CATHODE ACTIVE MATERIAL, CATHODE SLURRY AND CATHODE FOR SECONDARY BATTERY

FIELD OF THE INVENTION

[001] The present invention relates to the field of batteries. In particular, this invention relates to cathode active materials for lithium-ion batteries and other metal-ion batteries.

BACKGROUND OF THE INVENTION

[002] Over the past decades, lithium-ion batteries (LIBs) have come to be 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, lithium-ion battery electrodes comprise an electrode layer that contains an electrode active material, conductive carbon, and binder material. The binder material provides good electrochemical stability to the electrode layer, holds the electrode components together and adheres the electrode components to the current collector. The electrode layer is often formed by coating a slurry onto the current collector and drying it, the slurry comprising a solvent that is used to suspend and/or dissolve the various electrode components for easy processing and coating. Polyvinylidene fluoride (PVDF) is one of the most commonly used binders in the commercial lithium-ion battery industry, but PVDF can only dissolve in specific organic solvents, such as N-methyl-2-pyrrolidone (NMP). Accordingly, organic solvents such as NMP are currently the solvents of choice for 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 generates significant costs in the manufacturing process since a large capital investment is required.

[004] Given the drawbacks of organic solvent-based electrode slurries, water-based slurries comprising an aqueous solvent (most commonly water) have been considered instead. Water-based electrode slurries provide many benefits over organic solvent-based slurries; for example, water is much safer and easier to handle compared to organic solvents such as NMP. These advantages in turn make water a more economical solvent as no vapor recovery system or other special equipment is required for handling it.

[005] Chinese Patent Application Publication CN103618063A discloses a water-based electrode slurry suitable for forming an electrode. The slurry comprises lithium iron phosphate

(LFP) as a cathode active material, a conductive agent, and polyacrylonitrile (PAN) as a binder material. However, although LFP may be relatively chemically stable in water, many other cathode active materials are unstable in water and react with it to form unwanted impurities, such as lithium hydroxide (LiOH). This is particularly true for cathode active materials with a high nickel content, such as lithium nickel-manganese-cobalt oxides (NMC). The presence of these impurities leads to decreased battery electrochemical performance. NMC has a high specific capacity and is therefore a highly promising choice as a cathode active material suitable for producing batteries with excellent electrochemical performance. Yet, the reactivity of NMC with water presents a significant obstacle to the use of NMC in water-based electrode slurries.

[006] The reactivity of cathode active materials with water also presents difficulties in terms of logistics, since it means that a water-based cathode slurry has to be used to form a cathode as quickly as possible after being manufactured, so as to minimize the degradation of cathode active material in the water-based slurry. As a result, due to the long periods of time involved, the average water-based cathode slurry cannot be stored or transported from one location to another, rendering the cathode production process highly inflexible.

[007] In view of the above, the present inventors have studied the subject intensively. It was found that by pre-treating a cathode active material to form a self-assembled monolayer (SAM)—a coating layer on the cathode active material—that comprises a phosphorus-containing compound, the reaction of the cathode active material with water can be suppressed, thereby enabling such a coated cathode active material to be used in a water-based electrode slurry. This would in turn lead to better production flexibility. Furthermore, batteries comprising such a coated cathode active material were found to have exceptional electrochemical performance due to decreased cathode active material degradation in the slurry forming step.

[008] Accordingly, it is an aim of the present invention to present a coated cathode active material suitable for use in water-based electrode slurries that can be used to produce a cathode for a lithium-ion battery.

SUMMARY OF THE INVENTION

[009] The aforementioned needs are met by various aspects and embodiments disclosed herein. In one aspect, provided herein is a coated cathode active material in the form of particles, wherein a coated cathode active material particle comprises a cathode active material particle and a coating layer that is derived from a phosphorus-containing compound and surrounds the cathode active material particle. The coating layer reduces degradation of the cathode active material that results from reaction with water, thereby enabling the coated cathode active material to be used in a water-based electrode slurry. In some embodiments, said coating layer is formed through the mechanism of self-assembly, and the coating layer can therefore be termed a

self-assembled monolayer (SAM). In another aspect, a water-based electrode slurry comprising such a coated cathode active material is also disclosed.

BRIEF DESCRIPTION OF THE DRAWINGS

[0010] Figure 1 shows the amount of degradation of the coated cathode active materials prepared in Examples 3 and 4 in water, normalized with respect to the amount of degradation of uncoated cathode active material of Comparative Example 1 in water.

[0011] Figure 2 shows a Nyquist plot of the real and imaginary components of the impedance of a coin cell prepared according to Example 4 at different current frequencies.

[0012] Figure 3 shows a Nyquist plot of the real and imaginary components of the impedance of a coin cell prepared according to Comparative Example 14 at different current frequencies.

DETAILED DESCRIPTION OF THE INVENTION

[0013] In one aspect, provided herein is a coated cathode active material in the form of particles, wherein a coated cathode active material particle comprises a cathode active material particle and a coating layer that is derived from a phosphorus-containing compound and surrounds the cathode active material particle, and wherein the coating layer is formed through a mechanism of self-assembly. The coating layer can suppress the reaction of the cathode active material with water and therefore enable the coated cathode active material to be used in a water-based electrode slurry. In another aspect, a water-based electrode slurry comprising the coated cathode active material is disclosed.

[0014] The term "electrode" refers to a "cathode" or an "anode."

[0015] The term "electrode component" refers to any substance that is present in an electrode layer or an electrode slurry, including but not limited to electrode active materials, conductive agents, binders and solvents.

[0016] The term "positive electrode" is used interchangeably with cathode. Likewise, the term "negative electrode" is used interchangeably with anode.

