

US 20160293999A1

(19) **United States**(12) **Patent Application Publication**
KIM et al.(10) **Pub. No.: US 2016/0293999 A1**(43) **Pub. Date: Oct. 6, 2016**(54) **ELECTRODE ASSEMBLY HAVING
IMPROVED FLEXURAL RIGIDITY,
METHOD FOR PREPARING SAME, AND
ELECTROCHEMICAL BATTERY
COMPRISING SAME**(71) Applicant: **SAMSUNG SDI CO., LTD.**, Yongin-si,
Gyeonggi-do (KR)(72) Inventors: **Kee Wook KIM**, Suwon-si,
Gyeonggi-do (KR); **Nam Hyo KIM**,
Suwon-si, Gyeonggi-do (KR); **Hyo
Sang YUN**, Suwon-si, Gyeonggi-do
(KR); **Ji Hyun CHUN**, Suwon-si,
Gyeonggi-do (KR)(73) Assignee: **SAMSUNG SDI CO., LTD.**, Yongin-si,
Gyeonggi-do (KR)(21) Appl. No.: **15/038,352**(22) PCT Filed: **Nov. 21, 2014**(86) PCT No.: **PCT/KR2014/011234**

§ 371 (c)(1),

(2) Date: **May 20, 2016****H01M 2/16** (2006.01)**H01M 4/131** (2006.01)**H01M 4/587** (2006.01)**H01M 4/62** (2006.01)**H01M 4/1391** (2006.01)**H01M 4/04** (2006.01)**H01M 4/133** (2006.01)**H01M 4/1393** (2006.01)**H01M 10/0525** (2006.01)**H01M 4/525** (2006.01)(52) **U.S. Cl.**CPC **H01M 10/058** (2013.01); **H01M 10/0525**
(2013.01); **H01M 10/0565** (2013.01); **H01M**
2/162 (2013.01); **H01M 2/1653** (2013.01);
H01M 2/168 (2013.01); **H01M 4/131**
(2013.01); **H01M 4/525** (2013.01); **H01M**
4/623 (2013.01); **H01M 4/625** (2013.01);
H01M 4/1391 (2013.01); **H01M 4/0404**
(2013.01); **H01M 4/0435** (2013.01); **H01M**
4/133 (2013.01); **H01M 4/1393** (2013.01);
H01M 4/587 (2013.01); **H01M 4/622**
(2013.01); **H01M 2220/30** (2013.01); **H01M**
2004/028 (2013.01)

(57)

ABSTRACT

The present invention relates to an electrode assembly, a method for preparing the same, and an electrochemical battery including the same, wherein the electrode assembly comprises: a cathode including a cathode active material and a cathode current collector; an anode including an anode active material and an anode current collector; and a separator interposed between the cathode and the anode. The electrode assembly has a flexural rigidity of 15 kgf/cm² or more when pressed using a pressure of 1 kgf/cm² to 30 kgf/cm² for 1 second to 15 seconds at 20 ° C. to 110 ° C.

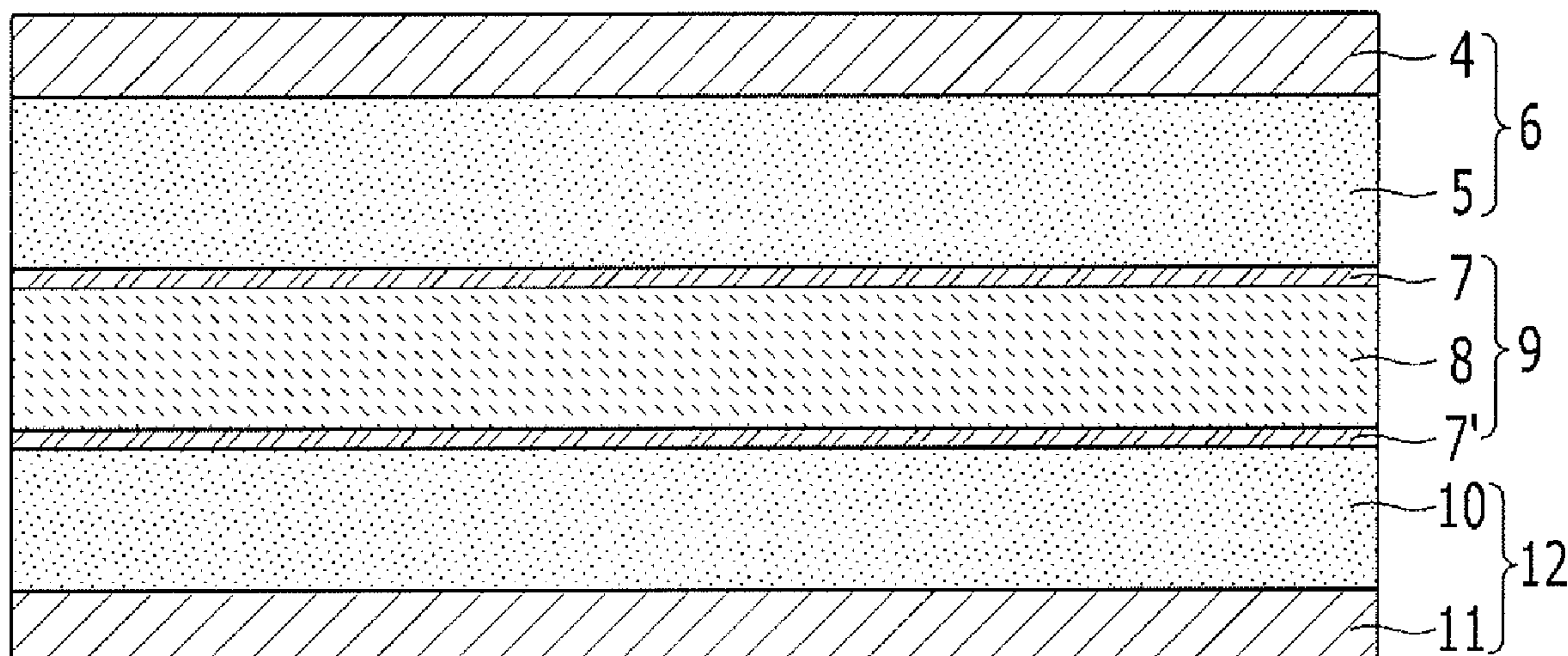
(30) **Foreign Application Priority Data**Nov. 21, 2013 (KR) 10-2013-0142324
Apr. 7, 2014 (KR) 10-2014-0041124
Oct. 2, 2014 (KR) 10-2014-0133339**Publication Classification**(51) **Int. Cl.****H01M 10/058** (2006.01)**H01M 10/0565** (2006.01)200

