BINDER COMPOSITION FOR SECONDARY BATTERY

FIELD OF THE INVENTION

[001] The present invention relates to the field of batteries. In particular, this invention relates to binder compositions that can be used in electrode mixtures for lithium-ion batteries and other metal-ion batteries.

BACKGROUND OF THE INVENTION

[002] Over the past decades, lithium-ion batteries (LIBs) have become widely utilized in various applications, especially consumer electronics, because of their outstanding energy density, long cycle life and high discharging capability. Due to rapid market development of electric vehicles (EV) and grid energy storage, high-performance, low-cost LIBs are currently offering one of the most promising options for large-scale energy storage devices.

[003] Generally, a lithium-ion battery electrode comprises an electrode layer adhered to a current collector, wherein the electrode layer comprises various electrode components such as an electrode active material, conductive material, and a binder material. The binder material provides a good electrochemical stability of the electrode layer, holds together the electrode components and adheres the electrode components to the current collector. A slurry is often used to form an electrode layer, wherein a solvent is used to suspend or dissolve the various electrode components for easy processing and coating onto the current collector. Polyvinylidene fluoride (PVDF) is one of the most commonly used binder materials in the commercial lithium-ion battery industry, but PVDF can only dissolve in some specific organic solvents such as N-methyl-2-pyrrolidone (NMP). Accordingly, these organic solvents would be used as the solvent in electrode slurries comprising PVDF. However, NMP is flammable and toxic and hence requires specific handling. An NMP recovery system must be in place during the drying process to recover NMP vapors. This will generate significant costs in the manufacturing process since a large capital investment is required.

[004] Considering the drawbacks of using organic solvent-based slurries to form electrodes, water-based slurries have been considered instead, wherein an aqueous solvent, most commonly water, is used in the slurry. A different polymer, one that is water-compatible in nature, must then be used as the binder material instead of PVDF because PVDF is insoluble and disperse poorly in water. Such water-based slurries would also often require a large amount of solvent in order to ensure good dispersion of the electrode components within, of which good dispersion of the electrode components is necessary to ensure good electrode performance. The presence of water however could also lead to degradation of cathode active materials, leading to

worsened battery performance.

[005] In light of the above, electrode material mixtures with much lower liquid contents have been devised. One common technique for electrode layer formation without the use of a solvent is polymer fibrillization, in which a polymer that can be fibrillized, such as polytetrafluoroethylene (PTFE), alone or in a composition containing it, are put under a high shear force together with other electrode components such as through jet-milling, calendaring, and extrusion. Through the application of the shear force, the polymer fibrillizes and forms a matrix which entraps and supports the other electrode components, thus functioning as a binder. US Patent No. 10,547,057 discloses an example of using polymer fibrillization in an electrode material mixture which is essentially free of water. A dry mixture of electrode active material, conductive agent, and PTFE as the binder material are fed through a nozzle by compressed air into the grinding chamber of a jet-mill. It was taught that collision among particles create a shearing force which causes the PTFE to fibrillize, leading to the electrode active material and conductive agent being embedded within the fibrous matrix of the PTFE, while at the same time the electrode active material and conductive agent particles also prevents the PTFE from re-agglomerating to a certain extent. However, PTFE forms strong van der Waals interactions with carbon-based conductive agents, which would reduce the effectiveness of PTFE fibrillization. Furthermore, as a fluorinated polymer, even if no solvent was used in the electrode manufacturing process, a toxic organic solvent or a toxic-chemical-releasing thermal process would have to be used in recycling of the electrode in order to recover the various electrode components which means significant environmental issues remain. In addition, equipment used to create the high shear force required for polymer fibrillization requires a large amount of energy to operate and is therefore expensive.

[006] Accordingly, the present inventors have intensively studied on the subject, and have found that a novel water-compatible copolymer disclosed herein can be effectively used as a binder material in an electrode material mixture with a specific and reduced aqueous solvent content, where the copolymer is readily fibrillized. Being water-compatible in nature, the copolymer can be readily recycled without having to resort to environmentally damaging methods. Surprisingly, and most significantly, using the copolymer to manufacture electrodes allows the electrode to be made thicker than conventional electrode material mixtures, thereby allowing for higher battery capacities.

SUMMARY OF THE INVENTION

[007] The aforementioned needs are met by various aspects and embodiments disclosed herein. In one aspect, provided herein is a binder composition comprising a copolymer as the binder material and a solvent; wherein the copolymer comprises a hydrophilic structural unit and a non-hydrophilic structural unit; and wherein the copolymer in the binder composition can be

fibrillized in the presence of a shear force. The solvent is water, while the copolymer is water-compatible in nature. In some embodiments, the proportion of the hydrophilic structural unit in the copolymer is from about 60% to about 90% by mole, based on the total number of moles of monomeric units in the copolymer. In certain embodiments, the weight-average molecular weight of the copolymer is from about 400,000 g/mol to about 700,000 g/mol. In certain embodiments, the solid content of the binder composition is from about 5% to about 30% by weight, based on the total weight of the binder composition.

[008] In another aspect, provided herein is a particularly illustrative embodiment of the binder composition of the present invention, wherein the hydrophilic structural unit is derived from an acid group-containing monomer and an amide group-containing monomer, and wherein the non-hydrophilic structural unit is derived from a nitrile group-containing monomer. In some embodiments, the proportion of monomeric units in the copolymer derived from an acid group-containing monomer is from about 10% to about 40% by mole, while the proportion of monomeric units in the copolymer derived from an amide group-containing monomer is from about 10% to about 55% by mole, based on the total number of moles of monomeric units in the copolymer.

[009] In a further aspect, an electrode material mixture utilizing the binder composition of the present invention is disclosed, wherein the electrode material mixture has a liquid content of from about 5% to about 22% by weight, based on the total weight of the electrode material mixture.

[0010] The copolymer in the binder composition can be readily fibrillized, meaning that the electrode components in an electrode layer manufactured using an electrode material mixture incorporating the binder composition of the present invention can be well-dispersed and adhere strongly to the current collector. As a result, the electrode layer could be made thicker, and batteries comprising such an electrode layer would have improved energy density. Following end-of-use, the copolymer can also be readily recycled without having to resort to environmentally damaging methods.

DETAILED DESCRIPTION OF THE INVENTION

[0011] The term “electrode” refers to a “cathode” or an “anode”.

[0012] The term “positive electrode” is used interchangeably with cathode. Likewise, the term “negative electrode” is used interchangeably with anode.

[0013] The term “current collector” refers to any conductive substrate that is in contact with an electrode layer and is capable of conducting an electrical current flowing to electrodes during discharging or charging a secondary battery. Some non-limiting examples of the current collector include a single conductive metal layer or substrate, and a single conductive metal

layer or substrate with an overlying conductive coating layer, such as a carbon black-based coating layer. The conductive metal layer or substrate may be in the form of a foil or a porous body having a three-dimensional network structure, and may be a polymeric or metallic material or a metalized polymer. In some embodiments, the three-dimensional porous current collector is covered with a conformal carbon layer.

[0014] The term "electrode layer" refers to a layer that is in contact with a current collector and comprises an electrochemically active material. In some embodiments, an electrode is made by applying an electrode layer on to a current collector. In some embodiments, an electrode layer is located on a surface of a current collector. In other embodiments, the three-dimensional porous current collector is coated conformally with an electrode layer.

[0015] The term "water-compatible" with respect to a chemical compound, mixture of compounds, or polymer refers to the ability of the chemical compound, mixture of compounds, or polymer to be well dispersed in an aqueous solvent to form a solution or colloid.

[0016] The term "binder material" refers to a chemical compound, mixture of compounds, or polymer that is used to hold a material in place and adhere the material onto a substrate. In some embodiments, a binder material is used to hold the electrode components in place and adhere them to a conductive metal part to form an electrode. In some embodiments, the binder material comprises a polymer. Such a polymer can be termed a "binder polymer". In some embodiments, the binder material is accompanied by a liquid, and which can be collectively known as a "binder composition". In certain embodiments, this liquid is water. In some embodiments, the binder material is dry or substantially free of liquid. In other embodiments, the binder material does not comprise liquid.

[0017] The term "dry" in the context of a mixture means that the mixture is substantially free of or does not comprise liquid. The term "substantially free of liquid" in the context of a mixture means that the mixture has a very low liquid content. In certain embodiments, "substantially free of liquid" means that the mixture has a liquid content of less than 1%, less than 0.8%, less than 0.6%, less than 0.5%, less than 0.4%, less than 0.3%, less than 0.2%, less than 0.15%, less than 0.1%, less than 0.05%. less than 0.03%, less than 0.02%, less than 0.015%, less than 0.01%, less than 0.1%, less than 0.075%, less than 0.05%, less than 0.025%, less than 0.02%, less than 0.015%, less than 0.01%, less than 0.0075%, less than 0.005%, less than 0.0025%, less than 0.002%, less than 0.0015%, or less than 0.001% by weight, based on the total weight of the mixture.

[0018] The term "conductive material" or "conductive agent" refers to a material that has good electrical conductivity. Therefore, a conductive agent is often mixed with an electrode active material at the time of forming an electrode to improve electrical conductivity of the electrode. In some embodiments, the conductive agent is chemically active. In some

embodiments, the conductive agent is chemically inactive.

[0019] The term "electrode material mixture" refers to a mixture of materials that can be used to form an electrode layer. In some embodiments, an electrode material mixture comprises an electrode active material and a binder material. In some embodiments, an electrode material mixture further comprises a conductive agent. In certain embodiments, an electrode material mixture further comprises a solvent, whereby the electrode material mixture can be termed an "electrode slurry".

[0020] With respect to a slurry, the term "water-based" means that the solvent of the slurry is an aqueous solvent. The term "aqueous solvent" refers to a solution containing water as the major component and one or more minor components in addition to water, or a solution that consists solely of water.

[0021] The term "unsaturated" as used herein refers to a moiety having one or more units of unsaturation.

[0022] The term "alkyl" or "alkyl group" refers to a univalent group having the general formula C_nH_{2n+1} and derived from the removal of a hydrogen atom from a saturated, unbranched or branched aliphatic hydrocarbon, where n is an integer.

[0023] The term "cycloalkyl" or "cycloalkyl group" refers to a saturated or unsaturated cyclic non-aromatic hydrocarbon radical having a single ring or multiple condensed rings. A cycloalkyl group can be unsubstituted or substituted by one or two suitable substituents. Furthermore, the cycloalkyl group can be monocyclic or polycyclic.

[0024] The term "alkenyl" refers to a univalent group derived from the removal of a hydrogen atom from any carbon atom of an unsaturated aliphatic hydrocarbon with at least one carbon-carbon double bond. Similarly, the term "alkynyl" refers to a univalent group derived from the removal of a hydrogen atom from any carbon atom of an unsaturated aliphatic hydrocarbon with at least one carbon-carbon triple bond. Furthermore, the term "enynyl" refers to a univalent group derived from the removal of a hydrogen atom from any carbon atom of an unsaturated aliphatic hydrocarbon with at least one carbon-carbon double bond and at least one carbon-carbon triple bond. The unsaturated aliphatic hydrocarbon of an alkenyl, alkynyl or enynyl may be branched or unbranched.

[0025] The term "alkylene" refers to a saturated divalent hydrocarbon group derived from a straight or branched chain saturated hydrocarbon by the removal of two hydrogen atoms, and may be optionally substituted with one or more substituents described herein.

[0026] The term "alkoxy" refers to an alkyl group, as previously defined, attached to the principal carbon chain through an oxygen atom. An alkoxy group may be substituted or unsubstituted.

[0027] The term "aryl" or "aryl group" refers to an organic radical derived from a monocyclic or polycyclic aromatic hydrocarbon by removing a hydrogen atom. An aryl group can be unsubstituted or substituted with one or more suitable substituents.

[0028] The term "alkylamino" refers to a group derived from the removal of a hydrogen atom from a primary or secondary amine. Alkylamino embraces the terms "N-alkylamino" and "N,N-dialkylamino", wherein the amino group is independently substituted with one or two alkyl groups, respectively. The alkylamino group is optionally substituted with one or more substituents.

[0029] The term "alkylthio" refers to a group containing a branched or unbranched alkyl group attached to a divalent sulfur atom. The alkylthio group is optionally substituted with one or more substituents.

[0030] The term "heteroatom" refers to one or more of oxygen (O), sulfur (S), nitrogen (N), phosphorus (P) or silicon (Si), including any oxidized form of nitrogen (N), sulfur (S) or phosphorus (P); the quaternized form of any basic nitrogen; or a substitutable nitrogen of a heterocyclic ring.

[0031] The term "aliphatic" refers to a non-aromatic hydrocarbon or groups derived therefrom.

[0032] The term "aromatic" refers to groups comprising aromatic hydrocarbon rings, optionally including heteroatoms or substituents.

[0033] The term "substituted" is used to describe a compound or chemical moiety wherein at least one hydrogen atom of that compound or chemical moiety is replaced with a second chemical moiety. Examples of substituents include, but are not limited to, halogen; alkyl; heteroalkyl; alkenyl; alkynyl; enynyl; aryl, heteroaryl, hydroxyl; alkoxyl; amino; nitro; thiol; alkylthio; imine; cyano; amido; phosphonato; phosphinato; carboxyl; thiocarbonyl; sulfonyl; sulfonamide; acyl; formyl; acyloxy; alkoxycarbonyl; oxo; haloalkyl; carbocyclic cycloalkyl; carbocyclic or heterocyclic aryl; amino, monoalkylamino and dialkylamino; o-lower alkyl; o-aryl, aryl; aryl-lower alkyl; $-CO_2CH_3$; $-CONH_2$; $-OCH_2CONH_2$; $-NH_2$; $-SO_2NH_2$; $-OCHF_2$; $-CF_3$; $-OCF_3$; –NH(alkyl); –N(alkyl)$_2$; –NH(aryl); –N(alkyl)(aryl); –N(aryl)$_2$; –CHO; –CO(alkyl); -CO(aryl); $-CO_2$(alkyl); and $-CO_2$(aryl); and such moieties can also be optionally substituted by a fused-ring structure or bridge. These substituents can optionally be further substituted with one or more substituents. All chemical groups disclosed herein can be substituted, unless it is specified otherwise.

[0034] The term "halogen" or "halo" refers to F, Cl, Br or I.

[0035] The term "polymer" refers to a polymeric compound prepared by polymerizing monomers, whether of the same or a different type. The generic term "polymer" embraces the

terms "homopolymer" as well as "copolymer".

[0036] The term "homopolymer" refers to a polymer prepared by the polymerization of the same type of monomer.

[0037] The term "copolymer" refers to a polymer prepared by the polymerization of two or more different types of monomers.

[0038] The term "monomeric unit" refers to the constitutional unit derived from a single monomer to the structure of a polymer.

[0039] The term "structural unit" refers to the total monomeric units derived from the same monomer type in a polymer.

[0040] The term "weight-average molecular weight" M_w of a polymer is defined mathematically as:

$$M_w = \frac{\sum N_i M_i^2}{\sum N_i M_i}$$

where N_i is the number of polymer molecules with a particular molecular weight M_i.

[0041] The term "hydrophilic" refers to the property of a substance or material that exhibits an affinity or attraction towards water. This property is often associated with substances that have polar or ionic characteristics, allowing them to form interactions with water molecules. Hydrophilic substances or materials possess a propensity to interact or mix with water, allowing them to be easily wetted or dissolved in water. In some embodiments, the hydrophilic substances or materials comprise functional groups that exhibit a tendency to interact with water molecules to form hydrogen bonds. These functional groups themselves exhibit polarity and are inherently hydrophilic, and can be termed "hydrophilic functional groups". Some non-limiting examples of the hydrophilic functional groups include acid, hydroxyl, and amide.

[0042] The term "hydroneutral" refers to the property of a substance or material that does not dissolve or only partially dissolve in polar solvents, particularly water. Hydroneutral functional groups are typically polar themselves and exhibit a tendency to interact with water molecules, but unlike hydrophilic functional groups, they cannot form hydrogen bonds with water molecules (but can accept hydrogen bonds from said water molecules) due to the lack of hydrogen. Some non-limiting examples of the hydroneutral functional groups include nitrile and nitro.

[0043] The term "homogenizer" refers to a piece of equipment that can be used to uniformly distribute a mixture of materials such that the composition of the mixture becomes constant. In some embodiments, homogenization takes place within a fluid to uniformly disperse materials throughout the fluid, for example during dispersing electrode components in a solvent

to form an electrode slurry. In certain embodiments, homogenization occurs through the blending of dry materials. In other embodiments, homogenization can also occur through the blending of a combination of dry materials and liquid materials and in varying ratios. Any conventional homogenizer can be used for the homogenization process as disclosed herein. Some non-limiting examples of the homogenizer include stirring mixers, planetary mixers, tumblers, and mills.

[0044] The term "applying" refers to an act of laying or spreading a substance on a surface.

[0045] The term "room temperature" refers to indoor temperatures from about 18 °C to about 30 °C, e.g., 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, or 30 °C. In some embodiments, room temperature refers to a temperature of about 20 °C +/- 1 °C or +/- 2 °C or +/- 3 °C. In other embodiments, room temperature refers to a temperature of about 22 °C or about 25 °C.