[0017] The term "binder" or "binder material" refers to a chemical compound, a mixture of compounds or a polymer that is used to hold an electrode material and/or a conductive agent in place and adhere them onto a conductive metal part to form an electrode. In some embodiments, the electrode does not comprise any conductive agent. In some embodiments, the binder material forms a solution or colloid in an aqueous solvent such as water. Such a solution or colloid may be referred to herein as a "binder composition".

[0018] The term "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 "polymer" refers to a polymeric compound prepared by polymerizing monomers, whether of the same type or of different types. The generic term "polymer" embraces the terms "homopolymer" and "copolymer".

[0020] The term "homopolymer" refers to a polymer prepared by the polymerization of the same type of monomer.

[0021] The term "copolymer" refers to a polymer prepared by the polymerization of two or more different types of monomers.

[0022] The term "binder polymer" refers to a polymer that is used as binder material. The term "binder copolymer" then refers to the copolymer specifically in a binder material comprising said copolymer.

[0023] As used herein, the term "cathode active material" refers to a substance containing a lithium metal oxide. In some embodiments, the cathode active material comprises primary particles, secondary particles, tertiary particles or a combination thereof. The term "primary particle" refers to an independently existing particle which is not composed of an aggregate. The term "secondary particle" refers to an aggregate particle formed by agglomeration of primary particles, and the term "tertiary particle" refers to an aggregate particle formed by agglomeration of secondary particles. In some embodiments, the cathode active material is composed of primary particles only. In some embodiments, the secondary particles are formed by agglomeration of several primary particles.

[0024] The term "particle size D50" refers to a volume-based accumulative 50% size (D50), which is a particle size at a point of 50% on an accumulative curve (i.e., a diameter of a particle in the 50th percentile (median) of the volumes of particles) when the accumulative curve is drawn so that a particle size distribution is obtained on the volume basis and the whole volume is 100%. Furthermore, with respect to the electrode active material of the present invention, the particle size D50 means a volume-averaged particle size of secondary particles which can be formed by mutual agglomeration of primary particles, and in a case where the particles are composed of the primary particles only, it means a volume-averaged particle size of the primary particles.

[0025] The term "alkyl" or "alkyl group" refers to a univalent group having the general formula C_nH_{2n+1} (n being an integer) and derived from the removal of a hydrogen atom from any carbon atom of a saturated aliphatic hydrocarbon, which may be branched or unbranched. In some embodiments, the alkyl group is a straight-chain alkyl group, i.e., the group is not

branched. Some examples of straight-chain alkyl groups include methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl and decyl. In other embodiments, the alkyl group is a branched alkyl group, i.e., side chains are present. Some examples of branched alkyl groups include isopropyl, 2-methyl-1-propyl, 2-methyl-2-propyl, 2-methyl-1-butyl, 3-methyl-1-butyl, 2-methyl-3-butyl, 2,2-dimethyl-1-propyl, 2-methyl-1-pentyl, 3-methyl-1-pentyl, 4-methyl-1-pentyl, 2-methyl-2-pentyl, 3-methyl-2-pentyl, 4-methyl-2-pentyl, 2,2-dimethyl-1-butyl, 3,3-dimethyl-1-butyl, 2-ethyl-1-butyl, isobutyl, *t*–butyl, isopentyl and neopentyl. An alkyl group can be unsubstituted or substituted with one or more suitable substituents.

[0026] 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. Examples of cycloalkyl groups include, but are not limited to, C_3-C_7 cycloalkyl groups, such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, and cycloheptyl; C_3-C_7 cycloalkenyl groups, such as cyclopropenyl, cyclobutenyl, cyclopentenyl, cyclohexenyl, and cycloheptenyl; and cyclic and bicyclic terpenes. A cycloalkyl group can be unsubstituted or substituted by one or two suitable substituents.

[0027] 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, which may be branched or unbranched. Non-limiting examples of alkenyl include vinyl, 1-propenyl, 2-propenyl, isobutenyl and butadienyl. 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, which may be branched or unbranched. Non-limiting examples of alkenyl include ethynyl, 3-methylpent-1-yn-3-yl ($HC\equiv C\text{-}C(CH_3)(C_2H_5)$-) and butadiynyl. Further, 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.

[0028] The term "alkoxy" refers to an alkyl group with an oxygen atom attached to its principal carbon chain. Some non-limiting examples of the alkoxy group include methoxy, ethoxy, propoxy, butoxy, and the like. An alkoxy may be substituted or unsubstituted; the substituent may be, but is not limited to, deuterium, hydroxy, amino, halo, cyano, alkoxy, alkyl, alkenyl, alkynyl, mercapto, nitro, and the like.

[0029] 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. The alkylene group is exemplified by methylene ($-CH_2-$), ethylene ($-CH_2CH_2-$), isopropylene ($-CH(CH_3)CH_2-$), and the like. The alkylene group is optionally substituted with one or more substituents described herein.

[0030] The term "aryl" or "aryl group" refers to an organic radical derived from a monocyclic or polycyclic aromatic hydrocarbon by removing a hydrogen atom. Non-limiting examples of an aryl group include phenyl, naphthyl, benzyl, tolanyl, sexiphenyl, phenanthrenyl, anthracenyl, coronenyl, and tolanylphenyl. An aryl group can be unsubstituted or substituted with one or more suitable substituents.

[0031] The term "alkylamino" embraces "*N*-alkylamino" and "*N,N*-dialkylamino", wherein the amino group is independently substituted with one or two alkyl groups, respectively. Some non-limiting examples of the alkylamino group include monoalkylamino or dialkylamino such as *N*-methylamino, *N*-ethylamino, *N,N*-dimethylamino, *N,N*-diethylamino, and the like. The alkylamino group is optionally substituted with one or more substituents described herein.

[0032] The term "alkylthio" refers to a group containing a linear or branched alkyl group attached to a divalent sulfur atom. Some non-limiting examples of the alkylthio group include methylthio (CH_3S-). The alkylthio group is optionally substituted with one or more substituents described herein.