FIG. 1

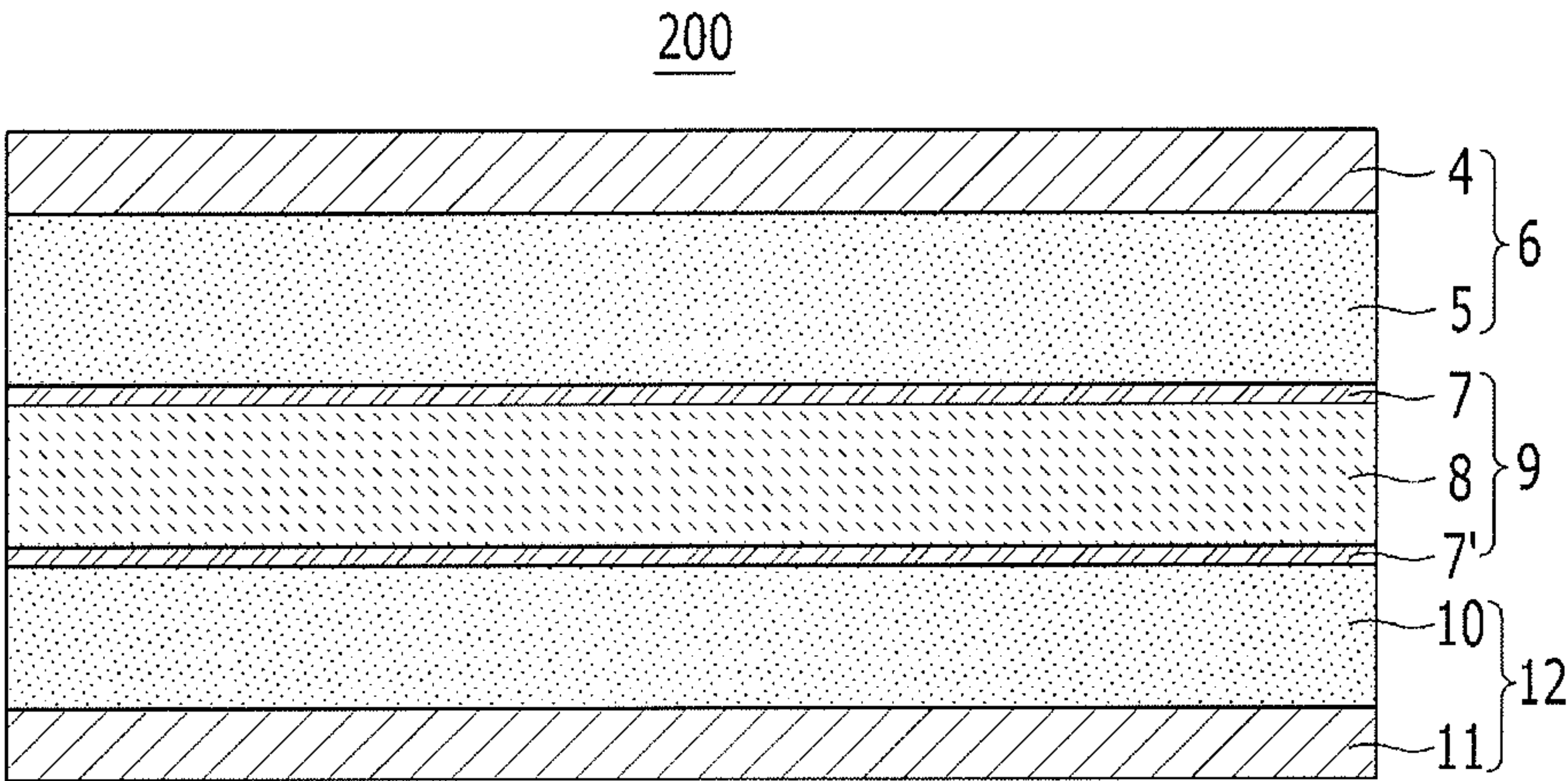
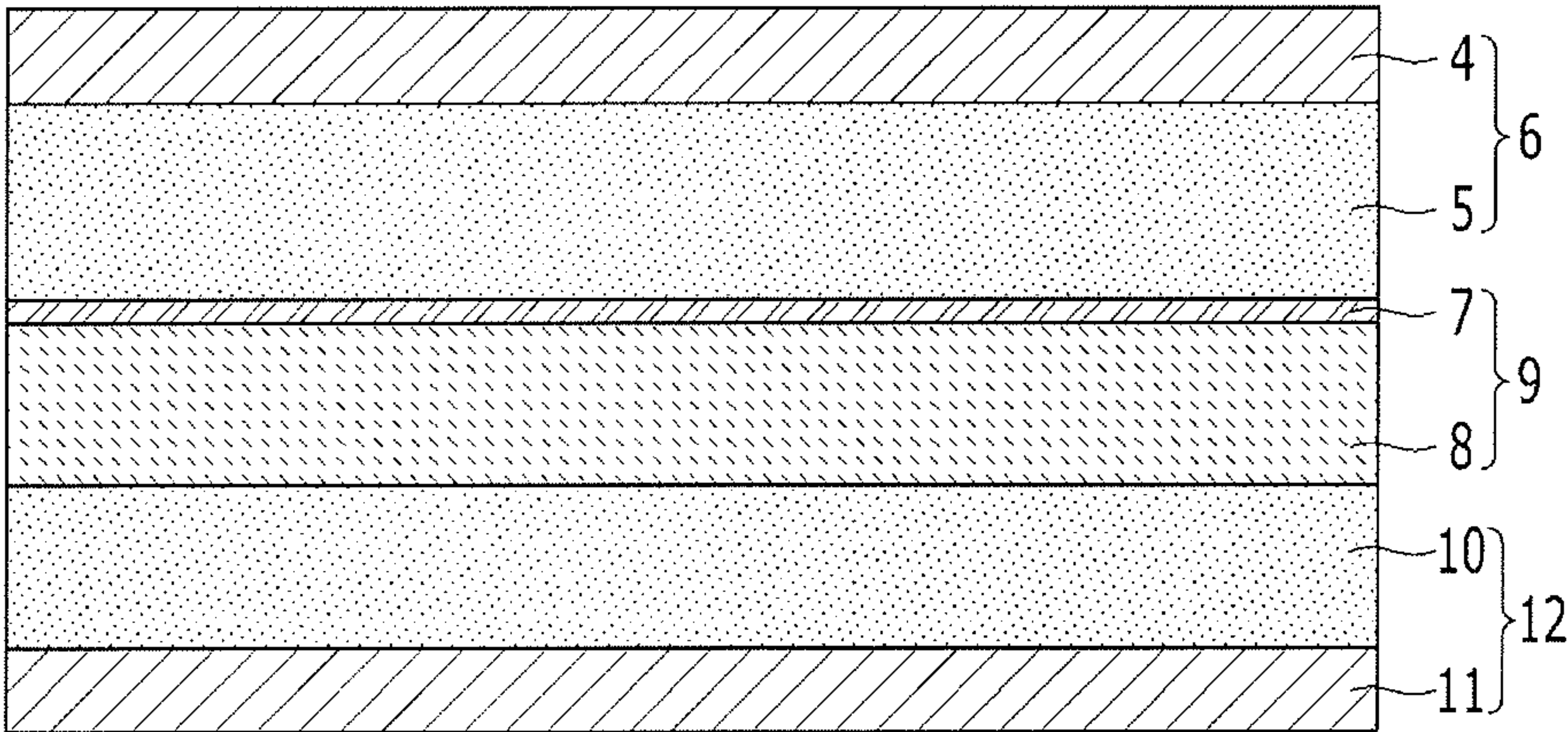


FIG. 2



**ELECTRODE ASSEMBLY HAVING
IMPROVED FLEXURAL RIGIDITY,
METHOD FOR PREPARING SAME, AND
ELECTROCHEMICAL BATTERY
COMPRISING SAME**

CROSS-REFERENCE TO RELATED
APPLICATION

[0001] Korean Patent Applications No. 10-2013-0142324, filed on Nov. 21, 2013, No. 10-2014-0041124, filed on Apr. 7, 2014, and No. 10-2014-0133339, filed on Oct. 2, 2014, in the Korean Intellectual Property Office are incorporated by reference herein in its entirety.

BACKGROUND OF THE INVENTION

[0002] 1. Field of the Invention

[0003] The present invention relates to an electrode assembly with improved flexural rigidity in a battery, a method of preparing the same, and an electrochemical battery including the same.

[0004] 2. Description of the Related Art

[0005] In general, as a portable electronic device such as a video camera, a cell phone, and a portable computer is lightened and conducts high performance, research on a secondary battery as a power source for the portable electronic device is actively being made. This secondary battery may include, for example, a nickel-cadmium battery, a nickel-hydrogen battery, a nickel-zinc battery, a lithium secondary battery, and the like. Among these batteries, the lithium secondary battery may be down-sized and enlarged and also has an advantage of a high voltage and high energy density per unit weight and thus is used in many fields.

[0006] This lithium secondary battery includes an electrode assembly as a main constituent element. However, in a process of manufacturing the electrode assembly by disposing an enlarged separator between electrodes and pressing them, since the separator wound between the electrodes may easily escape therefrom due to an area and/or weight increase according to enlargement of the separator, adherence of the separator to the electrodes needs to be increased. In addition, the electrode assembly requires excellent shape stability in order to prevent a shape change such as deformation of a battery and the like due to continuous charges and discharges.