[0046] In reference to a mixture containing volatile and non-volatile material, the term "solid content" refers to the amount of non-volatile material remaining after evaporation, while the term "solid portion" of a mixture or composition refers to this non-volatile material. Similarly, the term "liquid content" refers to the amount of material that has undergone evaporation, while the term "liquid portion" of a mixture or composition refers to this evaporated material. The sum of solid content and liquid content of the mixture adds up to the total mass of said mixture, and is often expressed as a proportion or percentage relative to the total mass of said mixture. In instances where a mixture is free of liquid, the solid content of a mixture is 100%, while its liquid content is then 0%.

[0047] The term "peeling strength" refers to the amount of force required to separate two materials that are adhered to each other, such as a current collector and an electrode layer. It is a measure of the binding strength between such two materials and is usually expressed in N/cm.

[0048] The term "adhesive strength" refers to the amount of force required to separate a substrate and a binder material adhered to the substrate. It is a measure of the adhesion between such two materials and is usually expressed in N/cm.

[0049] The term "C rate" refers to the charging or discharging rate of a cell or battery, expressed in terms of its total storage capacity in Ah or mAh. For example, a rate of 1 C means utilization of all of the stored energy in one hour; a 0.1 C means utilization of 10% of the energy in one hour or full energy in 10 hours; and a 5 C means utilization of full energy in 12 minutes.

[0050] The term "ampere-hour (Ah)" refers to a unit used in specifying the storage capacity of a battery. For example, a battery with 1 Ah capacity can supply a current of one ampere for one hour or 0.5 A for two hours, etc. Therefore, 1 ampere-hour (Ah) is the equivalent of 3,600 coulombs of electrical charge. Similarly, the term "milliampere-hour (mAh)" also refers

to a unit of the storage capacity of a battery and is 1/1,000 of an ampere-hour.

[0051] The term "capacity" is a characteristic of an electrochemical cell that refers to the total amount of electrical charge an electrochemical cell, such as a battery, is able to hold. Capacity is typically expressed in units of ampere-hours. The term "specific capacity" refers to the capacity output of an electrochemical cell, such as a battery, per unit weight, usually expressed in Ah/kg or mAh/g.

[0052] In the following description, all numbers disclosed herein are approximate values, regardless whether the word "about" or "approximate" is used in connection therewith. They may vary by 1 percent, 2 percent, 5 percent, or, sometimes, 10 to 20 percent. Whenever a numerical range with a lower limit, R^L, and an upper limit, R^U, is disclosed, any number falling within the range is specifically disclosed. In particular, the following numbers within the range are specifically disclosed: $R=R^L+k*(R^U-R^L)$, wherein k is a variable ranging from 0 percent to 100 percent. Moreover, any numerical range defined by two R numbers as defined in the above is also specifically disclosed.

[0053] In the present description, all references to the singular include references to the plural and vice versa. In the present description, where the context allows, references to an "aqueous solvent" may also specifically refer to water.

[0054] Currently, electrodes are often prepared by dispersing an electrode active material, a binder material and a conductive agent in a solvent to form an electrode slurry, then coating the electrode slurry onto a current collector and drying it. A widely-used electrode slurry formulation comprises PVDF as the binder material and a solvent of NMP, but the use of NMP presents significant environmental, health and safety risks, and necessitating a vapor recovery system which would incur additional costs. When applied to a dry electrode material mixture, these conventional fluorinated binders such as PVDF and PTFE are able to fibrillize, connecting the electrode components during the formation of the electrode layer. Nonetheless, the use of fluorinated binders necessitates the use of organic solvents in the recycling process. Attempts have also been made to utilize water-compatible polymers as binder materials, but so far, these binder polymers have to be used in a slurry with a relatively high aqueous solvent content, of which the aqueous solvent would react with and cause degradation of the cathode active material and leading to worsened battery performance.

[0055] Disclosed herein is a binder composition comprising a copolymer and a solvent, wherein the copolymer comprises a hydrophilic structural unit and a non-hydrophilic structural unit, within a particular weight-average molecular weight range. It has been observed that when used in an electrode material mixture and solvent is present in a certain proportion, for example through the incorporation of said binder composition in the electrode material mixture, the copolymer has an inherent capability to readily fibrillize, exhibiting exceptional binding

properties and hence allowing for thicker electrode layers.

[0056] There are no particular limitations to the method of producing the copolymer, but in some embodiments, the copolymer disclosed herein is produced via polymerization of monomers, polymers or monomer-polymer complexes dispersed in an aqueous solvent, the polymerization being initiated by free radicals generated by a water-soluble free radical initiator. Any suitable reaction conditions can be used in the polymerization process, as long as they result in the successful formation of the copolymer.

[0057] Free radical initiators and other chemicals required in the polymerization process are dispersed in the aqueous solvent. In some embodiments, the aqueous solvent is water. In some embodiments, the aqueous solvent is selected from the group consisting of tap water, bottled water, purified water, pure water, distilled water, deionized water (DI water), D_2O, and combinations thereof.

[0058] However, it is important to note that the aqueous solvent should preferably not contain any minor components, as the fibrillization performance of the copolymer relies on interactions between the copolymer and water. It has been discovered that the presence of a minor component in the aqueous solvent in the binder composition adversely affects the fibrillization of the copolymer. Therefore, in some embodiments, no minor component, apart from water, is present in the aqueous solvent in the binder composition.

[0059] In some embodiments, the minor component includes alcohols, lower aliphatic ketones, lower alkyl acetates, and combinations thereof. Some non-limiting examples of the alcohol include C_1-C_4 alcohols, such as methanol, ethanol, isopropanol, n-propanol, tert-butanol, n-butanol, 1,2-butanediol, 1,3-butanediol, 1,4-butanediol, 2,3-butanediol, ethylene glycol, propylene glycol, glycerol, and combinations thereof. Some non-limiting examples of the lower aliphatic ketones include acetone, dimethyl ketone, methyl ethyl ketone (MEK), and combinations thereof. Some non-limiting examples of the lower alkyl acetates include ethyl acetate (EA), isopropyl acetate, propyl acetate, butyl acetate (BA), and combinations thereof. Some other non-limiting examples of other minor components include 1,4-dioxane, diethyl ether, methyl tert-butyl ether, cyclopentyl methyl ether, tetrahydrofuran (THF), 2-methyl tetrahydrofuran, acetonitrile, dimethyl sulfoxide (DMSO), sulfolane, nitromethane, propylene carbonate, ethylene carbonate, dimethyl carbonate, pyridine, acetaldehyde, formic acid, acetic acid, propanoic acid, butyric acid, γ-valerolactone (GVL), furfuryl alcohol, methyl lactate, ethyl lactate, diethanolamine, dimethylacetamide (DMAc), dimethylformamide (DMF), N-methylpyrrolidone (NMP), dihydrolevoglucosenone (Cyrene™), N,N'-dimethylpropyleneurea (DMPU), and dimethyl isosorbide (DMI).

[0060] Surprisingly, it has been found that in order for the copolymer to exhibit desirable fibrillization characteristics, and consequently excellent binder performance, such as adhesive

strength, the presence of both a hydrophilic structural unit and a non-hydrophilic structural unit is essential. This adhesion is attributed to the combined effect of hydrophilic and non-hydrophilic interactions contributed by the respective structural units with water molecules. Accordingly, it is preferable for the copolymer in the binder composition of the present invention to comprise a hydrophilic structural unit and a non-hydrophilic structural unit. Some non-limiting examples of monomers which the hydrophilic structural unit can be derived from include an acid group-containing monomer, an amide group-containing monomer, and a combination thereof; and wherein the acid group is selected from the group consisting of carboxylic acid, sulfonic acid, sulfuric acid, phosphonic acid, phosphoric acid, nitric acid, their salts, their derivatives, and combinations thereof. Some non-limiting examples of monomers which the non-hydrophilic structural unit can be derived from include a nitrile group-containing monomer.

[0061] The proportion of each structural unit within the copolymer plays a critical role. When the proportion of the hydrophilic structural unit in the copolymer is too high, battery performance would be worsened as the copolymer would be overly hydrophilic. The copolymer would then exhibit an excessively high affinity for water, disrupting the intended fibrillization process and resulting in suboptimal performance when used to manufacture electrodes using a dry electrode material mixture. The presence of the non-hydrophilic structural unit in the copolymer thus plays a vital role to maintaining the desired fibrillization behavior of the copolymer during the preparation of electrodes using a dry electrode material mixture as excessive hydrogen bonding with water is prevented. On the other hand, when the proportion of the non-hydrophilic structural unit in the copolymer is too high, the copolymer loses its water compatibility. As a consequence, the copolymer would demonstrate inadequate fibrillization, rendering it incapable of acting as a binder material. Therefore, finding the right balance between these structural units is essential to ensure both water compatibility and optimal battery performance.

[0062] In some embodiments, the proportion of hydrophilic structural unit within the copolymer is from about 60% to about 90%, from about 63% to about 90%, from about 66% to about 90%, from about 69% to about 90%, from about 72% to about 90%, from about 75% to about 90%, from about 78% to about 90%, from about 81% to about 90%, from about 84% to about 90%, from about 87% to about 90%, from about 60% to about 87%, from about 63% to about 87%, from about 66% to about 87%, from about 69% to about 87%, from about 72% to about 87%, from about 75% to about 87%, from about 78% to about 87%, from about 81% to about 87%, from about 84% to about 87%, from about 60% to about 84%, from about 63% to about 84%, from about 66% to about 84%, from about 69% to about 84%, from about 72% to about 84%, from about 75% to about 84%, from about 78% to about 84%, from about 81% to about 84%, from about 60% to about 81%, from about 63% to about 81%, from about 66% to

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[0063] In some embodiments, the proportion of hydrophilic structural unit within the copolymer is about 60%, about 63%, about 66%, about 69%, about 72%, about 75%, about 78%, about 81%, about 84%, about 87%, or about 90% by mole, based on the total number of moles of monomeric units in the copolymer. In some embodiments, the proportion of hydrophilic structural unit within the copolymer is less than 90%, less than 87%, less than 84%, less than 81%, less than 78%, less than 75%, less than 72%, less than 69%, less than 66%, or less than 63% by mole, based on the total number of moles of monomeric units in the copolymer. In some embodiments, the proportion of hydrophilic structural unit within the copolymer is more than 60%, more than 63%, more than 66%, more than 69%, more than 72%, more than 75%, more than 78%, more than 81%, more than 84%, or more than 87% by mole, based on the total number of moles of monomeric units in the copolymer.

[0064] In a preferred embodiment, the hydrophilic structural unit of the copolymer comprises monomeric units derived from a monomer comprising a hydrophilic functional group. In certain embodiments, the monomer comprising a hydrophilic functional group is an acid group-containing monomer and an amide group-containing monomer. The acid group (e.g. a carboxylic acid group) is polar in nature because of the differences in electronegativities of the carbon (C) atom and oxygen (O) atom in the carbonyl subgroup, and between the oxygen (O) atom and hydrogen (H) atom in the hydroxyl subgroup. The unequal charge distribution within the acid group results in a net dipole moment, making the acid group highly polar in nature. This high polarity renders the acid group highly hydrophilic and enables it to participate in hydrogen bond formation (through the hydroxyl subgroup) and dipole-dipole interactions, making it a crucial functional group within the copolymer.

[0065] Similarly, an amide group is polar in nature because of the differences in electronegativities of the carbon (C) atom and oxygen (O) atom in the carbonyl subgroup, and between the nitrogen (N) atom and hydrogen (H) atom in the amine subgroup. The resulting unequal charge distributions lead to a net dipole moment, making the amide group also highly polar in nature. This high polarity makes the amide group highly hydrophilic. Like the acid group, the amide group can participate in hydrogen bond formation (through the amine subgroup) and dipole-dipole interactions. It is therefore another crucial functional group present

in the copolymer.

[0066] In some embodiments, the acid group is selected from the group consisting of carboxylic acid, sulfonic acid, sulfuric acid, phosphonic acid, phosphoric acid, nitric acid, salts of these acids, derivatives of these acids, and combinations thereof. In some embodiments, the salt of the acid comprises an alkali metal cation. Examples of the alkali metal include lithium, sodium, and potassium. In some embodiments, the salt of the acid comprises an ammonium cation. In some embodiments, the acid group is specifically a combination of one or more of the above acids and one or more salts of the above acids.

[0067] In some embodiments, the carboxylic acid is acrylic acid, methacrylic acid, crotonic acid, 2-butyl crotonic acid, cinnamic acid, maleic acid, maleic anhydride, fumaric acid, itaconic acid, itaconic anhydride, tetraconic acid, or combinations thereof. In certain embodiments, the carboxylic acid is 2-ethylacrylic acid, isocrotonic acid, cis-2-pentenoic acid, trans-2-pentenoic acid, angelic acid, tiglic acid, 3,3-dimethyl acrylic acid, 3-propyl acrylic acid, trans-2-methyl-3-ethyl acrylic acid, cis-2-methyl-3-ethyl acrylic acid, 3-isopropyl acrylic acid, trans-3-methyl-3-ethyl acrylic acid, cis-3-methyl-3-ethyl acrylic acid, 2-isopropyl acrylic acid, trimethyl acrylic acid, 2-methyl-3,3-diethyl acrylic acid, 3-butyl acrylic acid, 2-butyl acrylic acid, 2-pentyl acrylic acid, 2-methyl-2-hexenoic acid, trans-3-methyl-2-hexenoic acid, 3-methyl-3-propyl acrylic acid, 2-ethyl-3-propyl acrylic acid, 2,3-diethyl acrylic acid, 3,3-diethyl acrylic acid, 3-methyl-3-hexyl acrylic acid, 3-methyl-3-tert-butyl acrylic acid, 2-methyl-3-pentyl acrylic acid, 3-methyl-3-pentyl acrylic acid, 4-methyl-2-hexenoic acid, 4-ethyl-2-hexenoic acid, 3-methyl-2-ethyl-2-hexenoic acid, 3-tert-butyl acrylic acid, 2,3-dimethyl-3-ethyl acrylic acid, 3,3-dimethyl-2-ethyl acrylic acid, 3-methyl-3-isopropyl acrylic acid, 2-methyl-3-isopropyl acrylic acid, trans-2-octenoic acid, cis-2-octenoic acid, trans-2-decenoic acid, α-acetoxyacrylic acid, β-trans-aryloxyacrylic acid, α-chloro-β-E-methoxyacrylic acid, or combinations thereof. In some embodiments, the carboxylic acid is methyl maleic acid, dimethyl maleic acid, phenyl maleic acid, bromo maleic acid, chloromaleic acid, dichloromaleic acid, fluoromaleic acid, difluoro maleic acid, nonyl hydrogen maleate, decyl hydrogen maleate, dodecyl hydrogen maleate, octadecyl hydrogen maleate, fluoroalkyl hydrogen maleate, or combinations thereof. In some embodiments, the carboxylic acid is maleic anhydride, methyl maleic anhydride, dimethyl maleic anhydride, acrylic anhydride, methacrylic anhydride, or combinations thereof.

[0068] In some embodiments, the sulfonic acid is vinylsulfonic acid, methylvinylsulfonic acid, allylvinylsulfonic acid, allylsulfonic acid, methallylsulfonic acid, styrenesulfonic acid, 2-sulfoethyl methacrylic acid, 2-methylprop-2-ene-1-sulfonic acid, 2-acrylamido-2-methyl-1-propane sulfonic acid, 3-allyloxy-2-hydroxy-1-propane sulfonic acid, or combinations thereof.

[0069] In some embodiments, the sulfuric acid is allyl hydrogen sulfate, vinyl hydrogensulfate, 4-allyl phenol sulphate, or combinations thereof.

[0070] In some embodiments, the phosphonic acid is vinyl phosphonic acid, allyl phosphonic acid, vinyl benzyl phosphonic acid, acrylamide alkyl phosphonic acid, methacrylamide alkyl phosphonic acid, acrylamide alkyl diphosphonic acid, acryloylphosphonic acid, 2-methacryloyloxyethyl phosphonic acid, bis(2-methacryloyloxyethyl) phosphonic acid, ethylene 2-methacryloyloxyethyl phosphonic acid, ethyl-methacryloyloxyethyl phosphonic acid, or combinations thereof.