[0033] 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, for example N (as in 3,4-dihydro-2*H*-pyrrolyl), NH (as in pyrrolidinyl) or NR (as in *N*-substituted pyrrolidinyl).

[0034] The term "carbonyl" refers to -(C=O)-.

[0035] The term "acyl" refers to -(C=O)-R.

[0036] The term "amido" refers to -NH(C=O)-R.

[0037] The term "aliphatic" refers to a non-aromatic hydrocarbon or groups derived therefrom. Some non-limiting examples of aliphatic compounds include alkanes, alkenes, alkynes, alkyl, alkenyl, alkynyl, an alkylene group, an alkenylene group, or an alkynylene group.

[0038] The term "aromatic" refers to compounds and groups comprising aromatic hydrocarbon rings, optionally including heteroatoms or substituents. Examples of such groups include, but are not limited to, phenyl, tolyl, biphenyl, o-terphenyl, m-terphenyl, p-terphenyl, naphthyl, anthryl, phenanthryl, pyrenyl, triphenylenyl, and derivatives thereof.

[0039] The term "substituted" as 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. This second chemical moiety is known as a "substituent". Examples of substituents include, but are not limited to, halogen; alkyl; heteroalkyl; alkenyl; alkynyl; aryl, heteroaryl, hydroxyl; alkoxyl; amino; nitro; thiol; thioether; imine; cyano; amido; phosphonato; phosphinato; carboxyl; thiocarbonyl; sulfonyl; sulfonamide; acyl; formyl; acyloxy;

alkoxycarbonyl; oxo; haloalkyl (*e.g.*, trifluoromethyl); carbocyclic cycloalkyl, which can be monocyclic or fused or non-fused polycyclic (*e.g.*, cyclopropyl, cyclobutyl, cyclopentyl or cyclohexyl) or a heterocycloalkyl, which can be monocyclic or fused or non-fused polycyclic (*e.g.*, pyrrolidinyl, piperidinyl, piperazinyl, morpholinyl or thiazinyl); carbocyclic or heterocyclic aryl, which can be monocyclic or fused or non-fused polycyclic(*e.g.*, phenyl, naphthyl, pyrrolyl, indolyl, furanyl, thiophenyl, imidazolyl, oxazolyl, isoxazolyl, thiazolyl, triazolyl, tetrazolyl, pyrazolyl, pyridinyl, quinolinyl, isoquinolinyl, acridinyl, pyrazinyl, pyridazinyl, pyrimidinyl, benzimidazolyl, benzothiophenyl or benzofuranyl); amino (primary, secondary or tertiary); 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, for example $-OCH_2O-$. 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.

[0040] The term "straight-chain" refers to an organic compound or a moiety that does not comprise a side chain or a cyclic structure; i.e., the carbon atoms of the organic compound or moiety all form a single linear arrangement. A straight-chain compound or moiety can be substituted or unsubstituted, as well as saturated or unsaturated.

[0041] The term "halogen" or "halo" refers to F, Cl, Br or I.

[0042] The term "structural unit" refers to the total monomeric units contributed by the same monomer type in a polymer.

[0043] The term "acid salt group" refers to a functional group derived from an acid functional group, wherein the proton of the acid functional group is replaced with a cation. In some embodiments, the proton of the acid functional group is replaced with a metal cation. In some embodiments, the proton of the acid functional group is replaced with an ammonium ion.

[0044] The term "homogenizer" refers to an equipment that can be used to homogenize materials, i.e., to distribute materials uniformly throughout a fluid. Any conventional homogenizer can be used for the method disclosed herein. Some non-limiting examples of homogenizers include stirring mixers, planetary stirring mixers, blenders and ultrasonicators.

[0045] The term "planetary mixer" refers to an equipment that can be used to mix or stir different materials to produce a homogeneous mixture, the equipment comprising a vessel and blades that conduct a planetary motion within the vessel. In some embodiments, the planetary mixer comprises at least one planetary blade and at least one high-speed dispersion blade. The planetary and the high-speed dispersion blades rotate on their own axes as well as revolve continuously within the vessel. The rotation speed can be expressed in unit of rotations per minute (rpm), which refers to the number of rotations that a rotating body completes in one

minute.

[0046] The term "ultrasonicator" refers to an equipment that can apply ultrasound energy to agitate particles in a sample. Some non-limiting examples of the ultrasonicator include an ultrasonic bath, a probe-type ultrasonicator and an ultrasonic flow cell.

[0047] The term "ultrasonic bath" refers to an apparatus surrounded by a wall and is designed to hold a fluid within the wall. Ultrasonic energy is transmitted via the wall of the ultrasonic bath into the fluid.

[0048] The term "probe-type ultrasonicator" refers to an ultrasonic probe immersed into a fluid for direct sonication. The term "direct sonication" means that the ultrasound is directly produced in the fluid.

[0049] The term "ultrasonic flow cell" or "ultrasonic reactor chamber" refers to an apparatus through which sonication processes can be carried out in a flow-through mode. In some embodiments, the ultrasonic flow cell is in a single-pass, multiple-pass, or recirculating configuration.

[0050] The term "applying" refers to an act of laying or spreading a substance on a surface.

[0051] The term "current collector" refers to any conductive substrate, which 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 it 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.

[0052] 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, the electrode layer is made by applying a coating on to the current collector and drying the coating. In some embodiments, the electrode layer is located on the surface of the current collector. In other embodiments, a three-dimensional porous current collector is coated conformally with an electrode layer.

[0053] The term "doctor blading" refers to a process for fabrication of large area films on rigid or flexible substrates. A coating thickness can be controlled by an adjustable gap width between a coating blade and a coating surface, which allows the deposition of variable wet layer thicknesses.