[0007] In order to improve adherence of the separator to the electrodes and heat resistance of the separator, a method of forming an organic/inorganic mixed coating layer on one surface or both surfaces of the base film of the separator has been known (Korean Registration Patent No. 10-0775310) but may not sufficiently secure desired adherence and thus not be uniformly applied to variously-sized and-shaped separators.

[0008] Accordingly, development of an electrode assembly including a separator having adherence applicable to an enlarged electrochemical battery and thus capable of improving shape stability of the battery is required.

SUMMARY OF THE INVENTION

Technical Object

[0009] The present invention is to provide an electrode assembly having improved adherence between an electrode

and a separator in an electrode assembly and improved shape stability, and an electrochemical battery using the same.

Technical Solution

[0010] According to an example embodiment of the present invention, an electrode assembly includes a cathode on which a positive active material is coated, an anode on which an anode active material is coated, and a separator between the cathode and the anode, wherein the electrode assembly has a flexural rigidity of greater than or equal to 15 kgf/cm² when being compressed at 20° C. to 110° C. for 1 second to 15 seconds, with a pressure of 1 kgf/cm² to 30 kgf/cm². According to another example embodiment of the present invention, an electrochemical battery, particularly a lithium secondary battery includes the electrode assembly according to the example embodiment.

Advantageous Effect

[0011] In an electrode assembly according to example embodiments of the present invention, adherence between an electrode and a separator of the electrode assembly is improved. Accordingly, in a process of manufacturing the electrode assembly, the separator may be prevented from an escape and thus decrease a process inferiority rate and be stored for a long time.

[0012] In addition, the electrode assembly according to example embodiments of the present invention has excellent shape stability and may be minimized from a shape change despite charges and discharges repeated for a long time. Accordingly, a battery manufactured by using the electrode assembly may have highly efficient charge and discharge characteristics and be prevented from deterioration of battery performance.

BRIEF DESCRIPTION OF THE DRAWINGS

[0013] FIG. 1 is a cross-sectional view showing the internal part of an electrode assembly according to one example embodiment of the present invention, and the electrode assembly includes a cathode 6 having a positive active material layer 5 on a cathode current collector 4; an anode 12 having a negative active material layer 12 on an anode current collector 11; and a separator 9 disposed between the cathode 6 and the anode 12 and respectively adhered to the cathode or the anode, and including a porous substrate 8 and porous adhesive layers 7 and 7' on both surfaces of the porous substrate.

[0014] FIG. 2 is a cross-sectional view showing the internal part of an electrode assembly according to another example embodiment of the present invention, and the electrode assembly includes the cathode 6 having the positive active material layer 5 on the cathode current collector 4; the anode 12 having the negative active material layer 10 on the anode current collector 11; and the separator 9 disposed between the cathode 6 and the anode 12 and respectively adhered to the cathode or the anode, and including the porous substrate 8 and the porous adhesive layer 7 on one surface of the porous substrate.

DETAILED DESCRIPTION

[0015] Hereinafter, the present invention is described in detail. The disclosures that are not described in the present specification may be fully recognized and by conveyed by

those skilled in the art in a technical or similar field of the present invention and thus are omitted herein.

[0016] Hereinafter, referring to FIG. 1, an electrode assembly according to an example embodiment of the present invention is described. According to an example embodiment of the present invention, an electrode assembly includes a cathode including a positive active material and a cathode current collector, an anode including an anode active material and an anode current collector, and a separator disposed between the cathode and the anode, wherein the electrode assembly has a flexural rigidity of greater than or equal to 15 kgf/cm² when being compressed at 20° C. to 110° C. for 1 second to 15 seconds, with a pressure of 1 kgf/cm² to 30 kgf/cm².

[0017] The flexural rigidity of greater than or equal to 15 kgf/cm² when being compressed at 20° C. to 110° C. for 1 second to 15 seconds, with a pressure of 1 kgf/cm² to 30 kgf/cm² has a relation with a shape stability of the electrode assembly. This electrode assembly may minimize a battery shape change such as battery deformation and the like despite continuous charges and discharges for a long time and thus realize highly efficient charge and discharge characteristics and prevent deterioration of battery performance. The flexural rigidity may be measured using a 3 point bending machine (ex. UTM) according to ASTM D790, but is not limited thereto. The flexural rigidity may be specifically 17 kgf/cm² to 50 kgf/cm², and more specifically 20 kgf/cm² to 30 kgf/cm².

[0018] Referring to FIG. 1, the electrode assembly according to the embodiment includes a cathode 6 including a positive active material layer 5 on a cathode current collector 4; an anode 12 including an anode active material layer 10 formed on an anode current collector 11; and a separator 9 disposed between the cathode 6 and the anode 12 and respectively attached to the cathode or the anode. The separator 9 may include a porous substrate 8 and porous adhesive layers 7 and 7' on both surfaces of the porous substrate 8.

[0019] The porous substrate 8 may have a plurality of pore and may generally be a porous substrate used in an electrochemical device. Non-limiting examples of the porous substrate 8 may be a polymer film formed of a polymer or a mixture of two or more of polyethylene, polypropylene, polyethyleneterephthalate, polybutyleneterephthalate, polyester, polyacetal, polyamide, polyimide, polycarbonate, polyetheretherketone, polyaryletherketone, polyetherimide, polyamideimide, polybenzimidazole, polyethersulfone, polyphenyleneoxide, a cyclic olefin copolymer, polyphenylenesulfide, and polyethylenenaphthalene. For example, the porous substrate 8 may be a polyolefin-based substrate, and the polyolefin-based substrate may improve safety of a battery due to its improved shut-down function. The polyolefin-based substrate may be, for example, selected from a polyethylene single film, a polypropylene single film, a polyethylene/polypropylene double film, a polypropylene/polyethylene/polypropylene triple film, and a polyethylene/polypropylene/polyethylene triple film. For another example, the polyolefin-based resin may include a non-olefin resin in addition to an olefin resin or a copolymer of olefin and a non-olefin monomer.

[0020] A thickness of the porous substrate 8 may be 1 μm to 40 μm, for example 5 μm to 15 μm. Within the thickness range, a separator may have a desirable thickness that is

thick to prevent a short-circuit between the cathode and the anode of a battery and is also not thick to increase internal resistance.

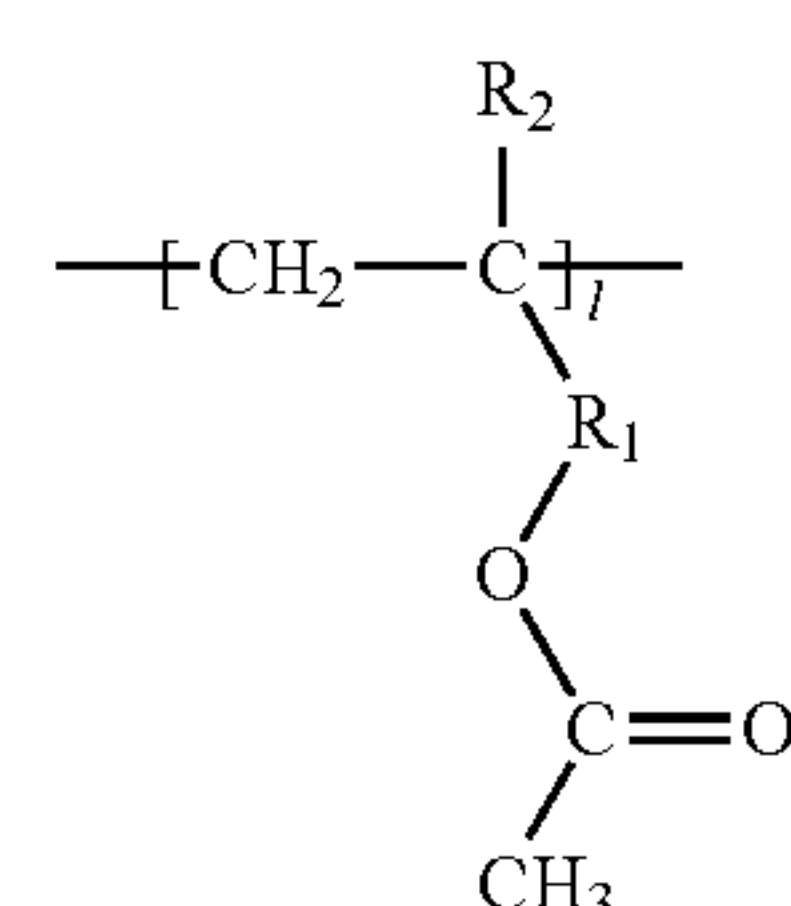
[0021] The porous adhesive layers 7 and 7' may be formed on both surfaces of the porous substrate 8, and may be formed of a porous adhesive layer composition. The porous adhesive layer composition may include an organic binder and a solvent.