[0071] In some embodiments, the phosphoric acid is mono (2-acryloyloxyethyl) phosphate, mono (2-methacryloyloxyethyl) phosphate, diphenyl (2-acryloyloxyethyl) phosphate, diphenyl (2-methacryloyloxyethyl) phosphate, phenyl (2-acryloyloxyethyl) phosphate, phosphoxyethyl methacrylate, 3-chloro-2-phosphoryloxy propyl methacrylate, phosphoryloxy poly(ethylene glycol) monomethacrylate, phosphoryloxy poly(propylene glycol) methacrylate, (meth)acryloyloxyethyl phosphate, (meth)acryloyloxypropyl phosphate, (meth)acryloyloxy-2-hydroxypropyl phosphate, (meth)acryloyloxy-3-hydroxypropyl phosphate, (meth)acryloyloxy-3-chloro-2 hydroxypropyl phosphate, allyl hydrogen phosphate, vinyl hydrogen phosphate, allyl hydrogen pyrophosphate, vinyl hydrogen pyrophosphate, allyl hydrogen tripolyphosphate, vinyl hydrogen tripolyphosphate, allyl hydrogen tetrapolyphosphate, vinyl hydrogen tetrapolyphosphate, allyl hydrogen trimetaphosphate, vinyl hydrogen trimetaphosphate, isopentenyl phosphate, isopentenyl pyrophosphate, or combinations thereof, wherein (meth)acryloyl- represents acryloyl- or methacryloyl-.

[0072] In some embodiments, the nitric acid is allyl hydrogen nitrate, ethenyl hydrogen nitrate, or combinations thereof.

[0073] In some embodiments, the proportion of monomeric units in the copolymer derived from an acid group-containing monomer is from about 10% to about 40%. When the proportion of monomeric units derived from an acid group-containing monomer in the copolymer exceeds the above range, the copolymer would lack the necessary structural organization and intermolecular interactions required for proper fibrillization, which would lead to a significant decrease in the copolymer's adhesive strength and lowered overall effectiveness as binder material in electrode manufacture using a dry electrode material mixture. Conversely, when the proportion of monomeric units derived from an acid group-containing monomer in the copolymer is below the above range, the copolymer would be insufficiently water-compatible for proper fibrillization as well. Therefore, it is crucial to carefully control and maintain the proportion of monomeric units derived from an acid group-containing monomer in the copolymer to within the specified range.

[0074] In certain embodiments, the proportion of monomeric units in the copolymer derived from an acid group-containing monomer is from about 10% to about 40%, from about 12% to about 40%, from about 15% to about 40%, from about 18% to about 40%, from about

20% to about 40%, from about 22% to about 40%, from about 25% to about 40%, from about 28% to about 40%, from about 30% to about 40%, from about 32% to about 40%, from about 35% to about 40%, from about 10% to about 35%, from about 12% to about 35%, from about 15% to about 35%, from about 18% to about 35%, from about 20% to about 35%, from about 22% to about 35%, from about 25% to about 35%, from about 28% to about 35%, from about 30% to about 35%, from about 10% to about 30%, from about 12% to about 30%, from about 15% to about 30%, from about 18% to about 30%, from about 20% to about 30%, from about 22% to about 30%, from about 25% to about 30%, from about 10% to about 25%, from about 12% to about 25%, from about 15% to about 25%, from about 18% to about 25%, from about 20% to about 25%, from about 10% to about 20%, from about 12% to about 20%, from about 15% to about 20%, from about 10% to about 15%, or from about 12% to about 15% by mole, based on the total number of moles of monomeric units in the copolymer.

[0075] In some embodiments, the proportion of monomeric units in the copolymer derived from an acid group-containing monomer is about 10%, about 12%, about 15%, about 18%, about 20%, about 22%, about 25%, about 28%, about 30%, about 32%, about 35%, about 38%, or about 40% by mole, based on the total number of moles of monomeric units in the copolymer. In some embodiments, the proportion of monomeric units in the copolymer derived from an acid group-containing monomer is less than 40%, less than 38%, less than 35%, less than 32%, less than 30%, less than 28%, less than 25%, less than 22%, less than 20%, less than 18%, less than 15%, or less than 12% by mole, based on the total number of moles of monomeric units in the copolymer. In some embodiments, the proportion of monomeric units in the copolymer derived from an acid group-containing monomer is more than 10%, more than 12%, more than 15%, more than 18%, more than 20%, more than 22%, more than 25%, more than 28%, more than 30%, more than 32%, more than 35%, or more than 38% by mole, based on the total number of moles of monomeric units in the copolymer.

[0076] In some embodiments, the amide group-containing monomer is acrylamide, methacrylamide, N-methyl methacrylamide, N-ethyl methacrylamide, N-n-propyl methacrylamide, N-isopropyl methacrylamide, isopropyl acrylamide, N-n-butyl methacrylamide, N-isobutyl methacrylamide, N,N-dimethyl acrylamide, N,N-dimethyl methacrylamide, N,N-diethyl acrylamide, N,N-diethyl methacrylamide, N-methylol methacrylamide, N-(methoxymethyl)methacrylamide, N-(ethoxymethyl)methacrylamide, N-(propoxymethyl)methacrylamide, N-(butoxymethyl)methacrylamide, N,N-dimethylaminopropyl methacrylamide, N,N-dimethylaminoethyl methacrylamide, N,N-dimethylol methacrylamide, diacetone methacrylamide, diacetone acrylamide, methacryloyl morpholine, N-hydroxyl methacrylamide, N-methoxymethyl acrylamide, N-methoxymethyl methacrylamide, N,N'-methylene-bis-acrylamide (MBA), N-hydroxymethyl acrylamide, or combinations thereof.

[0077] In some embodiments, the proportion of monomeric units in the copolymer

derived from an amide group-containing monomer is from about 10% to about 55%. When the proportion of monomeric units derived from an amide group-containing monomer in the copolymer exceeds the above range, the copolymer would lack the necessary structural organization and intermolecular interactions required for proper fibrillization, which would lead to a significant decrease in the copolymer's adhesive strength and lowered overall effectiveness as binder material in electrode manufacture using a dry electrode material mixture. Conversely, when the proportion of monomeric units derived from an amide group-containing monomer in the copolymer is below the above range, the copolymer would be insufficiently water-compatible for proper fibrillization as well. Therefore, it is crucial to carefully control and maintain the proportion of monomeric units derived from an amide group-containing monomer in the copolymer to within the specified range.

[0078] In certain embodiments, the proportion of monomeric units in the copolymer derived from an amide group-containing monomer is from about 10% to about 55%, from about 15% to about 55%, from about 20% to about 55%, from about 25% to about 55%, from about 30% to about 55%, from about 35% to about 55%, from about 40% to about 55%, from about 45% to about 55%, from about 50% to about 55%, from about 10% to about 50%, from about 15% to about 50%, from about 20% to about 50%, from about 25% to about 50%, from about 30% to about 50%, from about 35% to about 50%, from about 40% to about 50%, from about 45% to about 50%, from about 10% to about 45%, from about 15% to about 45%, from about 20% to about 45%, from about 25% to about 45%, from about 30% to about 45%, from about 35% to about 45%, from about 40% to about 45%, from about 10% to about 40%, from about 15% to about 40%, from about 20% to about 40%, from about 25% to about 40%, from about 30% to about 40%, from about 35% to about 40%, from about 10% to about 35%, from about 15% to about 35%, from about 20% to about 35%, from about 25% to about 35%, from about 30% to about 35%, from about 10% to about 30%, from about 15% to about 30%, from about 20% to about 30%, from about 25% to about 30%, from about 10% to about 25%, from about 15% to about 25%, from about 20% to about 25%, from about 10% to about 20%, from about 15% to about 20%, or from about 10% to about 15% by mole, based on the total number of moles of monomeric units in the copolymer.

[0079] In some embodiments, the proportion of monomeric units in the copolymer derived from an amide group-containing monomer is about 10%, about 15%, about 20%, about 25%, about 30%, about 35%, about 40%, about 45%, about 50%, or about 55% by mole, based on the total number of moles of monomeric units in the copolymer. In some embodiments, the proportion of monomeric units in the copolymer derived from an amide group-containing monomer is less than 55%, less than 50%, less than 45%, less than 40%, less than 35%, less than 30%, less than 25%, less than 20%, or less than 15% by mole, based on the total number of moles of monomeric units in the copolymer. In some embodiments, the proportion of

monomeric units in the copolymer derived from an amide group-containing monomer is more than 10%, more than 15%, more than 20%, more than 25%, more than 30%, more than 35%, more than 40%, more than 45%, or more than 50% by mole, based on the total number of moles of monomeric units in the copolymer.

[0080] A hydroxyl group is a hydrophilic functional group. Nevertheless, it has been discovered that the fibrillization of the copolymer deteriorates when it contains monomeric units derived from a hydroxyl group-containing monomer. As a result, in preferred embodiments, the copolymer is intentionally formulated without any monomeric units derived from a hydroxyl group-containing monomer.

[0081] In some embodiments, the non-hydrophilic structural unit of the copolymer comprises monomeric units derived from a nitrile group-containing monomer. The nitrile group is considered a hydroneutral functional group. It is not inherently hydrophilic but still retains sufficient polarity to avoid excessively compromising the water compatibility of the entire copolymer. The nitrile group exhibits polarity as the shared electrons in the covalent CN bond are more strongly attracted towards the N atom than the C atom. This allows the nitrile group to form strong intermolecular forces with acid and amide groups contained in another copolymer chain. Furthermore, the nitrogen (N) atom of a nitrile group (−C≡N) can accept hydrogen bonds with water molecules. However, without hydrogen in the nitrile group, a nitrile cannot itself form hydrogen bonds. Accordingly, the nitrile group is not considered hydrophilic, but is instead hydroneutral.

[0082] In some embodiments, the nitrile group-containing monomer includes α,β-ethylenically unsaturated nitrile monomers. In some embodiments, the nitrile group-containing monomer is acrylonitrile, α-halogenoacrylonitrile, α-alkylacrylonitrile, or combinations thereof. In some embodiments, the nitrile group-containing monomer is α-chloroacrylonitrile, α-bromoacrylonitrile, α-fluoroacrylonitrile, methacrylonitrile, α-ethylacrylonitrile, α-isopropylacrylonitrile, α-n-hexylacrylonitrile, α-methoxyacrylonitrile, 3-methoxyacrylonitrile, 3-ethoxyacrylonitrile, α-acetoxyacrylonitrile, α-phenylacrylonitrile, α-tolylacrylonitrile, α-(methoxyphenyl)acrylonitrile, α-(chlorophenyl)acrylonitrile, α-(cyanophenyl)acrylonitrile, vinylidene cyanide, or combinations thereof.

[0083] In some embodiments, the proportion of monomeric units in the copolymer derived from a nitrile group-containing monomer is from about 10% to about 40%. The presence of these nitrile group-derived monomeric units contributes to the overall polarity of the copolymer while avoiding excessive water compatibility, allowing the copolymer to interact appropriately with water without impeding the fibrillization process. This ensures that the copolymer retains its functionality as binder material when preparing electrodes using a dry electrode material mixture.

[0084] In certain embodiments, the proportion of monomeric units in the copolymer derived from a nitrile group-containing monomer from about 10% to about 40%, from about 12% to about 40%, from about 15% to about 40%, from about 18% to about 40%, from about 20% to about 40%, from about 22% to about 40%, from about 25% to about 40%, from about 28% to about 40%, from about 30% to about 40%, from about 32% to about 40%, from about 35% to about 40%, from about 10% to about 35%, from about 12% to about 35%, from about 15% to about 35%, from about 18% to about 35%, from about 20% to about 35%, from about 22% to about 35%, from about 25% to about 35%, from about 28% to about 35%, from about 30% to about 35%, from about 10% to about 30%, from about 12% to about 30%, from about 15% to about 30%, from about 18% to about 30%, from about 20% to about 30%, from about 22% to about 30%, from about 25% to about 30%, from about 10% to about 25%, from about 12% to about 25%, from about 15% to about 25%, from about 18% to about 25%, from about 20% to about 25%, from about 10% to about 20%, from about 12% to about 20%, from about 15% to about 20%, from about 10% to about 15%, or from about 12% to about 15% by mole, based on the total number of moles of monomeric units in the copolymer.

[0085] In some embodiments, the proportion of monomeric units in the copolymer derived from a nitrile group-containing monomer is about 10%, about 12%, about 15%, about 18%, about 20%, about 22%, about 25%, about 28%, about 30%, about 32%, about 35%, about 38%, or about 40% by mole, based on the total number of moles of monomeric units in the copolymer. In certain embodiments, the proportion of monomeric units in the copolymer derived from a nitrile group-containing monomer is less than 40%, less than 38%, less than 35%, less than 32%, less than 30%, less than 28%, less than 25%, less than 22%, less than 20%, less than 18%, less than 15%, or less than 12% by mole, based on the total number of moles of monomeric units in the copolymer. In some embodiments, the proportion of monomeric units in the copolymer derived from a nitrile group-containing monomer is more than 10%, more than 12%, more than 15%, more than 18%, more than 20%, more than 22%, more than 25%, more than 28%, more than 30%, more than 32%, more than 35%, or more than 38% by mole, based on the total number of moles of monomeric units in the copolymer.

[0086] Subsequent investigations revealed that the fibrillization of the copolymer was negatively affected when it consisted of monomeric units derived from certain specific types of monomers, even if these monomeric units could have otherwise been considered part of the non-hydrophilic structural unit. These included ester group-containing monomers, epoxy group-containing monomers, ether group-containing monomers, carbonyl group-containing monomers, and fluorine-containing monomers. In such cases, the presence of these monomers within the copolymer hindered the desired fibrillization process. Therefore, in preferred embodiments, it is advantageous for the copolymer to exclude any monomeric units derived from these aforementioned monomers. By avoiding the inclusion of these specific monomers, the

copolymer can circumvent the complications associated with their adverse impact on fibrillization. This ensures that the copolymer maintains its intended fibrillization behavior, thereby optimizing its performance and suitability in manufacturing electrodes using the dry electrode material mixture disclosed herein. In some embodiments, the non-hydrophilic structural unit does not comprise an ester group-containing monomer, an epoxy group-containing monomer, an ether group-containing monomer, a carbonyl group-containing monomer, a fluorine-containing monomer, or combinations thereof.

[0087] In further embodiments, the non-hydrophilic structural unit of the copolymer comprises monomeric units derived from an olefin. Any hydrocarbon that has at least one carbon-carbon double bond may be used as the olefin. In some embodiments, the olefin includes a C_2-C_{20} aliphatic compound, a C_8-C_{20} aromatic or cyclic compound containing vinylic unsaturation, a C_4-C_{40} diene, and combinations thereof. In some embodiments, the olefin is styrene, ethylene, propylene, isobutylene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene, 1-eicosene, 3-methyl-1-butene, cyclobutene, 3-methyl-1-pentene, 4-methyl-1-pentene, 4,6-dimethyl-1-heptene, 4-vinylcyclohexene, vinyl cyclohexane, norbornene, norbornadiene, ethylidene norbornene, cyclopentene, cyclohexene, dicyclopentadiene, cyclooctene, or combinations thereof. In some embodiments, the water-compatible copolymer does not comprise a structural unit derived from an olefin. A conjugated diene constitutes an olefin. In some embodiments, the conjugated diene is selected from the group consisting of C_4-C_{40} dienes; aliphatic conjugated dienes such as 1,3-butadiene, 1,3-pentadiene, 1,4-hexadiene, 1,5-hexadiene, 1,7-octadiene, 1,9-decadiene, isoprene, myrcene, 2-methyl-1,3-butadiene, 2,3-dimethyl-1,3-butadiene, 2-chloro-1,3-butadiene; substituted linear conjugated pentadienes; substituted branched conjugated hexadienes; and combinations thereof. It was discovered that the fibrillization process was not significantly influenced by the presence of monomeric units derived from an olefin, but the proportion of monomeric units derived from an olefin in the copolymer should be carefully controlled to maintain the desired fibrillization behavior of the copolymer, preferably not exceeding about 15% by mole, based on the total number of moles of monomeric units in the copolymer. By adhering to this limit, the copolymer can effectively undergo fibrillization without any substantial negative effects. This ensures that the copolymer retains its desired structural properties and functional characteristics, making it suitable for various applications where fibrillization is crucial.

[0088] The impact of the inclusion of monomeric units derived from an aromatic vinyl group-containing monomer in the copolymer on the fibrillization of the copolymer was also investigated. These monomeric units would be considered part of the non-hydrophilic structural unit. In some embodiments, the aromatic vinyl group-containing monomer is styrene, α-methylstyrene, vinyltoluene, divinylbenzene, or combinations thereof. However, it was found

that fibrillization of the copolymer was worsened when the copolymer comprised monomeric units derived from an aromatic vinyl group-containing monomer. This negative impact can be attributed to the steric hindrance caused by the bulky aromatic group. The steric hindrance interferes with the interaction between two copolymer chains and restricts the sliding movement between them during the fibrillization process. Therefore, it is advisable to exclude monomeric units derived from any of these aromatic vinyl group-containing monomers.

[0089] The copolymer has a weight-average molecular weight within the ranges set forth below. Surprisingly, it was discovered that this molecular weight range is critical to ensure good fibrilization performance, and when the copolymer has a weight-average molecular weight within the disclosed range, electrode layers comprising this copolymer exhibit strong adhesion and peeling strength. This allows for the electrode layer to be made thicker, increasing battery energy density while not sacrificing cycle characteristics or ability to maintain stable performance over repeated charge-discharge cycles. The copolymer disclosed herein thus not only serves as an effective binder material, but also contributes to the overall performance and durability of batteries.