[0054] The term "slot-die coating" refers to a process for fabrication of large area films on rigid or flexible substrates. A slurry is applied to the substrate by continuously pumping slurry through a nozzle onto the substrate, which is mounted on a roller and constantly fed toward the nozzle. The thickness of the coating is controlled by various methods, such as altering the slurry flow rate or the speed of the roller.

[0055] 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.

[0056] The term "solid content" refers to the amount of non-volatile material in a mixture that remains after evaporation. The term "solid portion" with respect to the mixture then refers to this non-volatile material.

[0057] The term "peeling strength" refers to the amount of force required to separate two materials that are bonded to each other, such as a current collector and an electrode layer. It is a measure of the adhesion strength between such two materials and is usually expressed in N/cm.

[0058] 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.

[0059] 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.

[0060] 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 per unit weight, usually expressed in Ah/kg or mAh/g.

[0061] In the following description, all numbers disclosed herein are approximate values, regardless of 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 above is also specifically disclosed.

[0062] In the present description, all references to the singular include references to the plural and vice versa.

[0063] 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 NMP as the solvent, but the use of NMP presents significant environmental, health and safety risks, in addition to incurring additional costs associated with a recovery system. Therefore, water-based electrode slurries comprising an aqueous solvent, such as water, have been proposed as an alternative. Batteries that comprise electrodes produced using such electrode slurries have excellent battery performance and the electrode production process has reduced environmental, health and safety risks.

[0064] In some embodiments, the electrode active material is a cathode active material. In certain embodiments, the cathode active material is in the form of particles. In some embodiments, the cathode active material is in the form of primary particles, secondary particles, tertiary particles, or combinations thereof. An electrode slurry comprising a cathode active material can also be known as a cathode slurry. Although the use of water as a solvent to produce an electrode slurry does pose significant benefits in terms of safety and environmental compatibility, many cathode active materials are unstable in water and would degrade by reacting with water to form lithium ions, which in turn form unwanted impurities such as lithium hydroxide (LiOH). This is particularly true for cathode active materials with a high nickel content, such as NMC. When a water-based electrode slurry comprising such a cathode active material with a high nickel content is used to produce an electrode, batteries comprising such electrodes have worsened electrochemical performance due to degradation of the cathode active material in water within the slurry. Furthermore, in order to minimize cathode active material degradation, water-based cathode slurries have to be used immediately following production, making the production process highly inflexible.

[0065] In some embodiments, the cathode active material is selected from the group consisting of $LiCoO_2$, $LiNiO_2$, $LiNi_{1-x}M_xO_2$, $LiNi_xMn_yO_2$, $LiCo_xNi_yO_2$, $Li_{1+z}Ni_xMn_yCo_{1-x-y}O_2$, $LiNi_xCo_yAl_zO_2$, 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; each z is independently from 0 to 0.4; and M is selected from the group consisting of Co, Mn, Al, Fe, Ti, Ga, Mg, and combinations thereof. In certain embodiments, each x in the above general formula is independently selected from 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375, 0.4, 0.425, 0.45, 0.475, 0.5, 0.525,

0.55, 0.575, 0.6, 0.625, 0.65, 0.675, 0.7, 0.725, 0.75, 0.775, 0.8, 0.825, 0.85, 0.875 and 0.9; each y in the above general formula is independently selected from 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375, 0.4, 0.425, 0.45, 0.475, 0.5, 0.525, 0.55, 0.575, 0.6, 0.625, 0.65, 0.675, 0.7, 0.725, 0.75, 0.775, 0.8, 0.825, 0.85, 0.875 and 0.9; each z in the above general formula is independently selected from 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375 and 0.4. In some embodiments, each x, y and z in the above general formula independently has a 0.01 interval.

[0066] In certain embodiments, the cathode active material is selected from the group consisting of $LiCoO_2$, $LiNiO_2$, $LiNi_xMn_yO_2$, $Li_{1+z}Ni_xMn_yCo_{1-x-y}O_2$ (NMC), $LiNi_xCo_yAl_zO_2$ (NCA), LiV_2O_5, $LiTiS_2$, $LiMoS_2$, $LiMnO_2$, $LiCrO_2$, $LiMn_2O_4$, $LiFeO_2$, $LiFePO_4$, $LiCo_xNi_yO_2$, and combinations thereof, wherein each x is independently from 0.4 to 0.6; each y is independently from 0.2 to 0.4; and each z is independently from 0 to 0.1. 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 0.45; and each z is independently from 0 to 0.2. 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 $Li_{1+x}Ni_aMn_bCo_cAl_{(1-a-b-c)}O_2$, with $0.33\leq a\leq 0.92$, $0.33\leq a\leq 0.9$, $0.33\leq a\leq 0.8$, $0.4\leq a\leq 0.92$, $0.4\leq a\leq 0.9$, $0.4\leq a\leq 0.8$, $0.5\leq a\leq 0.92$, $0.5\leq a\leq 0.9$, $0.5\leq a\leq 0.8$, $0.6\leq a\leq 0.92$, or $0.6\leq a\leq 0.9$; $0\leq b\leq 0.5$, $0\leq b\leq 0.4$, $0\leq b\leq 0.3$, $0\leq b\leq 0.2$, $0.1\leq b\leq 0.5$, $0.1\leq b\leq 0.4$, $0.1\leq b\leq 0.3$, $0.1\leq b\leq 0.2$, $0.2\leq b\leq 0.5$, $0.2\leq b\leq 0.4$, or $0.2\leq b\leq 0.3$; $0\leq c\leq 0.5$, $0\leq c\leq 0.4$, $0\leq c\leq 0.3$, $0.1\leq c\leq 0.5$, $0.1\leq c\leq 0.4$, $0.1\leq c\leq 0.3$, $0.1\leq c\leq 0.2$, $0.2\leq c\leq 0.5$, $0.2\leq c\leq 0.4$, or $0.2\leq c\leq 0.3$. 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.