[0022] The organic binder may be an acryl-based copolymer, for example a acryl-based copolymer including a (meth)acrylate-based monomer-derived repeating unit. In addition, the acryl-based copolymer may further include an acetate group-containing monomer-derived repeating unit in addition to the (meth)acrylate-based monomer-derived repeating unit. When the acryl-based copolymer having a (meth)acrylate-based monomer-derived repeating unit and/or acetate group-containing monomer-derived repeating unit is used as a binder, a separator may be strongly adhered to a cathode or an anode in a secondary battery environment wherein the separator is actually used and thus be prevented from an escape during an electrode assembly process, decrease a process inferiority rate, and realize a long term storage. In addition, the porous adhesive layer retains an electrolyte solution and thus may maintain satisfactory ion conductivity between the electrodes and not deteriorate a porosity of the porous substrate.

[0023] A glass transition temperature (T_g) of the acryl-based copolymer may be less than 100° C., for example, 20° C. to 60° C., specifically 30° C. to 45° C. Within the ranges, it is advantageous for good adherence and thus ensuring shape stability at a temperature where a separator is disposed between electrodes followed by being compressed.

[0024] The acryl-based copolymer having a (meth)acrylate-based monomer-derived repeating unit and/or an acetate group-containing monomer-derived repeating unit used in an example embodiment of the present invention is not particularly limited as long as it provides good adherence at a compression temperature between the cathode and the anode, and for example, the acryl-based copolymer may be a copolymer by polymerizing at least one (meth)acrylate-based monomer selected from the group consisting of butyl (meth)acrylate, propyl(meth)acrylate, ethyl(meth)acrylate and methyl(meth)acrylate. Or, the acryl-based copolymer may be a copolymer of at least one (meth)acrylate-based monomer selected from the group consisting of butyl (meth)acrylate, propyl (meth)acrylate, ethyl (meth)acrylate, and methyl (meth)acrylate and an acetate group-containing monomer selected from the group consisting of vinyl acetate and allyl acetate.

[0025] The acetate group-containing monomer-derived repeating unit may be a repeating unit of Chemical Formula 1:



[Chemical Formula 1]

[0026] In Chemical Formula 1, R_1 is a single bond or a linear or branched C1 to C6 alkyl, R_2 is hydrogen or methyl, and l is an integer of 1 to 100.

[0027] For example, the acetate group-containing monomer-derived repeating unit may be an acetate group-containing monomer-derived repeating unit selected from the group consisting of vinyl acetate and allyl acetate. The acryl-based copolymer may be prepared by polymerizing (meth)acrylate-based monomers or a (meth)acrylate-based monomer and other monomer in addition to the (meth)acrylate-based monomers. For example, the other monomer may be an acetate group-containing monomer. In this case, a (meth)acrylate-based monomer and another monomer, specifically, an acetate group-containing monomer may be polymerized in a mole ratio of 3:7 to 7:3, specifically 4:6 to 6:4, and more specifically about 5:5. The acryl-based copolymer may be, for example, prepared through a polymerization reaction of a butyl (meth)acrylate monomer, a methyl (meth)acrylate monomer, and a vinyl acetate and/or allyl acetate monomer in a mole ratio of 3 to 5:0.5 to 1.5:4 to 6, specifically, 4:1:5.

[0028] In an example embodiment of the present invention, the porous adhesive layer composition may further include an inorganic particle.

[0029] The inorganic particle used in an example embodiment of the present invention is not particularly limited, and may be an inorganic particle that is generally in this filed. Non-limiting examples of the inorganic particle used in the example embodiment of the present invention may be Al_2O_3 , SiO_2 , B_2O_3 , Ga_2O_3 , TiO_2 , or SnO_2 . These may be used alone or in a mixture of two or more. The inorganic particle used in the example embodiment of the present invention may be, for example, Al_2O_3 (alumina). A size of the inorganic particle used in the example embodiment of the present invention is not particularly limited, and its average particle diameter may be 1 nm to 2,000 nm, for example, 100 nm to 1,000 nm, 300 nm to 500 nm. When the inorganic particle having the size within the ranges, dispersibility of the inorganic particle in the porous adhesive layer composition and formation processibility of a porous adhesive layer may be prevented from being deteriorated, a thickness of the porous adhesive layer may be appropriately controlled and thus reduction of mechanical properties and increase of electrical resistance may be prevented. In addition, sizes of pores generated in the separator are appropriately controlled and thus internal a possibility of short-circuit may be reduced during charge and discharge of a battery. In the porous adhesive layer composition, the inorganic particle may be used in a form of inorganic dispersion liquid including the inorganic particle in an appropriate solvent.

[0030] The appropriate solvent is not particularly limited, and may be a general solvent in this art. The appropriate solvent to disperse the inorganic particle may be, for example, acetone. The inorganic dispersion liquid may be prepared by a general method without particular limitation, and may be prepared, for example by adding Al_2O_3 in acetone in an appropriate amount, and milling the same using beads mill to disperse it.

[0031] In the porous adhesive layer, the inorganic particle may be included in an amount of 70 wt % to 95 wt %, specifically 75 wt % to 90 wt %, and more specifically 80 wt % to 90 wt % based on the total weight of the porous adhesive layer. When the inorganic particle is included

within the range, heat dissipation properties of the inorganic particle may be sufficiently realized, and thermal shrinkage of the separator may be effectively suppressed when a porous adhesive layer is formed on a porous substrate using the same.

[0032] Non-limiting example of the solvent used in an example embodiment of the present invention may be acetone, dimethyl formamide, acetone, dimethylsulfoxide, dimethyl acetamide, dimethylcarbonate, or N-methylpyrrolidone. A content of the solvent may be 20 wt % to 99 wt %, specifically 50 wt % to 95 wt %, and more specifically 70 wt % to 95 wt % based on a weight of the porous adhesive layer composition. When the solvent is included within the range, a porous adhesive layer composition may be easily prepared and a drying process of a porous adhesive layer may be easily performed.

[0033] Thicknesses of the porous adhesive layers 7 and 7' may be 1 μm to 15 μm , specifically 1 μm to 10 μm , more specifically 1 μm to 8 μm , or 1 μm to 5 μm . When the porous adhesive layer has a thickness within the thickness range, excellent thermal stability and adherence may be obtained due to a porous adhesive layer having an appropriate thickness, and internal resistance of a battery is suppressed from being increase by preventing an entire thickness of a separator from being extremely thick.

[0034] The electrode assembly according to the present example embodiment may have a compression thickness variation ratio of greater than or equal to 10% according to Equation 1.

$$\text{Compression thickness variation ratio (\%)} = \left[\frac{\text{Thickness of an electrode assembly compressed at } 20^\circ \text{ C.} - \text{Thickness of the electrode assembly compressed at } 100^\circ \text{ C.}}{\text{Thickness of the electrode assembly compressed at } 20^\circ \text{ C.}} \right] \times 100 \quad [\text{Equation 1}]$$

[0035] In Equation 1, the thickness of an electrode assembly compressed at 20° C. is a thickness of a central portion of an electrode assembly including a stacked cathode/separator/anode after compressing it at 20° C. for 1 to 10 seconds, with a pressure of 1 to 30 kgf/cm² and measuring the thickness in one 1 hour, and the thickness of an electrode assembly compressed at 100° C. is a thickness of a central portion of the electrode assembly for 1 to 10 seconds, with a pressure of 1 to 30 kgf/cm² and measuring the thickness in one 1 hour. When the compression thickness variation ratio is within the range, the separator may be prevented from an escape in the electrode assembly due to excellent adherence during compression of the electrodes and the separator at a high temperature (ex. 100° C.) and thus deteriorate a process inferiority rate and accomplish a long term storage. The compression thickness variation ratio may be specifically greater than or equal to 13% and less than 50%, specifically, greater than or equal to 15% and less than 47%, and more specifically, greater than or equal to 20% and less than 45%. As the compression thickness variation ratio is larger, adherence of the separator to the cathode or the anode may be more improved.