[0090] In some embodiments, the weight-average molecular weight of the copolymer is from about 400,000 g/mol to about 700,000 g/mol, from about 450,000 g/mol to about 700,000 g/mol, from about 500,000 g/mol to about 700,000 g/mol, from about 550,000 g/mol to about 700,000 g/mol, from about 600,000 g/mol to about 700,000 g/mol, from about 650,000 g/mol to about 700,000 g/mol, from about 400,000 g/mol to about 650,000 g/mol, from about 450,000 g/mol to about 650,000 g/mol, from about 500,000 g/mol to about 650,000 g/mol, from about 550,000 g/mol to about 650,000 g/mol, from about 600,000 g/mol to about 650,000 g/mol, from about 400,000 g/mol to about 600,000 g/mol, from about 450,000 g/mol to about 600,000 g/mol, from about 500,000 g/mol to about 600,000 g/mol, from about 550,000 g/mol to about 600,000 g/mol, from about 400,000 g/mol to about 550,000 g/mol, from about 450,000 g/mol to about 550,000 g/mol, from about 500,000 g/mol to about 550,000 g/mol, from about 400,000 g/mol to about 500,000 g/mol, from about 450,000 g/mol to about 500,000 g/mol, or from about 400,000 g/mol to about 450,000 g/mol.

[0091] In some embodiments, the weight-average molecular weight of the copolymer is less than 700,000 g/mol, less than 650,000 g/mol, less than 600,000 g/mol, less than 550,000 g/mol, less than 500,000 g/mol, or less than 450,000 g/mol. In some embodiments, the weight-average molecular weight of the copolymer is more than 400,000 g/mol, more than 450,000 g/mol, more than 500,000 g/mol, more than 550,000 g/mol, more than 600,000 g/mol, or more than 650,000 g/mol.

[0092] In some embodiments, the current collector is an aluminium foil substrate, and accordingly the adhesive strength between the copolymer and the aluminium foil substrate

would be a good indicator of the copolymer's adhesive performance to a current collector. In some embodiments, the adhesive strength between the copolymer and an aluminium foil substrate is from about 1 N/cm to about 10 N/cm, from about 2 N/cm to about 10 N/cm, from about 3 N/cm to about 10 N/cm, from about 4 N/cm to about 10 N/cm, or from about 5 N/cm to about 10 N/cm. In certain embodiments, the adhesive strength between the copolymer and an aluminium foil substrate is less than 10 N/cm, less than 9 N/cm, less than 8 N/cm, less than 7 N/cm, less than 6 N/cm, less than 5 N/cm, less than 4 N/cm, less than 3 N/cm, or less than 2 N/cm. In some embodiments, the adhesive strength between the copolymer and an aluminium foil substrate is more than 1 N/cm, more than 2 N/cm, more than 3 N/cm, more than 4 N/cm, more than 5 N/cm, more than 6 N/cm, more than 7 N/cm, more than 8 N/cm, or more than 9 N/cm.

[0093] For many binder polymers, once they are completely dried, they undergo irreversible changes that prevent them from being redissolved in aqueous solvent. This can have an impact on binder performance, making complete drying not recommended. However, the copolymer disclosed herein can retain its functionality following rehydration even after undergoing complete drying. This means that it is possible to dry the post-reaction mixture resulting from the copolymerization process until there is substantially no liquid left, and subsequently use or store the dry binder material, instead of having to maintain the copolymer in water at all times. This reduces the volume required for storage. Subsequently, an aqueous solvent, preferably solely water as discussed above, can then be added to a dry mixture of the electrode components comprising the dry binder material during the manufacture of an electrode material mixture. Alternatively, the binder composition may be used directly in the manufacture of the electrode material mixture.

[0094] There is no particular limitation on the dryer used and the conditions used for drying, except the dryer should be able to reduce the liquid content of the post-reaction mixture at the relevant set of conditions without degrading the copolymer within the mixture. In some embodiments, the dryer is a spray dryer, freeze dryer, pan dryer, rotary dryer, screw dryer, fluidized bed dryer, drum dryer, vacuum dryer, or combinations thereof.

[0095] Regardless of the method for preparing the electrode material mixture, the presence of water is essential for fibrillization of the copolymer. Without water, the copolymer exists in the form of linear chains that strongly interact with one another through various intermolecular forces, such as dipole-dipole, ion-dipole, dipole-induced dipole, and induced dipole-induced dipole interactions, and hydrogen bonding. A significant amount of these interactions are by the functional groups located on each copolymer chain, such as alignment of interacting pairs between a nitrile (C≡N) group in one copolymer chain and a carboxylic acid (COOH) group in another copolymer chain, which may involve hydrogen bonds and dipole-

dipole interactions. Collectively, these strong intermolecular forces lock the copolymer chains in position, preventing fibrillization.

[0096] Water molecules disrupts the electrostatic attractions between copolymer chains. interacting with the hydrophilic groups in the copolymer chains and forming new hydrogen bonds with these hydrophilic groups. This interaction reduces the affinity of the functional groups between copolymer chains. For example, hydrogen bonding interactions can occur between a carboxylic acid group in one copolymer chain and a water molecule, instead of interacting with a nitrile group in another copolymer chain. This disruption weakens the intermolecular interactions between the copolymer chains, allowing them to slide over one another under shear force, leading to fibrillization. Accordingly, it is preferable for some water to present in the electrode material mixture comprising the copolymer disclosed herein, whether this water be present in the binder composition comprising the copolymer, or through rehydration of the dry binder material with water during the production of the electrode material mixture.

[0097] As discussed above, the copolymer readily fibrillizes in the presence of water. There are various methods known in the art to measure fibrillization of a polymer, of which the fibrillization method disclosed in the Example section was adopted. In some embodiments, the copolymer can be drawn to a height of 20-40 cm when using this fibrillization method. In certain embodiments, fibrillization of the copolymer as measured using the fibrillization method is from about 20 cm to about 40 cm, from about 22 cm to about 40 cm, from about 24 cm to about 40 cm, from about 26 cm to about 40 cm, from about 28 cm to about 40 cm, from about 30 cm to about 40 cm, from about 32 cm to about 40 cm, from about 34 cm to about 40 cm, from about 20 cm to about 35 cm, from about 22 cm to about 35 cm, from about 24 cm to about 35 cm, from about 26 cm to about 35 cm, from about 28 cm to about 35 cm, or from about 30 cm to about 35 cm.

[0098] In certain embodiments, fibrillization of the copolymer as measured using the fibrillization method is less than 40 cm, less than 39 cm, less than 38 cm, less than 37 cm, less than 36 cm, less than 35 cm, less than 34 cm, less than 33 cm, less than 32 cm, less than 31 cm, less than 30 cm, less than 28 cm, less than 26 cm, less than 24 cm, or less than 22 cm. In certain embodiments, fibrillization of the copolymer as measured using the fibrillization method is more than 20 cm, more than 21 cm, more than 22 cm, more than 23 cm, more than 24 cm, more than 25 cm, more than 26 cm, more than 27 cm, more than 28 cm, more than 29 cm, more than 30 cm, more than 32 cm, more than 34 cm, more than 36 cm, or more than 38 cm.

[0099] Incidentally, due to the necessity of the copolymer chains to slide over one another in order for fibrillization to occur, it is preferable that the copolymer does not form a single macromolecule through cross-linking since this would preclude the sliding of distinct

copolymer chains. Accordingly, in a preferable embodiment, the copolymer does not comprise any monomeric units derived from a cross-linking monomer. In some embodiments, the proportion of monomeric units in the copolymer derived from a crosslinking monomer is less than 0.25%, less than 0.2%, less than 0.15%, less than 0.1%, less than 0.05%, less than 0.02%, or less than 0.01% by mole, based on the total number of moles of monomeric units in the copolymer. In certain embodiments, the cross-linking monomer includes ethylene glycol diacrylate, ethylene glycol dimethacrylate, tri(propylene glycol) dimethacrylate, tetra(ethylene glycol) diacrylate, N,N'-methylenebis(acrylamide), poly(ethylene glycol) diacrylate, poly(ethylene glycol) dimethacrylate, poly(propylene glycol) diacrylate, poly(propylene glycol) dimethacrylate, trimethylolpropane ethoxylate triacrylate, trimethylolpropane propoxylate triacrylate, divinylbenzene, polyester dimethacrylate, divinyl ether, trimethylolpropane trimethacrylate, bisphenol A ethoxylate dimethacrylate, 1,4-butylene glycol diacrylate, 1,6-hexylene glycol diacrylate, di(trimethylolpropane) tetraacrylate, pentaerythritol tetraacrylate, pentaerythritol ethoxylate tetraacrylate, di(pentaerythritol) pentaacrylate, di(pentaerythritol) hexaacrylate, glycerol triacrylate, glycerol trimethacrylate, glycerol propoxylate triacrylate, derivatives thereof, isomers thereof, or combinations thereof.

[00100] The ratio of water to copolymer is crucial for the fibrillization process. Insufficient water results in strong interactions between copolymer chains, hindering fibrillization. Conversely, excessive water would mean that copolymer chains primarily interact with water molecules and with intermolecular interactions between copolymer chains lowered to the extent that such interactions are insufficient for fibrillization to occur. In some embodiments, this water is present as part of the binder composition of the present invention. In other embodiments, this water is present in an electrode material mixture comprising the copolymer disclosed herein.

[00101] In some embodiments, the solid content of the binder composition is from about 5% to about 30%, from about 8% to about 30%, from about 10% to about 30%, from about 12% to about 30%, from about 15% to about 30%, from about 18% to about 30%, from about 20% to about 30%, from about 22% to about 30%, from about 25% to about 30%, from about 28% to about 30%, from about 5% to about 28%, from about 8% to about 28%, from about 10% to about 28%, from about 12% to about 28%, from about 15% to about 28%, from about 18% to about 28%, from about 20% to about 28%, from about 22% to about 28%, from about 25% to about 28%, from about 5% to about 25%, from about 8% to about 25%, from about 10% to about 25%, from about 12% to about 25%, from about 15% to about 25%, from about 18% to about 25%, from about 20% to about 25%, from about 22% to about 25%, from about 5% to about 22%, from about 8% to about 22%, from about 10% to about 22%, from about 12% to about 22%, from about 15% to about 22%, from about 18% to about 22%, from about 20% to about 22%, from about 5% to about 20%, from about 8% to about 20%, from about 10% to

about 20%, from about 12% to about 20%, from about 15% to about 20%, from about 18% to about 20%, from about 5% to about 18%, from about 8% to about 18%, from about 10% to about 18%, from about 12% to about 18%, from about 15% to about 18%, from about 5% to about 15%, from about 8% to about 15%, from about 10% to about 15%, from about 12% to about 15%, from about 5% to about 12%, from about 8% to about 12%, from about 10% to about 12%, from about 5% to about 10%, from about 8% to about 10%, or from about 5% to about 8% by weight, based on the total weight of the binder composition.

[00102] In some embodiments, the solid content of the binder composition is less than 30%, less than 29%, less than 28%, less than 27%, less than 26%, less than 25%, less than 24%, less than 23%, less than 22%, less than 21%, less than 20%, less than 19%, less than 18%, less than 17%, less than 16%, less than 15%, less than 14%, less than 13%, less than 12%, less than 11%, less than 10%, less than 9%, or less than 8% by weight, based on the total weight of the binder composition. In some embodiments, the solid content of the binder composition is more than 5%, more than 6%, more than 7%, more than 8%, more than 9%, more than 10%, more than 11%, more than 12%, more than 13%, more than 14%, more than 15%, more than 16%, more than 17%, more than 18%, more than 19%, more than 20%, more than 21%, more than 22%, more than 23%, more than 24%, more than 25%, more than 26%, more than 27%, or more than 28% by weight, based on the total weight of the binder composition.

[00103] As discussed above, in some embodiments, the binder composition of the present invention can be added to an electrode active material to form an electrode material mixture, without the need to add additional solvent. In other embodiments, the copolymer disclosed herein is added in the form of a dry binder material to an electrode active material to form an electrode material mixture, in addition to a small amount of water in the specific proportion disclosed to enable fibrillization of the copolymer. The electrode active material can be either a cathode active material or an anode active material, resulting in the formation of a cathode mixture or an anode mixture respectively.

[00104] In the electrode material mixture, the copolymer disclosed herein, in conjunction with the water present in a specific proportion, readily fibrillizes and bind the various electrode components together. There is no particular limitation on the method used to produce the electrode material mixture, except that all electrode components should be well mixed to form the electrode material mixture; this can be achieved by using a homogenizer, for example. The use of a homogenizer facilitates the thorough blending of the electrode components, ensuring that they are well-mixed and homogeneously dispersed among the fibrillized copolymer network. This homogenization step is crucial for achieving a consistently and optimally distributed electrode material mixture. In turn, homogeneous distribution of the electrode components in a coated electrode layer plays an important role in fabricating batteries with good electrochemical performance.

[00105] In some embodiments, all the materials used to produce the electrode material mixture are added into the homogenizer in a single batch. This means that all the components are combined simultaneously and mixed together until a uniform mixture is obtained. In other embodiments, a step-wise approach is followed. Each component of the electrode material mixture, such as electrode active material, binder composition, and optionally conductive agent, can be added to the homogenizer in separate batches. Each batch may comprise more than one electrode component. This step-wise addition ensures that each component is properly integrated into the mixture, allowing for efficient mixing and uniform distribution of the materials. However, if the copolymer in the form of a dry binder material was used to form the electrode material mixture, it is preferable for all the electrode components to be added and mixed before adding water in order to ensure homogeneous distribution of the various electrode components.

[00106] The electrode active material can be either a cathode active material or an anode active material, resulting in the formation of a cathode mixture or an anode mixture respectively. Cathode active materials often exhibit instability in the presence of water, leading to unwanted reactions and the formation of impurities like lithium hydroxide (LiOH). These impurities have a negative impact on battery electrochemical performance, which is a significant drawback of water-based electrode slurries. To address this issue, various methods have been developed to waterproof cathode active materials. For instance, one approach involves coating cathode active materials to form cathode active materials with core-shell structures. However, these methods can increase production costs, prolong manufacturing times, and ironically could still potentially compromise battery performance.

[00107] The binder composition already contains a relatively low liquid content, and most of the water molecules in the binder composition are engaged in the intermolecular interactions with the copolymer chains, limiting their availability for interaction with the cathode active material when used to manufacture an electrode material mixture. Similarly, in other embodiments involving the preparation of an electrode material mixture using the copolymer in the form of a dry binder material, with a small amount of water added subsequently during the manufacture of the electrode material mixture in order to reach the desired liquid content, this amount is small enough to the extent that the water molecules would remain in the copolymer network and not react with the cathode active material. As a result, the degradation of cathode active material caused by its reaction with water can be effectively prevented. This approach then also offers advantages such as elimination of the necessity for protective measures for the cathode active materials, resulting in reduced material costs, and enabling shorter production times.

[00108] In some embodiments, the electrode active material is a cathode active material selected from the group consisting of $LiCoO_2$, $LiNiO_2$, $LiNi_xMn_yO_2$, $LiCo_xNi_yO_2$, $Li_{1+z}Ni_xMn_yCo_{1-x-y}O_2$ (NMC), $LiNi_xCo_yAl_zO_2$ (NCA), LiV_2O_5, $LiTiS_2$, $LiMoS_2$, $LiMnO_2$, $LiCrO_2$, $LiMn_2O_4$,

Li_2MnO_3, $LiFeO_2$, $LiFePO_4$, and combinations thereof, wherein each x is independently from 0.1 to 0.9; each y is independently from 0 to 0.9; and each z is independently from 0 to 0.4. In some embodiments, each x, y and z in the above general formula independently has a 0.01 interval. In other embodiments, the cathode active material is not $LiCoO_2$, $LiNiO_2$, LiV_2O_5, $LiTiS_2$, $LiMoS_2$, $LiMnO_2$, $LiCrO_2$, $LiMn_2O_4$, $LiFeO_2$, or $LiFePO_4$. In further embodiments, the cathode active material is not $LiNi_xMn_yO_2$, $Li_{1+z}Ni_xMn_yCo_{1-x-y}O_2$, $LiNi_xCo_yAl_zO_2$ or $LiCo_xNi_yO_2$, wherein each x is independently from 0.1 to 0.9; each y is independently from 0 to 0.9; and each z is independently from 0 to 0.4. In certain embodiments, the cathode active material is $Li_{1+x}Ni_aMn_bCo_cAl_{(1-a-b-c)}O_2$; wherein $-0.2\leq x\leq 0.2$, $0\leq a<1$, $0\leq b<1$, $0\leq c<1$, and $a+b+c\leq 1$. In some embodiments, the cathode active material has the general formula $LiMPO_4$, wherein M is selected from the group consisting of Fe, Co, Ni, Mn, Al, Mg, Zn, Ti, La, Ce, Sn, Zr, Ru, Si, Ge, or combinations thereof. In some embodiments, the cathode active material is selected from the group consisting of $LiFePO_4$, $LiCoPO_4$, $LiNiPO_4$, $LiMnPO_4$, $LiMnFePO_4$, $LiMn_xFe_{(1-x)}PO_4$, and combinations thereof; wherein $0<x<1$. In some embodiments, the cathode active material is $LiNi_xMn_yO_4$; wherein $0.1\leq x\leq 0.9$ and $0\leq y\leq 2$. In certain embodiments, the cathode active material is $xLi_2MnO_3\cdot(1-x)LiMO_2$, wherein M is selected from the group consisting of Ni, Co, Mn, and combinations thereof; and wherein $0<x<1$. In some embodiments, the cathode active material is $Li_3V_2(PO_4)_3$, or $LiVPO_4F$. In certain embodiments, the cathode active material has the general formula Li_2MsiO_4, wherein M is selected from the group consisting of Fe, Co, Mn, Ni, and combinations thereof.