[0067] 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.

[0068] In some embodiments, the cathode active material is $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ (NMC333), $LiNi_{0.4}Mn_{0.4}Co_{0.2}O_2$, $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ (NMC532), $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ (NMC622), $LiNi_{0.7}Mn_{0.15}Co_{0.15}O_2$, $LiNi_{0.7}Mn_{0.1}Co_{0.2}O_2$, $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (NMC811), $LiNi_{0.92}Mn_{0.04}Co_{0.04}O_2$, $LiNi_{0.85}Mn_{0.075}Co_{0.075}O_2$, $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, $LiNi_{0.88}Co_{0.1}Al_{0.02}O_2$, $LiNiO_2$ (LNO), or combinations thereof.

[0069] In other embodiments, the cathode active material is not $LiCoO_2$, $LiNiO_2$, $LiMnO_2$, $LiMn_2O_4$, or Li_2MnO_3. In further embodiments, the cathode active material is not $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$, $LiNi_{0.4}Mn_{0.4}Co_{0.2}O_2$, $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$, $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$, $LiNi_{0.7}Mn_{0.15}Co_{0.15}O_2$, $LiNi_{0.7}Mn_{0.1}Co_{0.2}O_2$, $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$, $LiNi_{0.92}Mn_{0.04}Co_{0.04}O_2$, $LiNi_{0.85}Mn_{0.075}Co_{0.075}O_2$, $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, or $LiNi_{0.88}Co_{0.1}Al_{0.02}O_2$.

[0070] In certain embodiments, the cathode active material comprises or is a core-shell composite having a core and shell structure, wherein the core and the shell each independently comprise a lithium transition metal oxide 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 certain embodiments, each x in the above general formula is independently selected from -0.2, -0.175, -0.15, -0.125, -0.1, -0.075, -0.05, -0.025, 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175 and 0.2; each a in the above general formula is independently selected from 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375, 0.4, 0.425, 0.45, 0.475, 0.5, 0.525, 0.55, 0.575, 0.6, 0.625, 0.65, 0.675, 0.7, 0.725, 0.75, 0.775, 0.8, 0.825, 0.85, 0.875, 0.9, 0.925, 0.95 and 0.975; each b in the above general formula is independently selected from 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375, 0.4, 0.425, 0.45, 0.475, 0.5, 0.525, 0.55, 0.575, 0.6, 0.625, 0.65, 0.675, 0.7, 0.725, 0.75, 0.775, 0.8, 0.825, 0.85, 0.875, 0.9, 0.925, 0.95 and 0.975; each c in the above general formula is independently selected from 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375, 0.4, 0.425, 0.45, 0.475, 0.5, 0.525, 0.55, 0.575, 0.6, 0.625, 0.65, 0.675, 0.7, 0.725, 0.75, 0.775, 0.8, 0.825, 0.85, 0.875, 0.9, 0.925, 0.95 and 0.975. In some embodiment, each x, a, b and c in the above general formula independently has a 0.01 interval. In other embodiments, the core and the shell each independently comprise two or more lithium transition metal oxides. In some embodiments, one of the core or shell comprises only one lithium transition metal oxide, while the other comprises two or more lithium transition metal oxides. The lithium transition metal oxide or oxides in the core and the shell may be the same, or they may be different or partially different. In some embodiments, the two or more lithium transition metal oxides are uniformly distributed over the core. In certain embodiments, the two or more lithium transition metal oxides are not uniformly

distributed over the core. In some embodiments, the cathode active material is not a core-shell composite.

[0071] In some embodiments, each of the lithium transition metal oxides in the core and the shell is independently 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 certain embodiments, the core and the shell each independently comprise two or more doped lithium transition metal oxides. In some embodiments, the two or more doped lithium transition metal oxides are uniformly distributed over the core and/or the shell. In certain embodiments, the two or more doped lithium transition metal oxides are not uniformly distributed over the core and/or the shell.

[0072] In some embodiments, the cathode active material comprises or is a core-shell composite comprising a core comprising a lithium transition metal oxide and a shell comprising a transition metal oxide. In certain embodiments, the lithium transition metal oxide is 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 certain embodiments, x in the above general formula is independently selected from -0.2, -0.175, -0.15, -0.125, -0.1, -0.075, -0.05, -0.025, 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175 and 0.2; each a in the above general formula is independently selected from 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375, 0.4, 0.425, 0.45, 0.475, 0.5, 0.525, 0.55, 0.575, 0.6, 0.625, 0.65, 0.675, 0.7, 0.725, 0.75, 0.775, 0.8, 0.825, 0.85, 0.875, 0.9, 0.925, 0.95 and 0.975; each b in the above general formula is independently selected from 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375, 0.4, 0.425, 0.45, 0.475, 0.5, 0.525, 0.55, 0.575, 0.6, 0.625, 0.65, 0.675, 0.7, 0.725, 0.75, 0.775, 0.8, 0.825, 0.85, 0.875, 0.9, 0.925, 0.95 and 0.975; each c in the above general formula is independently selected from 0, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25, 0.275, 0.3, 0.325, 0.35, 0.375, 0.4, 0.425, 0.45, 0.475, 0.5, 0.525, 0.55, 0.575, 0.6, 0.625, 0.65, 0.675, 0.7, 0.725, 0.75, 0.775, 0.8, 0.825, 0.85, 0.875, 0.9, 0.925, 0.95 and 0.975. In some embodiment, each x, a, b and c in the above general formula independently has a 0.01 interval. In some embodiments, the transition metal oxide is 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. In certain embodiments, the shell comprises a lithium transition metal oxide and a transition metal oxide.