[0036] Hereinafter, referring to FIG. 2, another electrode assembly according to another example embodiment of the present invention is described. Referring to FIG. 2, an electrode assembly according to another example embodiment of the present invention includes a cathode 6 including a positive active material layer 5 formed on a cathode current collector 4; an anode 12 including an anode active material layer 10 formed on a anode current collector 11; and

a separator **9** disposed between the cathode **6** and the anode **12** and attached to the cathode or the anode. The separator **9** may include a porous substrate **8** and a porous adhesive layer **7** formed on one surface of the porous substrate **8**. The electrode assembly according to the present example embodiment has substantially the same constituent elements as those of the electrode assembly according to the example embodiment of the present invention except for forming the porous adhesive layer **7** not on both surfaces but only on one surface of the porous substrate **8** of the separator **9** and thus will not be described in detail.

[0037] The electrode assembly may have a flexural rigidity of greater than or equal to 15 kgf/cm², for example, 17 kgf/cm² to 50 kgf/cm², specifically 20 kgf/cm² to 30 kgf/cm² when being compressed at 20° C. to 110° C. for 1 second to 15 seconds, with a pressure of 1 kgf/cm² to 30 kgf/cm². In other words, even when the porous adhesive layer **7** is formed only on one surface of the porous substrate **8**, sufficient adherence to an electrode (a cathode or an anode) is obtained, and a battery shape change may be minimized despite continuous charges and discharges.

[0038] Hereinafter, an electrode assembly according to another example embodiment of the present invention is described. The electrode assembly according to the present embodiment may additionally include a different kind of organic binder other than the acryl-based copolymer as an organic binder in the porous adhesive layer. This electrode assembly is substantially the same as the electrode assemblies according to the above example embodiment of the present invention or another example embodiment of the present invention except for additionally adding the organic binder to the porous adhesive layer. Accordingly, the additionally added binder other than the acryl-based copolymer may be mainly described hereinafter. The addition of the binder in the present example embodiment may further improve adherence and heat resistance.

[0039] Examples of an additional binder in addition to acryl-based copolymer may be one or mixture thereof selected from a polyvinylidene fluoride (PVdF) homopolymer, a polyvinylidene fluoride-hexafluoropropylene copolymer (PVdF-HFP), polymethylmethacrylate, polyacrylonitrile, polyvinylpyrrolidone, polyvinylacetate, polyethylene oxide, cellulose acetate, cellulose acetate butyrate, cellulose acetate propionate, cyanoethylpullulan, cyanoethylpolyvinyl alcohol, cyanoethyl cellulose, cyanoethylsucrose, pullulan, carboxyl methyl cellulose, and an acrylonitrile styrene butadiene copolymer. More specifically, a polyvinylidene fluoride-based binder may be used and examples thereof may be a polyvinylidene fluoride (PVDF) homopolymer, polyvinylidene fluoride-hexafluoropropylene (PVDF-HFP), polyvinylidene fluoride-trichloroethylene (PVDF-TCE), polyvinylidene fluoride-chlorotrifluoroethylene (PVDF-CTFE), and the like.

[0040] A weight ratio of the acryl-based copolymer and the additional binder may be 9.9:0.1 to 2.5:7.5. Specifically, it may be 9.9:0.1 to 5:5 and more specifically, 9:1 to 5.5:4.5, or 8:2 to 6:4. Within the range, the separator may provide an electrode assembly having excellent shape stability as well as maintaining sufficient adherence. Accordingly, the electrode assembly may prevent performance deterioration of a battery, and the battery may have highly efficient charge and discharge characteristics.

[0041] When the PVdF-based binder is further included, the PVdF-based binder may have a weight average molecu-

lar weight (Mw) of 500,000 to 1,500,000 (g/mol). For specific example, the PVdF-based binder may have a weight average molecular weight (Mw) of 1,000,000 to 1,500,000 (g/mol). For another example, two or more binders having different weight average molecular weights may be mixed. For example, a binder having a weight average molecular weight of less than or equal to 1,000,000 g/mol and a binder having a weight average molecular weight of greater than or equal to 1,000,000 g/mol may be mixed. When the PVdF-based binder having the molecular weight within the range is used, adherence between the porous adhesive layer and the porous substrate is fortified, a porous substrate that is weak against heat may be effectively prevented from being contracted, a separator having sufficiently improved electrolyte impregnation properties may be prepared, and a battery effectively generating electrical output may be manufactured.

[0042] Hereinafter, a method of preparing an electrode assembly according to an example embodiment of the present invention is described. The method of preparing an electrode assembly according to an example embodiment of the present invention may include manufacturing a cathode by forming a cathode active material layer on a cathode current collector, manufacturing an anode by forming an anode active material layer on an anode current collector, and disposing the separator according to the present invention between the cathode and the anode.

[0043] According to another example embodiment of the present invention, a method of preparing an electrode assembly may additionally include compressing a structure of the cathode/the separator/the anode at 20° C. to 110° C. for 1 second to 10 seconds with a pressure of 1 kgf/cm² to 30 kgf/cm² after disposing the separator between the cathode and the anode. When the separator manufactured in the above method is compressed with the cathode and the anode at 20° C. to 110° C. for 1 second to 10 seconds with a pressure of 1 kgf/cm² to 30 kgf/cm² after disposing the separator between the cathode and the anode, the acryl-based copolymer of the present invention may form strong adherence of the separator to the cathode or the anode and improve shape storage stability of the electrode assembly. The compression may be performed at a temperature determined considering a temperature where the porous substrate of the separator is not remarkably thermally shrunk and a temperature where the porous adhesive layer of the separator is adhered and specifically, at room temperature or 80° C. to 100° C. for 1 second to 5 seconds with a pressure of 5 kgf/cm² to 10 kgf/cm².

[0044] In addition, a method of manufacturing an electrode assembly according to still another example embodiment of the present invention may additionally include secondarily compressing the electrode assembly at 60° C. to 110° C. for 30 seconds to 180 seconds with a pressure of 1 kgf/cm² to 30 kgf/cm² after disposing the separator between cathode and anode, primarily compressing the electrode assembly at 20° C. to 110° C. for 1 second to 10 seconds with a pressure of 1 kgf/cm² to 30 kgf/cm², housing it in a battery case, and injecting an electrolyte solution into a battery case. Herein, the battery case may be an aluminum pouch, and the like but is not limited thereto.

[0045] In addition, a method of manufacturing an electrode assembly according to still another example embodiment of the present invention may additionally include storing the electrode assembly for 6 hours to 48 hours at 10°

C. to 30° C. after injecting the electrolyte solution and before secondarily compressing the electrode assembly. The secondary compression may form stronger adherence of the acryl-based copolymer of the present invention to the cathode or the anode and improve shape storage stability of the electrode assembly.

