum foil, copper foil, and active materials 5a and 6a to be finely crushed, with each material remaining unentangled. This is due to the differences in elastoplastic properties of each material. The impact of the flexible chain 2a causes the soft and thin plastic foil to be cut into large pieces without significant crushing, the hard stainless steel to deform significantly and be crushed into medium-sized pieces, and the aluminum foil, copper foil, and the active materials 5a and 6a coated thereon to be finely crushed, resulting in differences in the crushed shapes for each material.

[0030]

From the crushed material of the lithium-ion secondary battery 1 crushed by the chain-type crusher 2, the plastic foil forming the separator 4, which has the lightest weight, is separated and recovered using an air classifier as a separation means.

[0031]

50 Next, the crushed material, from which the plastic foil has been separated by air classification, is further separated and recovered using a two-stage sieving process, as another separation means, into materials with a shape dimension of 20 mm or larger, materials with a shape dimension smaller than 20 mm but 1 mm or larger, and materials with a shape dimension smaller than 1 mm. The materials separated by the first stage of the two-stage sieving have a crushed shape dimension of 20 mm or larger and consist of stainless steel, the material of the container 3.

[0032]

The materials with a crushed shape dimension smaller than 20 mm but 1 mm or larger are the aluminum foil 5b and copper foil 6b, which are the materials of the current collectors of the positive electrode 5 and negative electrode 6, respectively.

[0033]

60 The materials with a crushed shape dimension smaller than 1 mm include lithium, cobalt, nickel, manganese, and other components contained in the active material 5a of the positive electrode 5, as well as carbon and other components contained in the active material 6a of the negative electrode 6.

Additionally, the aluminum foil 5b and copper foil 6b with a crushed shape dimension smaller than 20 mm but 1 mm or larger are separated into aluminum foil 5b and copper foil 6b by an air classifier and recovered individually.

[0034]

(Example)

Next, the separation of the crushed material of the lithium-ion secondary battery 1 crushed by the chain-type crusher 2 by sieving is explained based on the analysis results of the proportion of materials by shape dimension. The chain-type crusher 2 and crushing time are under the same conditions as described in the embodiment above. The analysis methods used are combustion ion chromatography for fluorine (F), combustion infrared absorption for carbon (C), and ICP emission spectroscopy for other elements.

10 After the crushing process, large pieces of plastic foil and medium-sized pieces of stainless steel were removed in advance, and then sieve separation was performed, followed by analysis of the recovered materials.

[0035]

Based on this analysis, the proportions of crushed shape dimensions of 2 mm or larger, 1 to 2 mm, and 1 mm or smaller for each material are summarized in Table 1. Additionally, the weight proportions of the recovered materials for each shape dimension are summarized in Table 2.

[0036]

[Table 1]

Analyzed component		≥ 2 mm	2 mm to 1 mm	≤ 1 mm	Method of analysis
Li	wt%	1.64	0.80	3.06	ICP emission spectroscopy
Al	wt%	18.0	7.05	0.97	ICP emission spectroscopy
F	wt%	1.6	1.3	3.7	Combustion - ion chromatography method
Co	wt%	1.96	0.88	3.57	ICP emission spectroscopy
Mn	wt%	13.8	6.57	26.7	ICP emission spectroscopy
Ni	wt%	1.99	1.43	3.63	ICP emission spectroscopy
Cu	wt%	37.8	20.8	1.85	ICP emission spectroscopy
P	wt%	0.19	0.15	0.38	ICP emission spectroscopy
C	wt%	10.9	18.6	28.2	Combustion - Infrared absorption method

[0037]

[Table 2]

Shape/dimensions	Weight proportion of recovered materials (wt%)
≥ 2 mm	32.7
1-2 mm	4.3
≤ 1 mm	63.0
Total	100.0

20 [0038]

Based on the analysis results, the weight proportion of materials crushed to a shape dimension of 2 mm or larger is 32.7 wt% when the total recovered material is taken as 100 wt%. When the materials crushed to a shape dimension of 2 mm or larger are taken as 100 wt%, the contained materials are 18.0 wt% Al and 37.8 wt% Cu. The weight proportion of materials crushed to a shape dimension of 1 to 2 mm is 4.3 wt% when the total recovered material is taken as 100 wt%. When the materials crushed to a shape dimension of 1 to 2 mm are taken as 100 wt%, the contained materials are 7.05 wt% Al and 20.8 wt% Cu. Thus, by using a sieve capable of retaining materials with a shape dimension of 1 mm or larger, it is possible to separate and recover the aluminum foil 5b, which is the material of the current collector of the positive electrode 5, and the copper foil 6b, which is the material of the current collector of the negative electrode 6, from the crushed material.

30 [0039]

The weight proportion of materials crushed to a shape dimension of 1 mm or smaller is 63.0 wt% when the total recovered material is taken as 100 wt%. When the materials crushed to a shape dimension of 1 mm or smaller are taken as 100 wt%, the contained materials are 3.06 wt% Li, 3.57 wt% Co, 26.7 wt% Mn, 3.63 wt% Ni, and 28.2 wt% C. Thus, by using a sieve capable of passing materials with a shape dimension smaller than 1 mm, it is possible to separate and recover lithium, cobalt, nickel, and manganese contained in the active material 5a coated on the aluminum foil 5b of the positive electrode 5, as well as carbon contained in the active material 6a coated on the copper foil 6b of the negative electrode 6, from the crushed material.