[0073] In some embodiments, the average diameter of the cathode active material particles is from about 0.1 μm to about 100 μm, from about 0.1 μm to about 50 μm, from about 0.5 μm to about 50 μm, from about 0.5 μm to about 30 μm, from about 0.5 μm to about 20 μm, from about 1 μm to about 20 μm, from about 2.5 μm to about 20 μm, from about 5 μm to about

20 μm, from about 7.5 μm to about 20 μm, from about 10 μm to about 20 μm, from about 15 μm to about 20 μm, from about 2.5 μm to about 50 μm, from about 5 μm to about 50 μm, from about 10 μm to about 50 μm, from about 15 μm to about 50 μm, from about 20 μm to about 50 μm, or from about 50 μm to about 100 μm.

[0074] In some embodiments, the average diameter of the cathode active material particles is less than 100 μm, less than 80 μm, less than 60 μm, less than 50 μm, less than 40 μm, less than 30 μm, less than 20 μm, less than 15 μm, less than 10 μm, less than 7.5 μm, less than 5 μm, less than 2.5 μm, less than 1 μm, less than 0.75 μm, or less than 0.5 μm. In some embodiments, the average diameter of the cathode active material particles is more than 0.1 μm, more than 0.25 μm, more than 0.5 μm, more than 0.75 μm, more than 1 μm, more than 2.5 μm, more than 5 μm, more than 7.5 μm, more than 10 μm, more than 15 μm, more than 20 μm, more than 30 μm, more than 40 μm, or more than 50 μm.

[0075] In some embodiments, when the cathode active material comprises or is a core-shell composite having a core and shell structure, the diameter of the core is from about 1 μm to about 15 μm, from about 3 μm to about 15 μm, from about 3 μm to about 10 μm, from about 5 μm to about 10 μm, from about 5 μm to about 45 μm, from about 5 μm to about 35 μm, from about 5 μm to about 25 μm, from about 10 μm to about 45 μm, from about 10 μm to about 40 μm, from about 10 μm to about 35 μm, from about 10 μm to about 25 μm, from about 15 μm to about 45 μm, from about 15 μm to about 30 μm, from about 15 μm to about 25 μm, from about 20 μm to about 35 μm, or from about 20 μm to about 30 μm. In certain embodiments, when the cathode active material comprises or is a core-shell composite having a core and shell structure, the thickness of the shell 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 3 μm to about 15 μm, from about 3 μm to about 10 μm, from about 5 μm to about 10 μm, from about 10 μm to about 35 μm, from about 10 μm to about 20 μm, from about 15 μm to about 30 μm, from about 15 μm to about 25 μm, or from about 20 μm to about 35 μm. In certain embodiments, when the cathode active material comprises or is a core-shell composite having a core and shell structure, 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, when the cathode active material comprises or is a core-shell composite having a core and shell structure, 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.

[0076] In some embodiments, the specific surface area of the cathode active material particles is from about 0.1 m^2/g to about 3 m^2/g, from about 0.2 m^2/g to about 3 m^2/g, from about

0.3 m^2/g to about 3 m^2/g, from about 0.4 m^2/g to about 3 m^2/g, from about 0.5 m^2/g to about 3 m^2/g, from about 0.6 m^2/g to about 3 m^2/g, from about 0.7 m^2/g to about 3 m^2/g, from about 0.8 m^2/g to about 3 m^2/g, from about 0.9 m^2/g to about 3 m^2/g, from about 1 m^2/g to about 3 m^2/g, from about 1.1 m^2/g to about 3 m^2/g, from about 1.2 m^2/g to about 3 m^2/g, from about 1.3 m^2/g to about 3 m^2/g, from about 1.4 m^2/g to about 3 m^2/g, from about 1.5 m^2/g to about 3 m^2/g, 0.1 m^2/g to about 2 m^2/g, from about 0.2 m^2/g to about 2 m^2/g, from about 0.3 m^2/g to about 2 m^2/g, from about 0.4 m^2/g to about 2 m^2/g, from about 0.5 m^2/g to about 2 m^2/g, from about 0.6 m^2/g to about 2 m^2/g, from about 0.7 m^2/g to about 2 m^2/g, from about 0.8 m^2/g to about 2 m^2/g, from about 0.9 m^2/g to about 2 m^2/g, from about 1 m^2/g to about 2 m^2/g, from about 1.1 m^2/g to about 2 m^2/g, from about 1.2 m^2/g to about 2 m^2/g, from about 1.3 m^2/g to about 2 m^2/g, from about 1.4 m^2/g to about 2 m^2/g, from about 1.5 m^2/g to about 2 m^2/g, from about 0.1 m^2/g to about 1.5 m^2/g, from about 0.2 m^2/g to about 1.5 m^2/g, from about 0.3 m^2/g to about 1.5 m^2/g, from about 0.4 m^2/g to about 1.5 m^2/g, from about 0.5 m^2/g to about 1.5 m^2/g, from about 0.6 m^2/g to about 1.5 m^2/g, from about 0.7 m^2/g to about 1.5 m^2/g, from about 0.8 m^2/g to about 1.5 m^2/g, from about 0.9 m^2/g to about 1.5 m^2/g, from about 1 m^2/g to about 1.5 m^2/g, from about 0.3 m^2/g to about 1 m^2/g, from about 0.4 m^2/g to about 1 m^2/g, from about 0.5 m^2/g to about 1 m^2/g, from about 0.6 m^2/g to about 1 m^2/g, from about 0.7 m^2/g to about 1 m^2/g, from about 0.3 m^2/g to about 0.8 m^2/g, from about 0.4 m^2/g to about 0.8 m^2/g, or from about 0.5 m^2/g to about 0.8 m^2/g.