[00109] In certain embodiments, the cathode active material is doped with a dopant selected from the group consisting of Co, Cr, V, Mo, Nb, Pd, F, Na, Fe, Ni, Mn, Al, Mg, Zn, Ti, La, Ce, Sn, Zr, Ru, Si, Ge, and combinations thereof. In some embodiments, the dopant is not Co, Cr, V, Mo, Nb, Pd, F, Na, Fe, Ni, Mn, Mg, Zn, Ti, La, Ce, Ru, Si, or Ge. In certain embodiments, the dopant is not Al, Sn, or Zr.

[00110] In certain embodiments, the cathode active material comprises or is a core-shell composite having a core and shell structure. In some embodiments, the core comprises one or more lithium transition metal oxides. In some embodiments, the shell comprises one or more lithium transition metal oxides and/or one or more transition metal oxides. In some embodiments, the one or more lithium transition metal oxides are selected from the group consisting of $Li_{1+x}Ni_aMn_bCo_cAl_{(1-a-b-c)}O_2$, $LiCoO_2$, $LiNiO_2$, $LiMnO_2$, $LiMn_2O_4$, Li_2MnO_3, $LiCrO_2$, $Li_4Ti_5O_{12}$, LiV_2O_5, $LiTiS_2$, $LiMoS_2$, $LiCo_aNi_bO_2$, $LiMn_aNi_bO_2$, and combinations thereof; wherein $-0.2\leq x\leq 0.2$, $0\leq a<1$, $0\leq b<1$, $0\leq c<1$, and $a+b+c\leq 1$. In some embodiments, each of the lithium transition metal oxides is independently doped with one or more dopants selected from the group consisting of Co, Cr, V, Mo, Nb, Pd, F, Na, Fe, Ni, Mn, Al, Mg, Zn, Ti, La, Ce, Sn, Zr, Ru, Si, Ge, and combinations thereof. In some embodiments, the one or more transition metal oxides are selected from the group consisting of Fe_2O_3, MnO_2, Al_2O_3, MgO, ZnO, TiO_2,

La_2O_3, CeO_2, SnO_2, ZrO_2, RuO_2, and combinations thereof.

[00111] In some embodiments, each of the shell and the core comprises one or more lithium transition metal oxides. In some embodiments, the lithium transition metal oxides in the core and the shell may be the same, or they may be different or partially different. In some embodiments, when the core or the shell comprises two or more lithium transition metal oxides, the two or more lithium transition metal oxides are uniformly distributed over the core or the shell. In certain embodiments, when the core or the shell comprises two or more lithium transition metal oxides, the two or more lithium transition metal oxides are not uniformly distributed over the core or the shell. In some embodiments, the cathode active material is not a core-shell composite.

[00112] In some embodiments, the diameter of the core is from about 1 μm to about 45 μm, from about 1 μm to about 35 μm, from about 1 μm to about 25 μm, from about 1 μm to about 15 μm, from about 1 μm to about 10 μm, from about 1 μm to about 5 μm, from about 1 μm to about 3 μm, from about 1 μm to about 40 μm, from about 10 μm to about 40 μm, from about 20 μm to about 40 μm, from about 30 μm to about 40 μm, from about 1 μm to about 30 μm, from about 5 μm to about 30 μm, from about 10 μm to about 30 μm, from about 20 μm to about 30 μm, from about 1 μm to about 25 μm, from about 5 μm to about 25 μm, from about 10 μm to about 25 μm, from about 15 μm to about 25 μm, from about 1 μm to about 20 μm, from about 5 μm to about 20 μm, from about 10 μm to about 20 μm, or from about 15 μm to about 20 μm. In certain embodiments, the thickness of the shell is from about 1 μm to about 20 μm, from about 3 μm to about 20 μm, from about 5 μm to about 20 μm, from about 10 μm to about 20 μm, from about 1 μm to about 15 μm, from about 3 μm to about 15 μm, from about 5 μm to about 15 μm, from about 10 μm to about 15 μm, from about 1 μm to about 10 μm, from about 3 μm to about 10 μm, or from about 5 μm to about 10 μm. In some embodiments, each of the diameter of the core and shell is independently above or be below the values of 1 μm, 2 μm, 3 μm, 4 μm, 5 μm, 6 μm, 7 μm, 8 μm, 9 μm, 10 μm, 11 μm, 12 μm, 13 μm, 14 μm, 15 μm, 16 μm, 17 μm, 18 μm, 19 μm, and 20 μm. In certain embodiments, the diameter or thickness ratio of the core and the shell are in the range of 15:85 to 85:15, 25:75 to 75:25, 30:70 to 70:30, or 40:60 to 60:40. In certain embodiments, the volume or weight ratio of the core and the shell is 95:5, 90:10, 80:20, 70:30, 60:40, 50:50, 40:60, or 30:70.

[00113] In some embodiments, the electrode active material is an anode active material selected the group consisting of natural graphite particulate, synthetic graphite particulate, hard carbon, soft carbon, mesocarbon microbeads (MCMB), Sn particulate, SnO_2, SnO, $Li_4Ti_5O_{12}$ particulate, Si particulate, Si-C composite particulate, and combinations thereof.

[00114] In certain embodiments, the anode active material is doped with a dopant. In some

embodiments, the dopant is selected from the group consisting of Fe, Ni, Mn, Al, Mg, Zn, Ti, La, Ce, Sn, Zr, Ru, and combinations thereof. In some embodiments, the dopant is B, Si, Ge, N, P, F, S, Cl, I, Se, or combinations thereof. In other embodiments, the anode active material is not doped. In some embodiments, the anode active material is not doped with Fe, Ni, Mn, Al, Mg, Zn, Ti, La, Ce, Sn, Zr, Ru, B, Si, Ge, N, P, F, S, Cl, I, or Se.

[00115] In some embodiments, the anode active material comprises or is a core-shell composite having a core and shell structure. In some embodiments, the core is selected from the group consisting of natural graphite particulate, synthetic graphite particulate, hard carbon, soft carbon, mesocarbon microbeads (MCMB), Sn particulate, SnO_2, SnO, $Li_4Ti_5O_{12}$ particulate, Si particulate, Si-C composite particulate, and combinations thereof. In some embodiments, the shell is selected from the group consisting of soft carbon, hard carbon, natural graphite particulate, synthetic graphite particulate, mesocarbon microbeads (MCMB), Kish graphite, pyrolytic carbon, mesophase pitches, mesophase pitch-based carbon fiber, Sn particulate, SnO_2, SnO, $Li_4Ti_5O_{12}$ particulate, Si particulate, Si-C composite particulate, and combinations thereof.

[00116] In some embodiments, the electrode material mixture additionally comprises a conductive agent. The presence of a conductive agent in the electrode material mixture may be advantageous since the conductive agent enhances the electrically-conducting properties of an electrode. Any suitable material can act as the conductive agent. In some embodiments, the conductive agent is a carbonaceous material. Some non-limiting examples include carbon, carbon black, graphite, expanded graphite, graphene, graphene nanoplatelets, carbon fibers, carbon nano-fibers, graphitized carbon flake, carbon tubes, carbon nanotubes, activated carbon, Super P, KS6, vapor grown carbon fibers (VGCF), mesoporous carbon, and combinations thereof. In certain embodiments, the conductive agent does not comprise a carbonaceous material.

[00117] In some embodiments, the conductive agent is a conductive polymer selected from the group consisting of polypyrrole, polyaniline, polyacetylene, polyphenylene sulfide (PPS), polyphenylene vinylene (PPV), poly(3,4-ethylenedioxythiophene) (PEDOT), polythiophene, and combinations thereof. In some embodiments, the conductive agent also acts as a binder composition. In some embodiments, the conductive agent is a mixture of a carbonaceous material and a conductive polymer. In other embodiments, the conductive agent does not comprise a conductive polymer.

[00118] The electrode material mixture can further comprise additives as needed to obtain the desired electrode properties. In certain embodiments, the additive is a conductive polymer that is used in addition to the conductive agent.

[00119] In some embodiments, the proportion of copolymer in the electrode material mixture is from about 2% to about 10%, from about 3% to about 10%, from about 4% to about

10%, from about 5% to about 10%, from about 6% to about 10%, from about 7% to about 10%, from about 8% to about 10%, from about 9% to about 10%, from about 2% to about 9%, from about 3% to about 9%, from about 4% to about 9%, from about 5% to about 9%, from about 6% to about 9%, from about 7% to about 9%, from about 8% to about 9%, from about 2% to about 8%, from about 3% to about 8%, from about 4% to about 8%, from about 5% to about 8%, from about 6% to about 8%, from about 7% to about 8%, from about 2% to about 7%, from about 3% to about 7%, from about 4% to about 7%, from about 5% to about 7%, from about 6% to about 7%, from about 2% to about 6%, from about 3% to about 6%, from about 4% to about 6%, from about 5% to about 6%, from about 2% to about 5%, from about 3% to about 5%, from about 4% to about 5%, from about 2% to about 4%, from about 3% to about 4%, or from about 2% to about 3% by weight, based on the total weight of the solid portion of the electrode material mixture.

[00120] In some embodiments, the proportion of copolymer in the electrode material mixture is less than 10%, less than 9%, less than 8%, less than 7%, less than 6%, less than 5%, less than 4%, or less than 3% by weight, based on the total weight of the solid portion of the electrode material mixture. In some embodiments, the proportion of copolymer in the electrode material mixture is more than 2%, more than 3%, more than 4%, more than 5%, more than 6%, more than 7%, more than 8%, or more than 9% by weight, based on the total weight of the solid portion of the electrode material mixture.

[00121] In some embodiments, the proportion of conductive agent in the electrode material mixture is from about 1% to about 10%, from about 2% to about 10%, from about 3% to about 10%, from about 4% to about 10%, from about 5% to about 10%, from about 6% to about 10%, from about 7% to about 10%, from about 8% to about 10%, from about 9% to about 10%, from about 1% to about 9%, from about 2% to about 9%, from about 3% to about 9%, from about 4% to about 9%, from about 5% to about 9%, from about 6% to about 9%, from about 7% to about 9%, from about 8% to about 9%, from about 1% to about 8%, from about 2% to about 8%, from about 3% to about 8%, from about 4% to about 8%, from about 5% to about 8%, from about 6% to about 8%, from about 7% to about 8%, from about 1% to about 7%, from about 2% to about 7%, from about 3% to about 7%, from about 4% to about 7%, from about 5% to about 7%, from about 6% to about 7%, from about 1% to about 6%, from about 2% to about 6%, from about 3% to about 6%, from about 4% to about 6%, from about 5% to about 6%, from about 1% to about 5%, from about 2% to about 5%, from about 3% to about 5%, from about 4% to about 5%, from about 1% to about 4%, from about 2% to about 4%, from about 3% to about 4%, from about 1% to about 3%, from about 2% to about 3%, or from about 1% to about 2% by weight, based on the total weight of the solid portion of the electrode material mixture.

[00122] In some embodiments, the proportion of conductive agent in the electrode material mixture is less than 10%, less than 9%, less than 8%, less than 7%, less than 6%, less

than 5%, less than 4%, less than 3%, or less than 2% by weight, based on the total weight of the solid portion of the electrode material mixture. In some embodiments, the proportion of conductive agent in the electrode material mixture is more than 1%, more than 2%, more than 3%, more than 4%, more than 5%, more than 6%, more than 7%, more than 8%, or more than 9% by weight, based on the total weight of the solid portion of the electrode material mixture.

[00123] In some embodiments, the proportion of electrode active material in the electrode material mixture is from about 80% to about 99%, from about 82% to about 99%, from about 84% to about 99%, from about 86% to about 99%, from about 88% to about 99%, from about 90% to about 99%, from about 92% to about 99%, from about 94% to about 99%, from about 96% to about 99%, from about 80% to about 96%, from about 82% to about 96%, from about 84% to about 96%, from about 86% to about 96%, from about 88% to about 96%, from about 90% to about 96%, from about 92% to about 96%, from about 94% to about 96%, from about 80% to about 94%, from about 82% to about 94%, from about 84% to about 94%, from about 86% to about 94%, from about 88% to about 94%, from about 90% to about 94%, from about 92% to about 94%, from about 80% to about 92%, from about 82% to about 92%, from about 84% to about 92%, from about 86% to about 92%, from about 88% to about 92%, from about 90% to about 92%, from about 80% to about 90%, from about 82% to about 90%, from about 84% to about 90%, from about 86% to about 90%, from about 88% to about 90%, from about 80% to about 88%, from about 82% to about 88%, from about 84% to about 88%, from about 86% to about 88%, from about 80% to about 86%, from about 82% to about 86%, from about 84% to about 86%, from about 80% to about 84%, from about 82% to about 84%, or from about 80% to about 82% by weight, based on the total weight of the solid portion of the electrode material mixture.

[00124] In some embodiments, the proportion of electrode active material in the electrode material mixture is less than 99%, less than 98%, less than 96%, less than 94%, less than 92%, less than 90%, less than 88%, less than 86%, less than 84%, or less than 82% by weight, based on the total weight of the solid portion of the electrode material mixture. In some embodiments, the proportion of electrode active material in the electrode material mixture is more than 80%, more than 82%, more than 84%, more than 86%, more than 88%, more than 90%, more than 92%, more than 94%, more than 96%, or more than 98% by weight, based on the total weight of the solid portion of the electrode material mixture.

[00125] It is worth to note that that the homogenizer used for preparing the dry mixture with the binder material disclosed herein does not necessitate high shearing forces, which differs from the conventional fluorinated binder material like PVDF when mixed with other solid particles in a solid state. In such cases, a significant amount of force is required to promote the fibrillization of the binder. This distinction is important because the absence of a requirement for high shearing forces simplifies the mixing process and reduces the risk of damaging the

electrode components. The ease of the binder material to fibrillize without the need for excessive force thus allows for a more efficient and controlled production method, ensuring the optimal dispersion and integration of the binder within the electrode material mixture. Any homogenizer that can promote homogeneous distribution of the electrode components within the electrode material mixture can be used herein. In some embodiments, the homogenizer is a screw mixer, a mill, a stirring mixer, or a planetary mixer. Grounding helps prevent any unwanted effects of static electricity, which could potentially disrupt the uniform distribution and stability of the electrode components within the mixture. Accordingly, in some embodiments, the homogenizer is grounded to reduce the effect of static electricity on the electrode material mixture. Furthermore, since the dry mixture and the electrical material mixture have different properties, in certain embodiments, a combination of multiple homogenizers may be used to more effectively combine the electrode components and water to form the electrode material mixture.

[00126] There is no particular limitation to the duration of mixing for preparing the electrode material mixture, but it is necessary for the copolymer to be adequately fibrillized in order to bind all the electrode components together and ensure the electrode material mixture is uniformly distributed at the end of the mixing process. Accordingly, it is essential to ensure that this mixing duration is sufficiently long to do so. In some embodiments, the homogenization time after adding all the electrode components to produce the electrode material mixture is from about 1 minute to about 6 hours. In certain embodiments, the homogenization time after adding all the electrode components to produce the electrode material mixture is less than 24 hours, less than 16 hours, less than 12 hours, less than 10 hours, less than 8 hours, less than 6 hours, less than 4 hours, less than 2 hours, less than 60 minutes, less than 30 minutes, or less than 15 minutes. In some embodiments, the homogenization time after adding all the electrode components to produce electrode material mixture is more than 1 minute, more than 5 minutes, more than 10 minutes, more than 15 minutes, more than 30 minutes, more than 60 minutes, more than 2 hours, more than 4 hours, more than 6 hours, more than 8 hours, more than 10 hours, more than 12 hours, or more than 16 hours.

[00127] Maintaining an appropriate temperature during homogenization helps promote efficient fibrillization and mixing, and enhances the homogeneity and integration of the electrode components in the electrode material mixture. Accordingly, the homogenizer can be equipped with a temperature control system to perform the homogenization and fibrillization under elevated temperature. In some embodiments, the electrode material mixture is homogenized at a temperature of from about 20 °C to about 50 °C. In certain embodiments, the electrode material mixture is homogenized at a temperature of less than 50 °C, or less than 40 °C. In some embodiments, the electrode material mixture is homogenized at a temperature of more than 20 °C, more than 25 °C, more than 30 °C, more than 35 °C, or more than 40 °C.