[0040]

40 According to the recycling method for the lithium-ion secondary battery 1 described in the above embodiment, the lithium-ion secondary battery 1 is introduced into a chain-type crusher 2, wherein a flexible chain 2a rotates around a rotating shaft 2c within a cylindrical body 2b, and is crushed by the impact force of the chain 2a's striking action. This allows the plastic, stainless steel, aluminum foil 5b, copper foil 6b, and active materials 5a and 6a contained in the crushed material to be fragmented into distinct shape dimensions. By utilizing the properties of each material, separation means such as an air classifier or sieving can be employed to individually separate and recover each material, thereby facilitating recycling.

[0041]

Additionally, as a pretreatment before crushing the lithium-ion secondary battery 1, the explosion-proof valve 8 of the sealing plate 7 is perforated, the non-aqueous electrolyte is evaporated by vacuum drying, and the lithium-ion secondary battery 1, from which the non-aqueous electrolyte has been evaporated, is washed. This enables the

10 removal, stabilization, and detoxification of the electrolyte and fluorine without requiring heat treatment or incineration. As a result, large-scale equipment is not necessary, the number of processing steps can be reduced, and the pretreatment of the lithium-ion secondary battery can be performed safely and easily.

[0042]

In the above embodiment, the crusher was described as a chain-type crusher 2, but it is not limited to this. For example, a wire-type crusher using flexible wires instead of chains can also be used.

Additionally, while the separation means were described as using an air classifier and sieving, they are not limited to these. For example, a magnetic separator or a specific gravity separator can also be used.

Furthermore, while the lithium-ion secondary battery was described as a Co-Ni-Mn-based lithium-ion secondary battery, it is not limited to this. For example, Co-based lithium-ion secondary batteries or Ni-Co-based lithium-ion secondary batteries can also be used.

[Industrial Availability]

[0043]

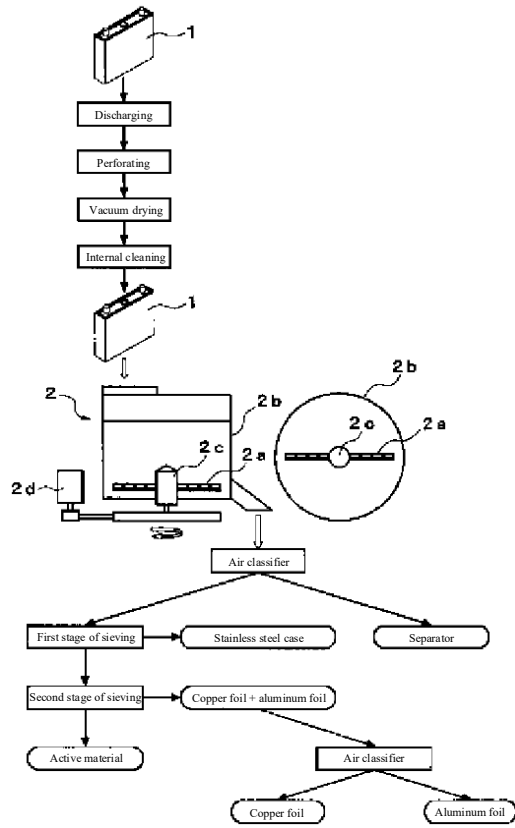
The present invention can be used for recycling lithium-ion secondary batteries.

[Reference Signs List]

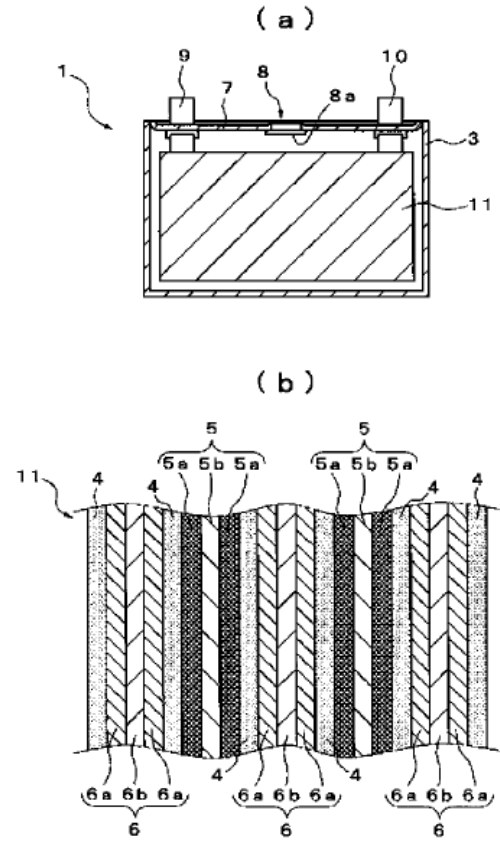
[0044]

- 1 Lithium-ion secondary battery
- 2 Chain-type crusher
- 2a Chain
- 2b Cylindrical body
- 2c Rotating shaft
- 3 Container
- 4 Separator
- 5 Positive electrode
- 5a Active material
- 5b Aluminum foil
- 6 Negative electrode
- 6a Active material
- 6b Copper foil
- 7 Sealing plate
- 8 Explosion-proof valve

[FIG. 1]



[FIG. 2]



Continued from the front page

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