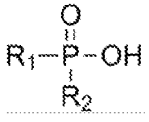
[0077] In some embodiments, the specific surface area of the cathode active material particles is less than 3 m^2/g, less than 2.75 m^2/g, less than 2.5 m^2/g, less than 2.25 m^2/g, less than 2 m^2/g, less than 1.9 m^2/g, less than 1.8 m^2/g, less than 1.7 m^2/g, less than 1.6 m^2/g, less than 1.5 m^2/g, less than 1.4 m^2/g, less than 1.3 m^2/g, less than 1.2 m^2/g, less than 1.1 m^2/g, less than 1 m^2/g, less than 0.9 m^2/g, less than 0.8 m^2/g, less than 0.7 m^2/g, less than 0.6 m^2/g, less than 0.5 m^2/g, less than 0.4 m^2/g, or less than 0.3 m^2/g. In some embodiments, the specific surface area of the cathode active material particles is more than 0.1 m^2/g, more than 0.2 m^2/g, more than 0.3 m^2/g, more than 0.4 m^2/g, more than 0.5 m^2/g, more than 0.6 m^2/g, more than 0.7 m^2/g, more than 0.8 m^2/g, more than 0.9 m^2/g, more than 1 m^2/g, more than 1.1 m^2/g, more than 1.2 m^2/g, more than 1.3 m^2/g, more than 1.4 m^2/g, more than 1.5 m^2/g, more than 1.6 m^2/g, more than 1.7 m^2/g, more than 1.8 m^2/g, more than 1.9 m^2/g, more than 2 m^2/g, more than 2.25 m^2/g, or more than 2.5 m^2/g.

[0078] The present invention is particularly applicable to cathode active materials that contain nickel, since it was found that cathode active materials that contain nickel are especially susceptible to degradation due to reaction with water.

[0079] In order to prevent degradation of cathode active materials in water and enable

such cathode active materials to be used in a water-based electrode slurry, it is proposed that the cathode active material is first pre-treated by coating the cathode active material with a coating layer, thereby forming a coated cathode active material. Each particle of the coated cathode active material comprises a particle of the cathode active material and a coating layer surrounding the particle of the cathode active material particle, wherein the coating layer is derived from a phosphorus-containing compound. In some embodiments, the particles of the coated cathode active material are in the form of primary particles, secondary particles, tertiary particles or a combination thereof.

[0080] In some embodiments, the phosphorus-containing compound has a chemical structure represented by general formula (1):



(1)

[0081] In some embodiments, R_1 is alkyl, alkenyl, alkynyl, enynyl, cycloalkyl, or alkylcarbonylalkyl. In some embodiments, R_2 is alkyl, alkenyl, alkynyl, enynyl, cycloalkyl, alkoxy, alkylcarbonylalkyl, or hydroxyl. In certain embodiments, each of the alkyl, alkenyl, alkynyl, enynyl, cycloalkyl, alkoxy, and alkylcarbonylalkyl is optionally substituted with one or more substituents. In some embodiments, the one or more substituents are independently selected from F, Cl, Br, I, cyano, hydroxyl, N_3, NO_2, NH_2, ester, amide, aldehyde, acyl, alkyl, alkoxy, alkylthio or alkylamino.

[0082] In some embodiments, R_1 is an alkyl group. In certain embodiments, R_1 is C_1-C_{40} alkyl, C_1-C_{35} alkyl, C_1-C_{30} alkyl, C_1-C_{25} alkyl, C_1-C_{20} alkyl, C_1-C_{15} alkyl, C_1-C_{10} alkyl, C_5-C_{40} alkyl, C_5-C_{30} alkyl, C_5-C_{20} alkyl, C_5-C_{10} alkyl, C_{10}-C_{40} alkyl, C_{10}-C_{30} alkyl, C_{10}-C_{20} alkyl, or C_{15}-C_{20} alkyl. In some embodiments, R_1 is a straight-chain group.

[0083] In some embodiments, R_1 is methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, nonadecyl, or icosyl.

[0084] In some embodiments, R_2 is a hydroxyl, alkyl, or alkoxy group. In certain embodiments, R_2 is C_1-C_{40} alkyl or alkoxy, C_1-C_{35} alkyl or alkoxy, C_1-C_{30} alkyl or alkoxy, C_1-C_{25} alkyl or alkoxy, C_1-C_{20} alkyl or alkoxy, C_1-C_{15} alkyl or alkoxy, C_1-C_{10} alkyl or alkoxy, C_5-C_{40} alkyl or alkoxy, C_5-C_{30} alkyl or alkoxy, C_5-C_{20} alkyl or alkoxy, C_5-C_{10} alkyl or alkoxy, C_{10}-C_{40} alkyl or alkoxy, C_{10}-C_{30} alkyl or alkoxy, C_{10}-C_{20} alkyl or alkoxy, or C_{15}-C_{20} alkyl or alkoxy.

[0085] In some embodiments, R_2 is methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, nonadecyl, or icosyl. In some embodiments, R_2 is methoxy, ethoxy, propoxy, butoxy,

pentoxy, hexoxy, heptoxy, octoxy, nonoxy, decoxy, undecoxy, dodecoxy, tridecoxy, tetradecoxy, pentadecoxy, hexadecoxy, heptadecoxy, octadecoxy, nonadecoxy, or icosoxy.

[0086] In some embodiments, when R_2 is an alkyl group, R_1 and R_2 are the same. In other embodiments, when R_2 is an alkyl group, R_1 and R_2 are different. In some embodiments, a phosphorus-containing compound having general formula (1) is used to coat the cathode active material. In other embodiments, two or more phosphorus-containing compounds are used in conjunction to coat the cathode active material, wherein each of the phosphorus-containing compounds has a chemical structure having general formula (1).