[00128] In the homogenization process, the rotational speed of the rotating elements of the

homogenizer can be customized within the specified range, wherein the homogenizer can be a screw mixer, stirring mixer or planetary mixer. By adjusting the rotational speed of the rotating element, the intensity of the mixing action can be modified. Higher speeds promote more vigorous mixing, while lower speeds offer a gentler mixing approach. This flexibility allows for precise control and optimization of the mixing conditions based on the specific requirements of the electrode material mixture. In certain embodiments, the rotational speed of each rotating element in the homogenizer is independently from about 10 rpm to about 3000 rpm, from about 10 rpm to about 100 rpm, from about 100 rpm to about 3000 rpm, from about 100 rpm to about 1000 rpm, from about 500 rpm to about 3000 rpm, or from about 500 rpm to about 2000 rpm. In certain embodiments, the rotational speed of each rotating element in the homogenizer is independently less than 3000 rpm, less than 2500 rpm, less than 2000 rpm, less than 1500 rpm, less than 1000 rpm, less than 500 rpm, less than 100 rpm, or less than 50 rpm. In certain embodiments, the rotational speed of each rotating element in the homogenizer is independently more than 100 rpm, more than 500 rpm, more than 1000 rpm, more than 1500 rpm, or more than 2000 rpm.

[00129] In some embodiments, the liquid content of the electrode material mixture only originates from the binder composition. Since no additional solvent is added in the manufacture of the electrode material mixture, the mixture would have an even lower liquid content. In certain embodiments, when a dry binder material consisting of the copolymer disclosed herein is used to manufacture the electrode material mixture instead, a small amount of water is indeed added but this amount is low. Accordingly, as discussed above, the water present would be sufficient to contribute to fibrillization of the copolymer without leading to side effects such as the degradation of the active material. Nonetheless, the relatively low amount of liquid present is important for achieving proper dispersion of the electrode components, adhesion of the binder material to the other electrode components, and subsequent processing steps in battery manufacturing, since it plays a crucial role in facilitating the binding and integration of the electrode components while ensuring the desired characteristics of the electrode material mixture.

[00130] In some embodiments, the liquid content of the electrode material mixture is from about 5% to about 22%, from about 8% to about 22%, from about 10% to about 22%, from about 12% to about 22%, from about 15% to about 22%, from about 18% to about 22%, from about 20% to about 22%, from about 5% to about 20%, from about 8% to about 20%, from about 10% to about 20%, from about 12% to about 20%, from about 15% to about 20%, from about 18% to about 20%, from about 5% to about 18%, from about 8% to about 18%, from about 10% to about 18%, from about 12% to about 18%, from about 15% to about 18%, from

about 5% to about 15%, from about 8% to about 15%, from about 10% to about 15%, from about 12% to about 15%, from about 5% to about 12%, from about 8% to about 12%, from about 10% to about 12%, from about 5% to about 10%, from about 8% to about 10%, or from about 5% to about 8% by weight, based on the total weight of the electrode material mixture. In certain embodiments, the liquid content of the electrode material mixture is less than 22%, less than 20%, less than 18%, less than 15%, less than 12%, less than 10%, or less than 8% by weight, based on the total weight of the electrode material mixture. In some embodiments, the liquid content of the electrode material mixture is more than 5%, more than 8%, more than 10%, more than 12%, more than 15%, more than 18%, or more than 20% by weight, based on the total weight of the electrode material mixture.

[00131] It was discovered that in order to maintain good fibrillization performance, it is necessary to increase the liquid content with increasing weight-average molecular weight of the copolymer. Accordingly, within the range of liquid contents of the electrode material mixture, if the copolymer has a relatively lower weight-average molecular weight, the liquid content of the electrode material mixture should tend towards the lower bound of the range disclosed. Conversely, if the copolymer has a relatively higher weight-average molecular weight, the liquid content of the electrode material mixture should tend towards the higher end of the range disclosed.

[00132] In some embodiments, after homogenization, the electrode material mixture is applied or calendered to form a free-standing electrode layer. The release film acts as a temporary support for the electrode material mixture. Subsequently, the free-standing electrode layer is laminated to a current collector and pressed together to form the final electrode structure. In some embodiments, formation of the free-standing electrode layer can be performed using a molding press, a roll press, or an extrusion press. In some embodiments, the molding press is a tablet press. In certain embodiments, the extrusion press is a screw extruder. The lamination step can be performed by applying pressure to bond the free-standing electrode layer to the current collector. This step ensures strong adhesion between the electrode layer and the current collector. The pressure can be applied using a laminating machine, hydraulic press, or other suitable equipment. The pressure and duration of pressing may vary depending on the materials used and the specific requirements of such materials. In some cases, the electrode may undergo a heat treatment process, such as drying or annealing, to enhance the electrode's stability and properties. The free-standing electrode layer can then be successfully applied onto the current collector, forming a cohesive and integrated electrode structure.

[00133] The current collector acts to collect electrons generated by electrochemical

reactions of the cathode active material or to supply electrons required for the electrochemical reactions. In some embodiments, the current collector can be in the form of a foil, sheet or film. In certain embodiments, the current collector is stainless steel, titanium, nickel, aluminum, copper, or alloys thereof; or electrically-conductive resin. In certain embodiments, the current collector has a two-layered structure comprising an outer layer and an inner layer, wherein the outer layer comprises a conductive material and the inner layer comprises an insulating material or another conductive material; for example, aluminum mounted with a conductive resin layer or a polymeric insulating material coated with an aluminum film. In some embodiments, the current collector has a three-layered structure comprising an outer layer, a middle layer and an inner layer, wherein the outer and inner layers comprise a conductive material and the middle layer comprises an insulating material or another conductive material; for example, a plastic substrate coated with a metal film on both sides. In certain embodiments, each of the outer layer, middle layer and inner layer is independently stainless steel, titanium, nickel, aluminum, copper, or alloys thereof; or electrically-conductive resin. In some embodiments, the insulating material is a polymeric material selected from the group consisting of polycarbonate, polyacrylate, polyacrylonitrile, polyester, polyamide, polystyrene, polyurethane, polyepoxy, poly(acrylonitrile-butadiene-styrene), polyimide, polyolefin, polyethylene, polypropylene, polyphenylene sulfide, poly(vinyl ester), polyvinyl chloride, polyether, polyphenylene oxide, cellulose polymer, and combinations thereof. In certain embodiments, the current collector has a structure comprising more than three layers.

[00134] In some embodiments, a conductive layer can be coated on the current collector to improve its current conductivity. In certain embodiments, the conductive layer comprises a material selected from the group consisting of carbon, carbon black, graphite, expanded graphite, graphene, graphene nanoplatelets, carbon fibers, carbon nano-fibers, graphitized carbon flake, carbon tubes, carbon nanotubes, activated carbon, mesoporous carbon, and combinations thereof. In some embodiments, the conductive layer does not comprise carbon, carbon black, graphite, expanded graphite, graphene, graphene nanoplatelets, carbon fibers, carbon nano-fibers, graphitized carbon flake, carbon tubes, carbon nanotubes, activated carbon, or mesoporous carbon.

[00135] In some embodiments, the conductive layer has a thickness of from about 0.5 μm to about 5.0 μm. Thickness of the conductive layer affects the volume occupied by the current collector within a battery, as well as the amount of the electrode active material needed and hence the capacity of the battery.

[00136] In certain embodiments, the thickness of the conductive layer on the current

collector is from about 0.5 μm to about 4.5 μm, from about 1.0 μm to about 4.0 μm, from about 1.0 μm to about 3.5 μm, from about 1.0 μm to about 3.0 μm, from about 1.0 μm to about 2.5 μm, from about 1.0 μm to about 2.0 μm, from about 1.1 μm to about 2.0 μm, from about 1.2 μm to about 2.0 μm, from about 1.5 μm to about 2.0 μm, from about 1.8 μm to about 2.0 μm, from about 1.0 μm to about 1.8 μm, from about 1.2 μm to about 1.8 μm, from about 1.5 μm to about 1.8 μm, from about 1.0 μm to about 1.5 μm, or from about 1.2 to about 1.5 μm. In some embodiments, the thickness of the conductive layer on the current collector is less than 4.5 μm, less than 4.0 μm, less than 3.5 μm, less than 3.0 μm, less than 2.5 μm, less than 2.0 μm, less than 1.8 μm, less than 1.5 μm, or less than 1.2 μm. In some embodiments, the thickness of the conductive layer on the current collector is more than 1.0 μm, more than 1.2 μm, more than 1.5 μm, more than 1.8 μm, more than 2.0 μm, more than 2.5 μm, more than 3.0 μm, or more than 3.5 μm.

[00137] The thickness of the current collector affects the volume it occupies within the battery, and hence the energy density of the battery. In some embodiments, the current collector has a thickness of from about 5 μm to about 30 μm. In certain embodiments, the current collector has a thickness of from about 5 μm to about 25 μm, from about 5 μm to about 20 μm, from about 5 μm to about 15 μm, from about 10 μm to about 30 μm, from about 10 μm to about 25 μm, or from about 10 μm to about 20 μm.

[00138] After the free-standing electrode layer has been applied onto the surface of the current collector, it may undergo a drying process. During the drying process, the water trapped between the copolymer chains, which was previously present, is eliminated. This removal of water leads to the development of strong intermolecular interactions among the different copolymer chains. As a result, these intermolecular interactions serve as anchors, holding the copolymer chains in position relative to one another and prevents further fibrillization. Moreover, the copolymer chains form bridge-link connections among the particles of the electrode components and with the current collector. Due to the random orientation of the copolymer chains, a meshed network is thus formed, which when combined with the bridge-links with the particles of the electrode components mean that the particles of the electrode components are in close contact with one another, thus forming a continuous and homogenous electrode layer. This close contact reduces interfacial resistance, facilitating efficient electron transfer within the electrode. Additionally, the electrode layer becomes firmly adhered to the current collector, ensuring mechanical stability and integrity.

[00139] Some non-limiting examples of equipment that can be used to heat and dry the coated film include a batch drying oven, a conveyor drying oven, and a microwave drying oven.

Some non-limiting examples of the conveyor drying oven include a conveyor hot air-drying oven, a conveyor resistance drying oven, a conveyor inductive drying oven, and a conveyor microwave drying oven. These ovens provide different heating mechanisms and can be used depending on specific requirements. There are no particular limitations on the conditions used to heat and dry the coated film, except that following the heating and drying process, the coated film should be securely fixed to the current collector without deformation or delamination. Alternatively, the laminated electrode, formed by combining the free-standing electrode layer and current collector, can undergo drying using a heated roller. Therefore, the temperature should be sufficiently high to complete the drying process within a reasonable timeframe. At the same time, the temperature should be low enough to prevent degradation of the electrode components in the resultant electrode layer and minimize the risk of uneven heating, which could lead to deformation or delamination of the electrode.

[00140] In some embodiments, the laminated electrode is heated and dried at a temperature of from about 30 °C to about 150 °C, from about 30 °C to about 100 °C, from about 50 °C to about 150 °C, or from about 50 °C to about 100 °C.

[00141] In some embodiments, the laminated electrode is heated and dried at a temperature of less than 150 °C, less than 140 °C, less than 130 °C, less than 120 °C, less than 110 °C, less than 100 °C, less than 90 °C, less than 80 °C, less than 70 °C, less than 60 °C, less than 50 °C, less than 40 °C, or less than 30 °C. In some embodiments, the coated film on the current collector is heated and dried at a temperature of more than 30 °C, more than 40 °C, more than 50 °C, more than 60 °C, more than 70 °C, more than 80 °C, more than 90 °C, more than 100 °C, more than 110 °C, more than 120 °C, more than 130 °C, more than 140 °C, or more than 150 °C.

[00142] In some embodiments, the laminated electrode is dried for a time period of from 10 seconds to 60 minutes. In certain embodiments, the laminated electrode is dried for a time period of from 1 minutes to 60 minutes, from 1 minutes to 50 minutes, from 1 minutes to 40 minutes, from 1 minutes to 30 minutes, from 1 minutes to 20 minutes, from 1 minutes to 10 minutes, or from 1 minutes to 5 minutes. In certain embodiments, the laminated electrode is dried for a time period of more than 10 seconds, more than 20 seconds, more than 30 seconds, more 40 seconds, more than 50 seconds, more than 1 minute, more than 2 minutes, more than 3 minutes, more than 4 minutes, more than 5 minutes, more than 10 minutes, more than 15 minutes, more than 20 minutes, more than 25 minutes, more than 30 minutes, more than 40 minutes, or more than 50 minutes. In certain embodiments, the laminated electrode is dried for a time period of less than 50 minutes, less than 40 minutes, less than 30 minutes, less than 20

minutes, less than 10 minutes, less than 5 minutes, less than 4 minutes, less than 3 minutes, less than 2 minutes, less than 1 minute, less than 50 seconds, less than 40 seconds, less than 30 seconds, or less than 20 seconds.

[00143] In some embodiments, after heating and drying, the electrode is compressed in order to enhance its density. The proportion of copolymer in the electrode layer is the same as the proportion of copolymer in the electrode material mixture described above. Likewise, the proportions of conductive agent and electrode active material in the electrode layer are the same as the respective proportions of conductive agent and electrode active material in the electrode material mixture described above. In any case, surprisingly, it was discovered that due to the presence of the copolymer in the electrode layer, the electrode layer could be made thicker compared to conventional electrode layers. This increased electrode layer thickness in turn increases battery overall energy density.

[00144] Regardless of whether the electrode has undergone compression, in certain embodiments, the thickness of the electrode layer is from about 20 μm to about 400 μm, from about 50 μm to about 400 μm, from about 100 μm to about 400 μm, from about 150 μm to about 400 μm, from about 200 μm to about 400 μm, from about 250 μm to about 400 μm, or from about 300 μm to about 400 μm. Regardless of whether the electrode has undergone compression, in some embodiments, the thickness of the electrode layer is more than 20 μm, more than 30 μm, more than 40 μm, more than 50 μm, more than 100 μm, more than 150 μm, more than 200 μm, more than 250 μm, more than 300 μm, or more than 350 μm. Regardless of whether the electrode has undergone compression, in some embodiments, the thickness of the electrode layer is less than 400 μm, less than 350 μm, less than 300 μm, less than 250 μm, less than 200 μm, less than 150 μm, less than 100 μm, less than 50 μm, or less than 30 μm.

[00145] Regardless of whether the electrode has undergone compression, in some embodiments, the surface density of the electrode layer is from about 1 mg/cm^2 to about 50 mg/cm^2, from about 3 mg/cm^2 to about 50 mg/cm^2, from about 5 mg/cm^2 to about 50 mg/cm^2, from about 10 mg/cm^2 to about 50 mg/cm^2, from about 15 mg/cm^2 to about 50 mg/cm^2, from about 20 mg/cm^2 to about 50 mg/cm^2, from about 30 mg/cm^2 to about 50 mg/cm^2, from about 40 mg/cm^2 to about 50 mg/cm^2, from about 1 mg/cm^2 to about 40 mg/cm^2, from about 1 mg/cm^2 to about 30 mg/cm^2, from about 5 mg/cm^2 to about 30 mg/cm^2, from about 10 mg/cm^2 to about 30 mg/cm^2, from about 15 mg/cm^2 to about 30 mg/cm^2, from about 20 mg/cm^2 to about 30 mg/cm^2, from about 1 mg/cm^2 to about 20 mg/cm^2, from about 5 mg/cm^2 to about 20 mg/cm^2, from about 10 mg/cm^2 to about 20 mg/cm^2, from about 1 mg/cm^2 to about 15 mg/cm^2, from about 3 mg/cm^2 to about 15 mg/cm^2, from about 5 mg/cm^2 to about 15 mg/cm^2, or from about 10 mg/cm^2 to

about 15 mg/cm^2.

[00146] Regardless of whether the electrode has undergone compression, in some embodiments, the surface density of the electrode layer is less than 50 mg/cm^2, less than 45 mg/cm^2, less than 40 mg/cm^2, less than 35 mg/cm^2, less than 30 mg/cm^2, less than 25 mg/cm^2, less than 20 mg/cm^2, less than 15 mg/cm^2, less than 10 mg/cm^2, less than 5 mg/cm^2, or less than 3 mg/cm^2. Regardless of whether the electrode has undergone compression, in some embodiments, the surface density of the electrode layer is more than 1 mg/cm^2, more than 3 mg/cm^2, more than 5 mg/cm^2, more than 10 mg/cm^2, more than 15 mg/cm^2, more than 20 mg/cm^2, more than 25 mg/cm^2, more than 30 mg/cm^2, more than 35 mg/cm^2, more than 40 mg/cm^2, or more than 45 mg/cm^2.