[0046] The cathode includes include a cathode current collector and a positive active material layer formed on the cathode current collector. The positive active material layer includes a positive active material, a binder, and optionally a conductive material. The cathode current collector may use aluminum (Al), nickel (Ni), and the like, but is not limited thereto. The positive active material may use a compound being capable of intercalating and deintercalating lithium. Specifically at least one of a composite oxide or a composite phosphate of a metal selected from cobalt, manganese, nickel, aluminum, iron, or a combination thereof and lithium may be used. More specifically, the positive active material may use lithium cobalt oxide, lithium nickel oxide, lithium manganese oxide, lithium nickel cobalt manganese oxide, lithium nickel cobalt aluminum oxide, lithium iron phosphate, or a combination thereof. The binder improves binding properties of positive active material particles with one another and with a current collector, and specific examples may be polyvinyl alcohol, carboxymethyl cellulose, hydroxypropyl cellulose, diacetyl cellulose, polyvinylchloride, carboxylated polyvinylchloride, polyvinylfluoride, an ethylene oxide-containing polymer, polyvinylpyrrolidone, polyurethane, polytetrafluoroethylene, polyvinylidene fluoride, polyethylene, polypropylene, a styrene-butadiene rubber, an acrylated styrene-butadiene rubber, an epoxy resin, nylon, and the like, but are not limited thereto. These may be used alone or as a mixture of two or more. The conductive material improves conductivity of an electrode and examples thereof may be natural graphite, artificial graphite, carbon black, a carbon fiber, a metal powder, a metal fiber, and the like, but are not limited thereto. These may be used alone or as a mixture of two or more. The metal powder and the metal fiber may use a metal of copper, nickel, aluminum, silver, and the like.

[0047] The anode includes a anode current collector and an anode active material layer formed on the anode current collector. The anode current collector may use copper (Cu), gold (Au), nickel (Ni), a copper alloy, and the like, but is not limited thereto. The anode active material layer may include an anode active material, a binder and optionally a conductive material. The anode active material may be a material that reversibly intercalates/deintercalates lithium ions, a lithium metal, a lithium metal alloy, a material being capable of doping and dedoping lithium, a transition metal oxide, or a combination thereof. The material that reversibly intercalates/deintercalates lithium ions may be a carbon material which is any generally-used carbon-based anode active material, and examples thereof may be crystalline carbon, amorphous carbon, or a combination thereof. Examples of the crystalline carbon may be graphite such as amorphous shape, plate shape, flake shape, spherical shape or fiber shape natural graphite or artificial graphite. Examples of the amorphous carbon may be soft carbon or hard carbon, a mesophase pitch carbonized product, fired coke, and the like. The lithium metal alloy may be an alloy of lithium and a metal selected from Na, K, Rb, Cs, Fr, Be, Mg, Ca, Sr, Si, Sb, Pb, In, Zn, Ba, Ra, Ge, Al, and Sn. The material being capable of doping and dedoping lithium may be Si, SiO_x

(0<x<2), a Si—C composite, a Si—Y alloy, Sn, SnO₂, a Sn—C composite, a Sn—Y, and the like, and at least one of these may be mixed with SiO₂. Specific examples of the element Y may be selected from Mg, Ca, Sr, Ba, Ra, Sc, Y, Ti, Zr, Hf, Rf, V, Nb, Ta, Db, Cr, Mo, W, Sg, Tc, Re, Bh, Fe, Pb, Ru, Os, Hs, Rh, Ir, Pd, Pt, Cu, Ag, Au, Zn, Cd, B, Al, Ga, Sn, In, Tl, Ge, P, As, Sb, Bi, S, Se, Te, Po, and a combination thereof. The transition metal oxide may be vanadium oxide, lithium vanadium oxide, and the like. The binder and the conductive material used in the anode may be the same as the binder and conductive material of the cathode.

[0048] The cathode and the anode may be manufactured by mixing each active material composition including each active material and a binder, and optionally a conductive material in a solvent, and coating the active material composition on each current collector. Herein, the solvent may be N-methylpyrrolidone, and the like, but is not limited thereto.

[0049] The electrolyte solution may include a salt having a structure of A⁺B⁻ dissolved or dissociated in an organic solvent.

[0050] The organic solvent serves as a medium for transmitting ions taking part in the electrochemical reaction of a battery. Specific examples thereof may be selected from a carbonate-based solvent, an ester-based solvent, an ether-based solvent, a ketone-based solvent, an alcohol-based solvent, and an aprotic solvent. Examples of the carbonate-based solvent may be dimethyl carbonate (DMC), diethyl carbonate (DEC), dipropyl carbonate (DPC), methylpropyl carbonate (MPC), ethylpropyl carbonate (EPC), ethylmethyl carbonate (EMC), ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), and the like. Particularly, when the linear carbonate compounds and cyclic carbonate compounds are mixed, an organic solvent having a high dielectric constant and a low viscosity may be provided. The cyclic carbonate compound and the linear carbonate compound are mixed together in a volume ratio ranging from 1:1 to 1:9. Examples of the ester-based solvent may be methylacetate, ethylacetate, n-propylacetate, dimethylacetate, methylpropionate, ethylpropionate, γ-butyrolactone, decanolide, valerolactone, mevalonolactone, caprolactone, and the like. Examples of the ether-based solvent may be dibutylether, tetraglyme, diglyme, dimethoxyethane, 2-methyltetrahydrofuran, tetrahydrofuran, and the like. Examples of the ketone-based solvent may be cyclohexanone, and the like, and examples of the alcohol-based solvent may be ethanol, isopropyl alcohol, and the like. The organic solvent may be used singularly or in a mixture of two or more, and when the organic solvent is used in a mixture of two or more, the mixture ratio may be controlled in accordance with a desirable cell performance.

[0051] Non-limiting examples of the A⁺ may be a cation of an alkali metal cation such as Li⁺, Na⁺, or K⁺, or a combination thereof. Non-limiting examples of the B⁻ may be an anion of PF₆⁻, SbF₆⁻, AlCl₄⁻, AlO₂⁻, Cl⁻, Br⁻, I⁻, ClO₄⁻, AsF₆⁻, B(C₂O₄)₂⁻, CH₃CO₂⁻, N(SO₃C₂F₅)₂⁻, C₄F₉SO₃⁻, CF₃SO₃⁻, N(CF₃SO₂)₂⁻, or C(CF₂SO₂)₃⁻, or a combination thereof. For example, a lithium salt may be used, and the lithium salt supplies lithium ions in a battery, basically operates an electrochemical battery, and improves lithium ion transportation between positive and anodes therein. Examples of the lithium salt may be LiPF₆, LiBF₄, LiSbF₆, LiAsF₆, LiN(SO₃C₂F₅)₂, LiN(CF₃SO₂)₂, LiC₄F₉SO₃, LiClO₄, LiAlO₂, LiAlCl₄, LiN(C_xF_{2x+1}SO₂)

($C_xF_{2y+1}SO_2$), x and y are natural numbers, LiCl, LiI, $LiB(C_2O_4)_2$, or a combination thereof. The lithium salt may be used in a concentration ranging from 0.1 M to 2.0 M. When the lithium salt is included within the above concentration range, an electrolyte may have excellent performance and lithium ion mobility due to optimal electrolyte conductivity and viscosity.

[0052] The electrochemical battery according to an example embodiment of the present invention may be specifically a lithium secondary battery such as a lithium metal secondary battery, a lithium ion secondary battery, a lithium polymer secondary battery, or a lithium ion polymer secondary battery.

DETAILED DESCRIPTION

[0053] Hereinafter, Examples, Comparative Examples and Experimental Examples are provided in order to illustrate the present invention in detail. However, the following Examples, Comparative Examples, and Experimental Examples are examples of the present invention and are not to be construed as limiting the present invention.

PREPARATION EXAMPLE

Preparation Example 1

Preparation of Separator

[0054] A first binder solution was prepared by polymerizing butyl methacrylate (BMA), methyl methacrylate (MMA), and vinyl acetate (VAc) in a mole ratio of 4/1/5 to obtain an acryl-based copolymer binder (Tg: 35° C., Mw: 600K (GPC)), dissolving the acryl-based copolymer binder in a solid amount of 10 wt % in acetone, and stirring the solution with an agitator at 40° C. for 2 hours. On the other hand, an alumina dispersion liquid was prepared by adding alumina (LS235, Nippon Light Metal Co., Ltd.) in an amount of 25 wt % to acetone and dispersing it therein at 25° C. for 2 hours with a bead mill. The first binder solution and the alumina dispersion liquid were mixed in 1/5 of a ratio between the binder solid and alumina solid, and acetone was added to the mixture to be an entire solid of 10 wt %, preparing a porous adhesive layer composition. The porous adhesive layer composition was coated to be respectively 2 μ m thick on both surfaces of a 12 μ m-thick polyethylene fabric panel (W scope) to manufacture a separator having a total thickness of 16 μ m.