[00147] Regardless of whether the electrode has undergone compression, in some embodiments, the density of the electrode layer is from about 0.5 g/cm^3 to about 7.5 g/cm^3, from about 1 g/cm^3 to about 7.5 g/cm^3, from about 1.5 g/cm^3 to about 7.5 g/cm^3, from about 2 g/cm^3 to about 7.5 g/cm^3, from about 2.5 g/cm^3 to about 7.5 g/cm^3, from about 3 g/cm^3 to about 7.5 g/cm^3, from about 3.5 g/cm^3 to about 7.5 g/cm^3, from about 4 g/cm^3 to about 7.5 g/cm^3, from about 4.5 g/cm^3 to about 7.5 g/cm^3, from about 0.5 g/cm^3 to about 5.5 g/cm^3, from about 1 g/cm^3 to about 5.5 g/cm^3, from about 1.5 g/cm^3 to about 5.5 g/cm^3, from about 2 g/cm^3 to about 5.5 g/cm^3, from about 2.5 g/cm^3 to about 5.5 g/cm^3, from about 0.5 g/cm^3 to about 2.5 g/cm^3, from about 1 g/cm^3 to about 2.5 g/cm^3, or from about 1.5 g/cm^3 to about 2.5 g/cm^3. Regardless of whether the electrode has undergone compression, in some embodiments, the density of the electrode layer is less than 7.5 g/cm^3, less than 6.5 g/cm^3, less than 6 g/cm^3, less than 5.5 g/cm^3, less than 5 g/cm^3, less than 4.5 g/cm^3, less than 4 g/cm^3, less than 3.5 g/cm^3, less than 3 g/cm^3, less than 2.5 g/cm^3, less than 2 g/cm^3, less than 1.5 g/cm^3, or less than 1 g/cm^3. Regardless of whether the electrode has undergone compression, in some embodiments, the density of the electrode layer is more than 0.5 g/cm^3, more than 1 g/cm^3, more than 1.5 g/cm^3, more than 2 g/cm^3, more than 2.5 g/cm^3, more than 3 g/cm^3, more than 3.5 g/cm^3, more than 4 g/cm^3, more than 4.5 g/cm^3, more than 5 g/cm^3, or more than 5.5 g/cm^3.

[00148] In addition, as discussed above, an electrode prepared using the binder material disclosed herein exhibits strong adhesion of the electrode layer to the current collector. It is important for the electrode layer to have good peeling strength to the current collector to ensure that the electrode layer remains firmly attached to the current collector, preventing any delamination or separation. Maintaining a strong connection between the electrode layer and the current collector is crucial, as this significantly impacts both the mechanical stability of the electrodes and the battery's performance. Therefore, the electrodes should have sufficient

peeling strength to withstand the rigors of and demanding conditions encountered during battery manufacture, and must endure various handling and assembly steps without experiencing any detachment or weakening of the bonds between the electrode layer and the current collector. Delamination or separation of the electrode layer can lead to performance issues and reduced lifespan of the battery.

[00149] Regardless of whether the electrode has undergone compression, in some embodiments, the peeling strength between the current collector and the electrode layer is from about 1.0 N/cm to about 8.0 N/cm, from about 1.0 N/cm to about 6.0 N/cm, from about 1.0 N/cm to about 5.0 N/cm, from about 1.0 N/cm to about 4.0 N/cm, from about 1.0 N/cm to about 3.0 N/cm, from about 1.0 N/cm to about 2.5 N/cm, from about 1.0 N/cm to about 2.0 N/cm, from about 1.2 N/cm to about 3.0 N/cm, from about 1.2 N/cm to about 2.5 N/cm, from about 1.2 N/cm to about 2.0 N/cm, from about 1.5 N/cm to about 3.0 N/cm, from about 1.5 N/cm to about 2.5 N/cm, from about 1.5 N/cm to about 2.0 N/cm, from about 1.8 N/cm to about 3.0 N/cm, from about 1.8 N/cm to about 2.5 N/cm, from about 2.0 N/cm to about 6.0 N/cm, from about 2.0 N/cm to about 5.0 N/cm, from about 2.0 N/cm to about 3.0 N/cm, from about 2.0 N/cm to about 2.5 N/cm, from about 2.2 N/cm to about 3.0 N/cm, from about 2.5 N/cm to about 3.0 N/cm, from about 3.0 N/cm to about 8.0 N/cm, from about 3.0 N/cm to about 6.0 N/cm, or from about 4.0 N/cm to about 6.0 N/cm.

[00150] Regardless of whether the electrode has undergone compression, in some embodiments, the peeling strength between the current collector and the electrode layer is more than 1.0 N/cm, more than 1.2 N/cm, more than 1.5 N/cm, more than 2.0 N/cm, more than 2.2 N/cm, more than 2.5 N/cm, more than 3.0 N/cm, more than 3.5 N/cm, more than 4.0 N/cm, more than 4.5 N/cm, more than 5.0 N/cm, more than 5.5 N/cm, more than 6.0 N/cm, more than 6.5 N/cm, or more than 7.0 N/cm. Regardless of whether the electrode has undergone compression, in some embodiments, the peeling strength between the current collector and the electrode layer is less than 8.0 N/cm, less than 7.5 N/cm, less than 7 N/cm, less than 6.5 N/cm, less than 6.0 N/cm, less than 5.5 N/cm, less than 5.0 N/cm, less than 4.5 N/cm, less than 4.0 N/cm, less than 3.5 N/cm, less than 3.0 N/cm, less than 2.8 N/cm, less than 2.5 N/cm, less than 2.2 N/cm, less than 2.0 N/cm, less than 1.8 N/cm, or less than 1.5 N/cm. By ensuring that the electrodes exhibit adequate peeling strength, manufacturers can achieve reliable and stable electrode structures.

[00151] After the electrode has been manufactured, it can be assembled with separator and the corresponding counter electrode to create the electrode assembly. The selection of the counter electrode is determined by whether the electrode itself functions as a cathode or an anode.

[00152] The copolymer disclosed herein can be readily fibrillized, meaning that when an electrode material mixture incorporating the copolymer is utilized, whether by adding the copolymer as a dry binder material or as a binder composition, the resulting electrode layer demonstrates strong binding between its constituent components and exhibits robust adhesion to the current collector. This ensures that the electrode components are well integrated and firmly attached, preventing delamination or separation, and even allows for a thicker electrode layer, thereby increasing battery energy density. This electrode material mixture also has a low liquid content in the form of water, thereby minimizing the degradation of the cathode active material due to reaction with water and the resultant loss of battery performance. The aforementioned binder composition itself also has a lower liquid content, which contributes to enhanced efficiency in terms of storage and transport. Additionally, electrodes comprising the copolymer can also be readily recycled without resorting to environmentally harmful methods. By adopting the binder composition of the present invention, significant improvements can therefore be achieved in the electrode manufacturing process, without sacrificing the electrochemical performance of the battery.

[00153] The following examples are presented to exemplify embodiments of the invention but are not intended to limit the invention to the specific embodiments set forth. Unless indicated to the contrary, all parts and percentages are by weight. All numerical values are approximate. When numerical ranges are given, it should be understood that embodiments outside the stated ranges may still fall within the scope of the invention. Specific details disclosed in each example should not be construed as necessary features of the invention.

[00154] While the invention has been described with respect to a limited number of embodiments, the specific features of one embodiment should not be attributed to other embodiments of the invention. In some embodiments, the methods may include numerous steps not mentioned herein. In other embodiments, the methods do not include, or are substantially free of, any steps not enumerated herein. Variations and modifications from the described embodiments exist. The appended claims intend to cover all those modifications and variations as falling within the scope of the invention.

EXAMPLES

[00155] The peeling strengths of the electrode layers were measured by a tensile testing machine (DZ-106A, obtained from Dongguan Zonhow Test Equipment Co. Ltd., China). This test measures the average force required to peel an electrode layer from the current collector at 180° angle in newtons. The mean roughness depth (R_z) of the current collector is 2 μm. A strip of adhesion tape (3M; US; model no. 810) with a width of 18 mm and a length of 20 mm was attached onto the surface of the electrode layer. The electrode strip was clipped onto the testing

machine, and the tape was folded back on itself at 180 degrees, then placed in a moveable jaw and pulled at room temperature and a peel rate of 200 mm per minute. The maximum stripping force measured was taken as the peeling strength. Measurements were repeated three times to find the average value.

[00156] Fibrillization of a copolymer can be measured by the following procedure, through the use of a device as disclosed in CN 216978992 U. The binder composition (i.e., filtered sixth suspension) is normalized to a solid content of 12%, and then is poured into the vessel to a depth of 40 mm. Afterwards, a pin with a diameter of 2 mm is dipped into the binder composition to a depth of 10 mm, rested for 30 seconds, before being raised at a constant speed of 5 mm/s. Fibrillization of the copolymer could then be quantified by the maximum height above the liquid level to which the copolymer could be drawn by the pin without breaking.

[00157] The solid content of a binder composition or an electrode material mixture was calculated from the change in mass of the binder composition or electrode material mixture respectively before and after drying. Approximately 1 g of the binder composition or electrode material mixture was weighed in a weighing bottle and dried at 110 ± 5 °C and -0.09 MPa for more than 5 hours by a vacuum dryer, then cooled in a desiccator for about 15 minutes and its mass was measured. The difference in mass of the binder composition or electrode material mixture before and after drying was determined, and the solid content of the binder composition or electrode material mixture was calculated according to the following formula:

$$\text{Solid content of x} = \frac{\text{Mass of x after drying}}{\text{Mass of x before drying}} \times 100\%$$

wherein x may refer to the binder composition or electrode material mixture.

[00158] The weight-average molecular weight of the copolymers in the binder compositions was measured by gel permeation chromatography. A dry binder material was first dissolved in dimethylformamide at room temperature. Once dissolution was complete, the solution was gently filtered through a 0.45 μm filter to prepare a measurement sample. A polystyrene standard was used to prepare a calibration curve against which the weight-average molecular weight of the copolymer were calculated. The obtained measurement sample was analyzed with an Agilent PLgel 5um MIXED-C column. The flow rate was 1 ml/min and the weight of the sample was 2 mg. The detector used was Waters 2414 Refractive Index (RI) Detector and the detection temperature was 35 °C.

Example 1

A) Preparation of binder material

[00159] 11.90 g of sodium hydroxide (NaOH) was added into a round-bottom flask containing 380 g of distilled water. The mixture was stirred at 80 rpm for 30 mins to obtain a

first suspension.

[00160] 24.78 g of acrylic acid was added into the first suspension. The mixture was further stirred at 80 rpm for 30 mins to obtain a second suspension.

[00161] 28.76 g of acrylamide was dissolved in 10 g of DI water to form an acrylamide solution. Thereafter, all of the acrylamide solution was added into the second suspension. The mixture was further heated to 55 °C and stirred at 80 rpm for 45 mins to obtain a third suspension.

[00162] 13.95 g of acrylonitrile was added into the third suspension. The mixture was further stirred at 80 rpm for 10 mins to obtain a fourth suspension.

[00163] Further, 0.015 g of water-soluble free radical initiator (ammonium persulfate, APS; obtained from Aladdin Industries Corporation, China) was dissolved in 3 g of DI water and 0.0075 g of reducing agent (sodium bisulfite; obtained from Tianjin Damao Chemical Reagent Factory, China) was dissolved in 1.5 g of DI water. All of the APS solution and the sodium bisulfite solution were added into the fourth suspension. The mixture was stirred at 200 rpm for 24 h at 55 °C to obtain a fifth suspension.

[00164] After the complete reaction, the temperature of the fifth suspension was lowered to 25 °C. 3.72 g of NaOH was dissolved in 200 g of DI water and all of this sodium hydroxide solution was added dropwise into the fifth suspension to adjust pH to 7.3 and form a sixth suspension. The sixth suspension was filtered using 200 μm nylon mesh. The solid content of the filtered sixth suspension was 11.59 wt.%.

[00165] The filtered sixth suspension was dried in a vacuum dryer overnight at 60 °C, then ground using a mortar and pestle to form a dry binder material in the form of a fine powder. The weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 32 cm.

B) Preparation of positive electrode

[00166] 0.21 kg of conductive agent (KS6; obtained from ANR Technologies Pte. Ltd., Singapore), 0.15 kg of the dry binder material, and 2.64 kg of NMC532 (obtained from Shandong Tianjiao New Energy Co., Ltd, China), were sequentially added into a screw mixer. Subsequently, 0.42 kg of DI water was added to the mixed electrode components, and the electrode components were discharged from the screw mixer as an electrode material mixture. The process took about 5 minutes.

[00167] The homogenized electrode material mixture was pressed onto one side of an aluminum foil having a thickness of 16 μm as a current collector using a hot press. The coated film on the aluminum foil was vacuum dried at about 60 °C to form a cathode electrode layer. The thickness of the electrode layer was 76 μm. The surface density of the electrode layer was

about 20 mg/cm^2 and, and the density of the electrode layer was about 3.3 g/cm^3.

C) Preparation of Negative Electrode

[00168] A negative electrode slurry was prepared by mixing 93 wt.% of graphite (BTR New Energy Materials Inc., Shenzhen, Guangdong, China) with 1 wt.% carboxymethyl cellulose (CMC, BSH-12, DKS Co. Ltd., Japan) and 3 wt.% SBR (AL-2001, NIPPON A&L INC., Japan) as a binder, and 3 wt.% carbon black as a conductive agent in deionized water. The solid content of the anode slurry was 51.5 wt.%. The slurry was coated onto one side of a copper foil having a thickness of 8 μm using a doctor blade coater. The coated slurry on the copper foil was dried at about 80 °C by a hot air dryer to obtain a negative electrode. The electrode was then pressed to decrease the thickness of the anode electrode layer to 75 μm. The surface density of the anode electrode layer was about 10 mg/cm^2.

D) Assembly of coin cell

[00169] CR2032 coin-type Li cells were assembled in an argon-filled glove box. The coated cathode and anode sheets were cut into disc-form positive and negative electrodes respectively. The cathode and anode were kept apart by a separator. The separator was a ceramic coated microporous membrane made of nonwoven fabric (MPM, Japan), which had a thickness of about 25 μm. The electrode assembly was dried in a box-type resistance oven under vacuum (DZF-6020, obtained from Shenzhen Kejing Star Technology Co. Ltd., China) at 105 °C for about 16 hours. The water content of the electrode assembly after drying was 180 ppm.

[00170] An electrolyte was then injected into the case holding the packed electrodes under a high-purity argon atmosphere with a moisture and oxygen content of less than 3 ppm respectively. The electrolyte was a solution of $LiPF_6$ (1 M) in a mixture of ethylene carbonate (EC), ethyl methyl carbonate (EMC) and dimethyl carbonate (DMC) at a volume ratio of 1:1:1. After electrolyte filling, the coin cell was mechanically pressed using a punch tooling with a standard circular shape.

E) Electrochemical measurements

[00171] The coin cell was analyzed in a constant current mode using a multi-channel battery tester (BTS-4008-5V50mA, obtained from Neware Electronics Co. Ltd, China). The first cycle was completed at C/20, performed between 3.0 and 4.3 V at 25 °C, with the corresponding discharging capacity of the cycle measured. The coin cell was then cycled for 100 cycles and the 100 cycle capacity retention rate was measured. The electrochemical performance of the coin cell of Example 1 is shown in Table 1 below.

Preparation of Binder Material of Example 2

[00172] The binder material was prepared by the method described in Example 1, except that the polymerization process was controlled such that the weight-average molecular weight of the copolymer was about 480,000 g/mol. The copolymer was able to be fibrillized to 23 cm.

Preparation of Binder Material of Example 3

[00173] The binder material was prepared by the method described in Example 1, except that the polymerization process was controlled such that the weight-average molecular weight of the copolymer was about 630,000 g/mol. The copolymer was able to be fibrillized to 39 cm.

Preparation of Binder Material of Examples 4-5, 10-12, and Comparative Examples 10-11

[00174] The binder material was prepared by the method described in Example 1.

Preparation of Binder Material of Example 6

[00175] The binder material was prepared by the method described in Example 1, except that 11.09 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 23.32 g of acrylic acid was added in the preparation of the second suspension, 22.29 g of acrylamide was added in the preparation of the third suspension, and 19.86 g of acrylonitrile was added in the preparation of the fourth suspension. The solid content of the binder material was 11.25 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 26 cm.

Preparation of Binder Material of Example 7

[00176] The binder material was prepared by the method described in Example 1, except that 3.40 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 9.48 g of acrylic acid was added in the preparation of the second suspension, 36.67 g of acrylamide was added in the preparation of the third suspension, and 19.32 g of acrylonitrile was added in the preparation of the fourth suspension. The solid content of the binder material was 10.57 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 38 cm.

Preparation of Binder Material of Example 8

[00177] The binder material was prepared by the method described in Example 1, except that 13.92 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 28.43 g of acrylic acid was added in the preparation of the second suspension, 19.41 g of acrylamide was added in the preparation of the third suspension, and 18.25 g of acrylonitrile was added in the preparation of the fourth suspension. The solid content of the binder material was 11.32 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 22 cm.

Preparation of Binder Material of Example 9

[00178] The binder material was prepared by the method described in Example 1, except that 13.92 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 28.43 g of acrylic acid was added in the preparation of the second suspension, 17.97 g of acrylamide was added in the preparation of the third suspension, and 12.88 g of acrylonitrile and 10.22 g of tetramethylethylene was added in the preparation of the fourth suspension. The solid content of the binder material was 11.57 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 21 cm.

Preparation of Binder Material of Example 13

[00179] The binder material was prepared by the method described in Example 1, except all of the acrylic acid was replaced by 34.43 g of 2-ethylacrylic acid. The solid content of the binder material was 12.98 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 34 cm.

Preparation of Binder Material of Example 14

[00180] The binder material was prepared by the method described in Example 1, except all of the acrylic acid was replaced by 37.18 g of vinylsulfonic acid. The solid content of the binder material was 13.29 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 29 cm.

Binder Material of Comparative Example 1

[00181] PVDF-HFP with a weight-average molecular weight of about 400,000 g/mol was used as the dry binder material.

Binder Material of Comparative Example 2

[00182] CMC with a weight-average molecular weight of about 700,000 g/mol was used as the dry binder material.

Preparation of Binder Material of Comparative Example 3

[00183] The binder material was prepared by the method described in Example 1, except that 16.35 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 32.80 g of acrylic acid was added in the preparation of the second suspension, 15.10 g of acrylamide was added in the preparation of the third suspension, and 18.25 g of acrylonitrile was added in the preparation of the fourth suspension. The solid content of the binder material was 11.63 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 17 cm.

Preparation of Binder Material of Comparative Example 4

[00184] The binder material was prepared by the method described in Example 1, except

that 1.38 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 5.83 g of acrylic acid was added in the preparation of the second suspension, 38.11 g of acrylamide was added in the preparation of the third suspension, and 20.93 g of acrylonitrile was added in the preparation of the fourth suspension. The solid content of the binder material was 10.67 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 12 cm.

Preparation of Binder Material of Comparative Example 5

[00185] The binder material was prepared by the method described in Example 1, except that 13.92 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 28.43 g of acrylic acid was added in the preparation of the second suspension, 5.75 g of acrylamide was added in the preparation of the third suspension, and 28.45 g of acrylonitrile was added in the preparation of the fourth suspension. The solid content of the binder material was 11.18 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 3 cm.

Preparation of Binder Material of Comparative Example 6

[00186] The binder material was prepared by the method described in Example 1, except that 13.52 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 27.70 g of acrylic acid was added in the preparation of the second suspension, 11.50 g of acrylamide was added in the preparation of the third suspension, and 17.17 g of acrylonitrile and 11.92 g of tetramethylethylene was added in the preparation of the fourth suspension. The solid content of the binder material was 12.63 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 14 cm.

Preparation of Binder Material of Comparative Example 7

[00187] The binder material was prepared by the method described in Example 1, except that 8.66 g of sodium hydroxide (NaOH) was added in the preparation of the first suspension, 18.95 g of acrylic acid was added in the preparation of the second suspension, 18.69 g of acrylamide was added in the preparation of the third suspension, and 25.76 g of acrylonitrile was added in the preparation of the fourth suspension. The solid content of the binder material was 10.23 wt.%, the weight-average molecular weight of the copolymer was about 550,000 g/mol, and the copolymer was able to be fibrillized to 15 cm.

Preparation of Binder Material of Comparative Example 8

[00188] The binder material was prepared by the method described in Example 1, except that the polymerization process was controlled such that the weight-average molecular weight of the copolymer was about 320,000 g/mol. The copolymer was able to be fibrillized to 6 cm.

Preparation of Binder Material of Comparative Example 9

[00189] The binder material was prepared by the method described in Example 1, except that the polymerization process was controlled such that the weight-average molecular weight of the copolymer was about 750,000 g/mol. The copolymer was able to be fibrillized to 13 cm.

Assembly of coin cells of Examples 2-3, 6-9, 13-14, and Comparative Examples 2-9

[00190] The coin cells of the respective examples and comparative examples were assembled in the same manner as Example 1, except that the respective binder materials were used in the manufacture of the cathodes.

Assembly of coin cells of Example 4

[00191] The coin cells were assembled in the same manner as Example 1, except that 0.25 kg of DI water was added in the preparation of the electrode material mixture during the manufacture of the cathodes. The liquid content of the cathode electrode material mixture was about 7.7 wt.%.

Assembly of coin cells of Example 5

[00192] The coin cells were assembled in the same manner as Example 1, except that 0.80 kg of DI water was added in the preparation of the electrode material mixture during the manufacture of the cathodes. The liquid content of the cathode electrode material mixture was about 21.1 wt.%.

Assembly of coin cells of Example 10

[00193] The coin cells were assembled in the same manner as Example 1, except that all of the NMC532 was replaced by the same mass of NMC333 (obtained from Shandong Tianjiao New Energy Co., Ltd, China).

Assembly of coin cells of Example 11

[00194] The coin cells were assembled in the same manner as Example 1, except that all of the NMC532 was replaced by the same mass of NMC811 (obtained from Shandong Tianjiao New Energy Co., Ltd, China).

Assembly of coin cells of Example 12

[00195] The coin cells were assembled in the same manner as Example 5, except that all of the NMC532 was replaced by the same mass of LFP (Tianjin Sitelan Energy Technology Co. Ltd., China).

Assembly of coin cells of Comparative Example 1

[00196] The coin cells were assembled in the same manner as Example 1, except that N-methyl-2-pyrrolidone was used instead of water in the manufacture of the cathodes.

Assembly of coin cells of Comparative Example 10

[00197] The cathode electrode material mixture was prepared in the same manner as Example 1, except that only 0.10 kg of DI water was added. The liquid content of the cathode electrode material mixture was about 3.2 wt.%. A viable cathode could not be formed using this cathode electrode material mixture and accordingly the assembly of the coin cell was not proceeded further.

Assembly of coin cells of Comparative Example 11

[00198] The coin cells were assembled in the same manner as Example 1, except that 1.00 kg of DI water was added in the preparation of the electrode material mixture during the manufacture of the cathodes. The liquid content of the cathode electrode material mixture was about 25.0 wt.%.

Electrochemical measurements of Examples 2-11 and 13-14

[00199] The coin cells of the respective Examples were analyzed in the same manner as Example 1 and the results are shown in Table 1 below.

Electrochemical measurements of Examples 12

[00200] The coin cell of Example 12 was analyzed in the same manner as Example 1, except the voltage range was between 2.00 and 3.65 V, and the results are shown in Table 1 below.

Electrochemical measurements of Comparative Examples 1-11

[00201] The coin cells of the respective Comparative Examples were analyzed in the same manner as Example 1 where possible, and the results are shown in Table 2 below.

Table 1

	Proportion of monomeric units in copolymer comprising functional group (mol%)				M_w	Fibrillization performance (cm)	Cathode active material	Slurry Liquid Content (wt%)	0.05C Initial discharging capacity (mAh/g)	100 cycle capacity retention (%)
	Derived from acid	Derived from amide	Derived from nitrile	Derived from olefin						
Example 1	34	40	26	0	550,000	32	NMC532	12.3	165	85
Example 2	34	40	26	0	480,000	23	NMC532	12.3	166	81
Example 3	34	40	26	0	630,000	39	NMC532	12.3	165	86
Example 4	34	40	26	0	550,000	32	NMC532	7.7	164	84
Example 5	34	40	26	0	550,000	32	NMC532	21.1	161	83
Example 6	32	31	37	0	550,000	26	NMC532	12.3	169	85
Example 7	13	51	36	0	550,000	38	NMC532	12.3	167	88
Example 8	39	27	34	0	550,000	22	NMC532	12.3	167	84
Example 9	39	25	24	12	550,000	21	NMC532	12.3	159	80
Example 10	34	40	26	0	550,000	32	NMC333	12.3	154	91
Example 11	34	40	26	0	550,000	32	NMC811	12.3	205	81
Example 12	34	40	26	0	550,000	32	LFP	21.1	157	96
Example 13	34[1]	40	26	0	550,000	34	NMC532	12.3	163	84
Example 14	34[2]	40	26	0	550,000	29	NMC532	12.3	164	83

[1]: 2-ethylacrylic acid instead of acrylic acid.
[2]: vinylsulfonic acid instead of acrylic acid.

Table 2

	Proportion of monomeric units in copolymer comprising functional group (mol%)			M_w	Fibrillization performance (cm)	Cathode active material	Slurry Liquid Content (wt%)	0.05C Initial discharging capacity (mAh/g)	100 cycle capacity retention (%)
	Derived from acid	Derived from amide	Derived from nitrile	Derived from olefin					
Comparative Example 1[3]	/	/	/	/	400,000	33	NMC532	12.3	168	76
Comparative Example 2[4,5]	/	/	/	/	700,000	2	NMC532	12.3	N/A	N/A
Comparative Example 3	45	21	34	0	550,000	17	NMC532	12.3	162	71
Comparative Example 4	8	53	39	0	550,000	12	NMC532	12.3	143	69
Comparative Example 5[5]	39	8	53	0	550,000	3	NMC532	12.3	N/A	N/A
Comparative Example 6	38	16	32	14	550,000	14	NMC532	12.3	163	68
Comparative Example 7	26	26	48	0	550,000	15	NMC532	12.3	165	67
Comparative Example 8[5]	34	40	26	0	320,000	6	NMC532	12.3	N/A	N/A
Comparative Example 9	34	40	26	0	750,000	13	NMC532	12.3	157	70
Comparative Example 10[5]	34	40	26	0	550,000	32	NMC532	3.2	N/A	N/A
Comparative Example 11	34	40	26	0	550,000	32	NMC532	25.0	160	70

[3]: PVDF-HFP instead of a copolymer disclosed herein
[4]: CMC instead of a copolymer disclosed herein
[5]: A viable lithium-ion battery could not be formed.

CLAIMS

What is claimed is:

1. A binder composition for a secondary battery, comprising a copolymer and water; wherein the copolymer comprises a hydrophilic structural unit and a non-hydrophilic structural unit; and wherein the copolymer in the binder composition can be fibrillized in the presence of a shear force.

2. The binder composition according to claim 1, wherein the hydrophilic structural unit comprises monomeric units derived from a monomer comprising a hydrophilic functional group, wherein the monomer comprising a hydrophilic functional group is selected from the group consisting of an acid group-containing monomer, an amide group-containing monomer, and a combination thereof; and wherein the acid is selected from the group consisting of carboxylic acid, sulfonic acid, sulfuric acid, phosphonic acid, phosphoric acid, nitric acid, their salts, their derivatives, and combinations thereof.

3. The binder composition according to claim 1, wherein the non-hydrophilic structural unit comprises monomeric units derived from a monomer selected from the group consisting of a nitrile group-containing monomer, an olefin, and a combination thereof.

4. The binder composition according to claim 1, wherein the proportion of the hydrophilic structural unit in the copolymer is from about 60% to about 90% by mole, based on the total number of moles of monomeric units in the copolymer.

5. The binder composition according to claim 2, wherein the proportion of monomeric units in the copolymer derived from an acid group-containing monomer is from about 10% to about 40% by mole, based on the total number of moles of monomeric units in the copolymer in the binder composition.

6. The binder composition according to claim 2, wherein the proportion of monomeric units in the copolymer derived from an amide group-containing monomer is from about 10% to about 55% by mole, based on the total number of moles of monomeric units in the copolymer in the binder composition.

7. The binder composition according to claim 3, wherein the proportion of monomeric units in the copolymer derived from a nitrile group-containing monomer is from about 10% to

about 40% by mole, based on the total number of moles of monomeric units in the copolymer in the binder composition.

8. The binder composition according to claim 2, wherein the carboxylic acid is selected from the group consisting of acrylic acid, methacrylic acid, crotonic acid, 2-butyl crotonic acid, cinnamic acid, maleic acid, fumaric acid, itaconic acid, tetraconic acid, 2-ethylacrylic acid, isocrotonic acid, cis-2-pentenoic acid, trans-2-pentenoic acid, angelic acid, tiglic acid, 3,3-dimethyl acrylic acid, 3-propyl acrylic acid, trans-2-methyl-3-ethyl acrylic acid, cis-2-methyl-3-ethyl acrylic acid, 3-isopropyl acrylic acid, trans-3-methyl-3-ethyl acrylic acid, cis-3-methyl-3-ethyl acrylic acid, 2-isopropyl acrylic acid, trimethyl acrylic acid, 2-methyl-3,3-diethyl acrylic acid, 3-butyl acrylic acid, 2-butyl acrylic acid, 2-pentyl acrylic acid, 2-methyl-2-hexenoic acid, trans-3-methyl-2-hexenoic acid, 3-methyl-3-propyl acrylic acid, 2-ethyl-3-propyl acrylic acid, 2,3-diethyl acrylic acid, 3,3-diethyl acrylic acid, 3-methyl-3-hexyl acrylic acid, 3-methyl-3-tert-butyl acrylic acid, 2-methyl-3-pentyl acrylic acid, 3-methyl-3-pentyl acrylic acid, 4-methyl-2-hexenoic acid, 4-ethyl-2-hexenoic acid, 3-methyl-2-ethyl-2-hexenoic acid, 3-tert-butyl acrylic acid, 2,3-dimethyl-3-ethyl acrylic acid, 3,3-dimethyl-2-ethyl acrylic acid, 3-methyl-3-isopropyl acrylic acid, 2-methyl-3-isopropyl acrylic acid, trans-2-octenoic acid, cis-2-octenoic acid, trans-2-decenoic acid, α-acetoxyacrylic acid, β-trans-aryloxyacrylic acid, α-chloro-β-E-methoxyacrylic acid, methyl maleic acid, dimethyl maleic acid, phenyl maleic acid, bromo maleic acid, chloromaleic acid, dichloromaleic acid, fluoromaleic acid, difluoro maleic acid, nonyl hydrogen maleate, decyl hydrogen maleate, dodecyl hydrogen maleate, octadecyl hydrogen maleate, fluoroalkyl hydrogen maleate, and combinations thereof.

9. The binder composition according to claim 2, wherein the amide group-containing monomer is selected from the group consisting of acrylamide, methacrylamide, N-methyl methacrylamide, N-ethyl methacrylamide, N-n-propyl methacrylamide, N-isopropyl methacrylamide, isopropyl acrylamide, N-n-butyl methacrylamide, N-isobutyl methacrylamide, N,N-dimethyl acrylamide, N,N-dimethyl methacrylamide, N,N-diethyl acrylamide, N,N-diethyl methacrylamide, N-methylol methacrylamide, N-(methoxymethyl)methacrylamide, N-(ethoxymethyl)methacrylamide, N-(propoxymethyl)methacrylamide, N-(butoxymethyl)methacrylamide, N,N-dimethylaminopropyl methacrylamide, N,N-dimethylaminoethyl methacrylamide, N,N-dimethylol methacrylamide, diacetone methacrylamide, diacetone acrylamide, methacryloyl morpholine, N-hydroxyl methacrylamide, N-methoxymethyl acrylamide, N-methoxymethyl methacrylamide, N,N'-methylene-bis-acrylamide, N-hydroxymethyl acrylamide and combinations thereof.

10. The binder composition according to claim 3, wherein the nitrile group-containing monomer is selected from the group consisting of acrylonitrile, α-halogenoacrylonitrile, α-alkylacrylonitrile, α-chloroacrylonitrile, α-bromoacrylonitrile, α-fluoroacrylonitrile, methacrylonitrile, α-ethylacrylonitrile, α-isopropylacrylonitrile, α-n-hexylacrylonitrile, α-methoxyacrylonitrile, 3-methoxyacrylonitrile, 3-ethoxyacrylonitrile, α-acetoxyacrylonitrile, α-phenylacrylonitrile, α-tolylacrylonitrile, α-(methoxyphenyl)acrylonitrile, α-(chlorophenyl)acrylonitrile, α-(cyanophenyl)acrylonitrile, vinylidene cyanide and combinations thereof.

11. The binder composition according to claim 1, wherein the weight-average molecular weight of the copolymer in the binder composition is from about 400,000 g/mol to about 700,000 g/mol.

12. The binder composition according to claim 1, wherein the solid content of the binder composition is from about 5% to about 30% by weight, based on the total weight of the binder composition.

13. The binder composition according to claim 1, wherein the adhesive strength between the copolymer in the binder composition and an aluminium foil substrate is from about 1 N/cm to about 10 N/cm.

14. The binder composition according to claim 1, wherein the fibrillization of the copolymer occurs within a range of from about 20 cm to about 40 cm.

15. An electrode material mixture for a secondary battery, comprising an electrode active material, and the binder composition according to claim 1.

16. The electrode material mixture according to claim 15, further comprising a conductive agent.

17. The electrode material mixture according to claim 15, wherein the proportion of copolymer in the electrode material mixture is from about 2% to about 10% by weight, based on the total weight of the electrode material mixture.

18. The electrode material mixture according to claim 15, wherein the proportion of electrode active material in the electrode material mixture is from about 80% to about 99% by weight, based on the total weight of the electrode material mixture.

19. The electrode material mixture according to claim 16, wherein the proportion of

conductive agent in the electrode material mixture is from about 1% to about 10% by weight, based on the total weight of the electrode material mixture.

20. The electrode material mixture according to claim 15, wherein the electrode material mixture has a liquid content of from about 5% to about 22% by weight, based on the total weight of the electrode material mixture.