Preparation Example 2

Preparation of Separator

[0055] A first binder solution was prepared by polymerizing butyl methacrylate (BMA), methyl methacrylate (MMA), and vinyl acetate (VAc) in a mole ratio of 4/1/5 to prepare an acryl-based copolymer binder (Tg: 35° C., Mw: 600K (GPC)), dissolving the acryl-based copolymer binder in a solid amount of 10 wt % in acetone, and stirring the solution with an agitator at 40° C. for 2 hours. A second binder solution was prepared by dissolving KF9300 (Kureha Corp., Mw: 1,200,000 g/mol) as a PVdF-based binder in a solid amount of 7 wt % in a mixed solvent of acetone and DMAc and stirring the solution at 40° C. for 4 hours with an agitator. On the other hand, an alumina dispersion liquid was prepared by adding alumina (LS235, Nippon Light Metal

Co., Ltd.) in an amount of 25 wt % in acetone and dispersing it therein at 25° C. for 2 hours with a bead mill. The first and second binder solutions and the alumina dispersion liquid were mixed to have 8/2 of a weight ratio between the acryl-based binder and the PVdF-based binder and 1/5 of a ratio between the binder solid and the alumina solid, and acetone was added thereto to prepare a porous adhesive layer composition to have an entire solid of 10 wt %. The porous adhesive layer composition was coated to be respectively on 2 μ m thick both surfaces of a 12 μ m-thick polyethylene fabric panel (W scope), manufacturing a separator having a total thickness of 16 μ m.

Preparation Example 3

Preparation of Separator

[0056] A separator was manufactured according to the same method as Preparation Example 2 except for using the acryl-based binder and the PVdF-based binder in a weight ratio of 7/3.

Preparation Example 4

Preparation of Separator

[0057] A separator was manufactured according to the same method as Preparation Example 2 except for using the acryl-based binder and the PVdF-based binder in a weight ratio of 6/4.

Preparation Example 5

Preparation of Separator

[0058] A separator was manufactured according to the same method as Preparation Example 2 except for using the acryl-based binder and the PVdF-based binder in a weight ratio of 3/7.

Preparation Example 6

Preparation of Separator

[0059] A separator was manufactured according to the same method as Preparation Example 1 except for coating the porous adhesive layer composition to be 2 μ m thick on one surface of a polyethylene fabric panel to have the total thickness of the separator of 14 μ m.

Comparative Preparation Example 1

Preparation of Separator

[0060] A separator was manufactured according to the same method as Preparation Example 2 except for mixing the acryl-based binder and the PVdF-based binder in a weight ratio of 1/9.

Comparative Preparation Example 2

Preparation of Separator

[0061] A separator was manufactured according to the same method as Preparation Example 2 except for mixing the acryl-based binder and the PVdF-based binder in a weight ratio of 0.5/9.5.

Comparative Preparation Example 3

Preparation of Separator

[0062] A separator was manufactured according to the same method as Preparation Example 2 except for using only the PVdF-based binder.

[0063] Each binder composition of the separators according to Preparation Examples 1 to 6 and Comparative Preparation Examples 1 to 3 are provided in Table 1.

TABLE 1

	Acryl-based binder	PVdF-based binder
Preparation Example 1	100	0
Preparation Example 2	80	20
Preparation Example 3	70	30
Preparation Example 4	60	40
Preparation Example 5	30	70
Preparation Example 6	100	0
Comparative Preparation Example 1	10	90
Comparative Preparation Example 2	5	95
Comparative Preparation Example 3	0	100

EXAMPLES

Example 1

Preparation of Electrode Assembly

[0064] A positive active material coating composition was prepared by using LCO (LiCoO_2) as a positive active material, PVdF (polyvinylidene fluoride) as a binder, and carbon black as a conductive agent. The positive active material coating composition was prepared by dispersing an active material, a binder, and a conductive material in a weight ratio of 94:3:3 in N-methyl-2-pyrrolidone with a mixer (Planetary Despa Mixer) into slurry and coating the slurry to be 94 μm thick on both surfaces of a 14 μm -thick aluminum foil with a doctor blade and drying it. Then, the coated foil was pressed with a roll presser and dried with a vacuum dryer to remove moisture in a coating layer, manufacturing a cathode. On the other hand, an anode active material coating composition was prepared by using graphite as an anode active material and SBR (styrene-butadiene rubber) and CMC (carboxy methyl cellulose) as a binder. Herein, the anode active material and the binder were used in a weight ratio of 96:4, and the SBR and the CMC were used in a weight ratio of 1:1. Then, an anode was manufactured according to the same method as the cathode except for forming a 120 μm -thick coating layer on both surfaces of a 8 μm -thick copper foil. The cathode and the anode were respectively cut into a size of 100 cm \times 4.2 cm, and the separator according to Preparation Example 1 was cut into a size of 100 cm \times 4.4 cm and then, disposed between the cathode and the anode and wound therewith into a size 7 cm (a length direction) \times 4.4 cm (a width direction), manufacturing an electrode assembly.

Example 2

Preparation of Electrode Assembly

[0065] An electrode assembly according to Example 2 was manufactured according to the same method as Example 1 except for using the separator according to Preparation Example 2.

Example 3

Preparation of Electrode Assembly

[0066] An electrode assembly according to Example 3 was manufactured according to the same method as Example 1 except for using the separator according to Preparation Example 3.

Example 4

Preparation of Electrode Assembly

[0067] An electrode assembly according to Example 4 was manufactured according to the same method as Example 1 except for using the separator according to Preparation Example 4.

Example 5

Preparation of Electrode Assembly

[0068] An electrode assembly according to Example 5 was manufactured according to the same method as Example 1 except for using the separator according to Preparation Example 5.

Example 6

Preparation of Electrode Assembly

[0069] An electrode assembly according to Example 6 was manufactured according to the same method as Example 1 except for using the separator according to Preparation Example 6, positioning a cathode to face the surface of the separator having a porous adhesive layer and an anode to face the other surface of the separator having no porous adhesive layer.

Comparative Example 1

Preparation of Electrode Assembly

[0070] An electrode assembly according to Comparative Example 1 was manufactured according to the same method as Example 1 except for using the separator according to Comparative Preparation Example 1.

Comparative Example 2

Preparation of Electrode Assembly

[0071] An electrode assembly according to Comparative Example 2 was manufactured according to the same method as Example 1 except for using the separator according to Comparative Preparation Example 2.

Comparative Example 3

Preparation of Electrode Assembly

[0072] An electrode assembly according to Comparative Example 3 was manufactured according to the same method as Example 1 except for using the separator according to Comparative Preparation Example 3.

EXPERIMENTAL EXAMPLE

[0073] Each flexural rigidity and compression thickness variation ratio of the electrode assemblies according to

Examples 1 to 6 and Comparative Examples 1 to 3 was measured in the following method, and the results are provided in Table 2.

[0074] Flexural Rigidity

[0075] The electrode assemblies according to Examples 1 to 6 and Comparative Examples 1 to 3 were respectively compressed for 10 seconds under a pressure of 9 kgf/cm² at 80° C., 90° C., 100° C., and 110° C., respectively. Subsequently, flexural rigidity of each electrode assemblies was put in an MD direction toward a right-left direction in a distance of 60 mm and measured according to ASTM D790 with a 3 point bending machine (UTM), while dropped at a speed of 2.8 mm/min.

[0076] Compression Thickness Variation Ratio

[0077] Each electrode assembly according to Examples 1 to 6 and Comparative Examples 1 to 3 was compressed at 20° C. under a pressure of 9 kgf/cm² for 3 seconds, and one hour later, its thickness in the middle was measured with a 15 cm steel ruler. Each electrode assembly was also compressed at 100° C. under a pressure of 9 kgf/cm² for 10 seconds, and one hour later, its thickness in the middle was measured with a 15 cm steel ruler. The thicknesses compressed at 20° C. and 100° C. were used to calculate a compression thickness variation ratio through the following formula.

$$\begin{aligned} \text{A compression thickness variation ratio (\%)} = & \\ & [(\text{Thickness of an electrode assembly com-} \\ & \text{pressed at 20° C.} - \text{Thickness of an electrode} \\ & \text{assembly compressed at 100° C.}) / \text{Thickness of} \\ & \text{an electrode assembly compressed at 20°} \\ & \text{C}] \times 100 \end{aligned}$$

TABLE 2

	Flexural rigidity depending on a press				Compression thickness (mm) and compression thickness variation ratio (%)		
	temperature (kgf/cm ²)				20°	100°	Variation ratio (%)
	80° C.	90° C.	100° C.	110° C.	C. press	C. press	
Example 1	43	47	50	50	6.5	5	23
Example 2	41	44	47	48	6.5	5	23
Example 3	36	39	40	40	6.5	5	23
Example 4	17	20	23	24	6.5	5.5	15.38
Example 5	15	20	23	25	6.5	5.5	15.38
Example 6	39	40	42	43	6.5	5	23
Comparative Example 1	7	8	9	10	6.5	6.0	7.69
Comparative Example 2	3	5	6	6	6.5	6.0	7.69
Comparative Example 3	(no flexural point, non-measurable)				6.5	6.0	7.69

[0078] Thickness Variation Ratio after Cycles

[0079] The electrode assemblies according to Examples 2 and 3 and Comparative Example 3 were manufactured in the following method, their thickness variation ratios after cycles were measured, and the results are provided in Table 3.

[0080] Each electrode assembly according to Examples 2 and 3 and Comparative Example 3 was compressed at 100° C. for 3 seconds under a pressure of 9 kgf/cm² and put in an aluminum pouch, an electrolyte solution was injected thereinto, and the pouch was sealed. Herein, the electrolyte solution was prepared by dissolving 1.1 M LiPF₆ in 2.7 g of

an organic solvent of ethylene carbonate (EC)/ethylmethyl carbonate (EMC) in a EC:EMC volume ratio of 30/70. Then, the pouched electrode assembly was stored at room temperature for 12 hours, compressed at 100° C. for 30 seconds under a pressure of 9 kgf/cm², and then, stored at room temperature for 12 hours. Subsequently the pouched electrode assembly was precharged under a condition of 0.2 C for one hour in a charge/discharger, and its thickness of the electrode assembly in the middle was measured with a 15 cm steel ruler. Then, the pouch was open to remove gas, charge and discharge were 500 times repeated under a condition of 0.7 C, and the thickness of the electrode assembly in the middle was measured.

TABLE 3

	Thickness before pre- charging (mm)	Thickness after 500 cycles (mm)	Thickness variation ratio after cycling (%)
Example 2	3.0	3.5	16.7
Example 3	3.0	3.5	16.7
Comparative Example 3	3.0	4.0	33.3

[0081] Referring to Tables 2 and 3, when an acryl-based copolymer was used alone or mixed with the polyvinylidene fluoride-based binder in a weight ratio of 9.9:0.1 to 2.5:7.5, a compression thickness variation ratio of greater than or equal to 10% and thus excellent adherence of electrodes to a separator was found. In addition, flexural rigidity of greater than or equal to 15 kgf/cm² and thus excellent shape stability of an electrode assembly was found.

[0082] Accordingly, a separator may be prevented from an escape and thus may decrease an inferiority rate in an electrode assembly process and accomplish a long time storage. The electrode assembly may have a minimized from a shape change during charges and discharges repeated for a long time. This is supported by a thickness variation ratio of less than or equal to 20% after cycles. Accordingly, a battery cell using the electrode assembly may have highly efficient charge and discharge characteristics and be prevented from deterioration of battery performance.

What is claimed is:

1. An electrode assembly comprising a cathode including a positive active material and a cathode current collector, an anode including an anode active material and a anode current collector, and a separator disposed between the cathode and the anode, wherein the electrode assembly has a flexural rigidity of greater than or equal to 15 kgf/cm² when being compressed at 20° C. to 110° C. for 1 second to 15 seconds, with a pressure of 1 kgf/cm² to 30 kgf/cm².

2. The electrode assembly of claim 1, wherein the separator comprises a porous substrate and a porous adhesive layer disposed on one surface or both surfaces of the porous substrate and including an acryl-based copolymer including a(meth)acrylate-based monomer-derived repeating unit.

3. The electrode assembly of claim 2, wherein the acryl-based copolymer further comprises an acetate group-containing monomer-derived repeating unit.

4. The electrode assembly of claim 2, wherein the (meth)acrylate-based monomer-derived repeating unit is a repeating unit derived from at least one monomer selected from the group consisting of methyl(meth)acrylate, ethyl(meth)acrylate, propyl(meth)acrylate, and butyl(meth)acrylate.

5. The electrode assembly of claim 3, wherein the acetate group-containing monomer-derived repeating unit is a repeating unit derived from allyl acetate or vinyl acetate.

6. The electrode assembly of claim 2, wherein the porous adhesive layer further comprises an inorganic particle, and the inorganic particle is included in an amount of 70 wt % to 95 wt % based on the total weight of the porous adhesive layer.

7. The electrode assembly of claim 2, wherein the porous adhesive layer further comprises a polyvinylidene fluoride-based binder.

8. The electrode assembly of claim 7, wherein the polyvinylidene fluoride-based binder is at least one selected from a polyvinylidene fluoride (PVDF) homopolymer, polyvinylidene fluoride-hexafluoropropylene (PVDF-HFP), polyvinylidene fluoride-trichloroethylene (PVDF-TCE), and polyvinylidene fluoride-chlorotrifluoroethylene (PVDF-CTFE).

9. The electrode assembly of claim 7, wherein a weight ratio of the acryl-based copolymer and the polyvinylidene fluoride-based binder is 9.9:0.1 to 2.5:7.5.

10. The electrode assembly of claim 1, wherein the electrode assembly has a compression thickness variation ratio of greater than or equal to 10% according to Equation 1:

$$\text{Compression thickness variation ratio (\%)} = \left[\frac{\text{Thickness of an electrode assembly compressed at } 20^{\circ} \text{ C.} - \text{Thickness of an electrode assembly compressed at } 100^{\circ} \text{ C.}}{\text{Thickness of an electrode assembly compressed at } 20^{\circ} \text{ C.}} \right] \times 100 \quad [\text{Equation 1}]$$

wherein, in Equation 1, the thickness of an electrode assembly compressed at 20° C. is a thickness of a central portion of an electrode assembly including a stacked cathode/separator/anode after compressing it at

20° C. for 1 second to 10 seconds, with a pressure of 1 kgf/cm² to 30 kgf/cm² and measuring the thickness in one 1 hour, and the thickness of an electrode assembly compressed at 100° C. is a thickness of a central portion of the electrode assembly for 1 second to 10 seconds, with a pressure of 1 kgf/cm² to 30 kgf/cm² and measuring the thickness in one 1 hour.

11. An electrochemical battery comprising the electrode assembly of claim 1.

12. The electrochemical battery of claim 11, wherein the electrochemical battery is a lithium polymer secondary battery or a lithium ion polymer secondary battery.

13. A method of preparing an electrode assembly, comprising forming a positive active material layer on a cathode current collector to prepare a cathode, forming an anode active material layer on an anode current collector to prepare an anode, disposing a separator between the cathode and the anode, and compressing the cathode/separator/anode structure at 20° C. to 110° C. for 1 second to 10 seconds with 1 kgf/cm² to 30 kgf/cm².

14. The method of claim 13, wherein the method further comprises secondarily compressing the structure at 60° C. to 110° C. for 30 seconds to 180 seconds, with 1 kgf/cm² to 30 kgf/cm² after the compressing the structure and injecting an electrolyte.

15. The method of claim 13, wherein the separator comprises a porous substrate and a porous adhesive layer disposed on one surface or both surfaces of the porous substrate and including an acryl-based copolymer including a (meth)acrylate-based monomer-derived repeating unit.

16. The method of claim 15, wherein the acryl-based copolymer further comprises an acetate group-containing monomer-derived repeating unit.

* * * * *