[0087] In some embodiments, the phosphorus-containing compound(s) is/are dissolved in a polar organic solvent to form a coating mixture. Using a polar solvent is advantageous since the phosphorus-containing compounds represented by general formula (1) are polar and would therefore dissolve more easily in a polar solvent to form a well-dispersed coating mixture. In some embodiments, the polar organic solvent is a green solvent, i.e., a solvent that has a reduced environmental impact. The use of a green solvent specifically may be preferable since it can help minimize the potential environmental impact of using said solvent in the coating reaction.

[0088] Some non-limiting examples of suitable green polar organic solvents include 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), methyl isobutyl ketone (MIBK) 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.

[0089] Some other non-limiting examples of suitable polar organic solvents include tetrahydrofuran (THF), 2-methyl tetrahydrofuran, methyl tert-butyl ether, cyclopentyl methyl ether, acetonitrile, dimethyl sulfoxide, sulfolane, propylene carbonate, ethylene carbonate, dimethyl carbonate, diethyl carbonate, ethyl methyl carbonate, furfuryl alcohol, formic acid, acetic acid, γ-valerolactone (GVL), methyl lactate, ethyl lactate, dihydrolevoglucosenone (Cyrene™), dibutoxymethane, N,N'-dimethylpropyleneurea (DMPU), dimethyl isosorbide (DMI) and combinations thereof.

[0090] In some embodiments, the molar proportion of phosphorus-containing compound with respect to the sum of moles of the phosphorus-containing compound and the cathode active material is from about 0.01% to about 1%, from about 0.01% to about 0.9%, from about 0.01% to about 0.8%, from about 0.01% to about 0.7%, from about 0.01% to about 0.6%, from about 0.01% to about 0.5%, from about 0.01% to about 0.4%, from about 0.01% to about 0.3%, from

about 0.01% to about 0.2%, from about 0.01% to about 0.1%, from about 0.05% to about 1%, from about 0.05% to about 0.9%, from about 0.05% to about 0.8%, from about 0.05% to about 0.7%, from about 0.05% to about 0.6%, from about 0.05% to about 0.5%, from about 0.05% to about 0.4%, from about 0.05% to about 0.3%, from about 0.05% to about 0.2%, from about 0.05% to about 0.1%, from about 0.1% to about 1%, from about 0.1% to about 0.9%, from about 0.1% to about 0.8%, from about 0.1% to about 0.7%, from about 0.1% to about 0.6%, from about 0.1% to about 0.5%, from about 0.1% to about 0.4%, or from about 0.1% to about 0.3%.

[0091] In some embodiments, the molar proportion of phosphorus-containing compound with respect to the sum of moles of the phosphorus-containing compound and the cathode active material is less than 1%, less than 0.9%, less than 0.8%, less than 0.7%, less than 0.6%, less than 0.5%, less than 0.4%, less than 0.3%, less than 0.2%, less than 0.1%, less than 0.075%, or less than 0.05%. In some embodiments, the molar proportion of phosphorus-containing compound with respect to the sum of moles of the phosphorus-containing compound and the cathode active material is more than 0.01%, more than 0.025%, more than 0.05%, more than 0.075%, more than 0.1%, more than 0.2%, more than 0.3%, more than 0.4%, more than 0.5%, more than 0.6%, more than 0.7%, or more than 0.8%.

[0092] In some embodiments, the proportion of the phosphorus-containing compound in the coating mixture is from about 0.01% to about 1%, from about 0.01% to about 0.9%, from about 0.01% to about 0.8%, from about 0.01% to about 0.7%, from about 0.01% to about 0.6%, from about 0.01% to about 0.5%, from about 0.01% to about 0.4%, from about 0.01% to about 0.3%, from about 0.01% to about 0.2%, from about 0.01% to about 0.1%, from about 0.05% to about 1%, from about 0.05% to about 0.9%, from about 0.05% to about 0.8%, from about 0.05% to about 0.7%, from about 0.05% to about 0.6%, from about 0.05% to about 0.5%, from about 0.05% to about 0.4%, from about 0.05% to about 0.3%, from about 0.05% to about 0.2%, from about 0.05% to about 0.1%, from about 0.1% to about 1%, from about 0.1% to about 0.9%, from about 0.1% to about 0.8%, from about 0.1% to about 0.7%, from about 0.1% to about 0.6%, from about 0.1% to about 0.5%, from about 0.1% to about 0.4%, or from about 0.1% to about 0.3% by weight, based on the total weight of the coating mixture.

[0093] In some embodiments, the proportion of the phosphorus-containing compound in the coating mixture is less than 1%, less than 0.9%, less than 0.8%, less than 0.7%, less than 0.6%, less than 0.5%, less than 0.4%, less than 0.3%, less than 0.2%, less than 0.1%, less than 0.08%, or less than 0.05% by weight, based on the total weight of the coating mixture. In some embodiments, the proportion of the phosphorus-containing compound in the coating mixture is more than 0.01%, more than 0.03%, more than 0.05%, more than 0.08%, more than 0.1%, more than 0.2%, more than 0.3%, more than 0.4%, more than 0.5%, more than 0.6%, more than 0.7%, or more than 0.8% by weight, based on the total weight of the coating mixture.

[0094] In some embodiments, the cathode active material is added into the coating mixture in order to form a coating layer surrounding the particles of the cathode active material, wherein the coating layer is derived from phosphorus-containing compound. The addition of the cathode active material to the coating mixture forms a reaction mixture. In some embodiments, after the reaction mixture is formed, it is continuously stirred using a homogenizer to allow for the coating reaction to occur. There is no particular limitation on the homogenizer used, except that the homogenizer should be able to disperse the cathode active material in the reaction mixture to ensure that the surfaces of all the cathode active material particles can be in contact with the phosphorus-containing compound to enable the formation of the coating layer around each cathode active material particle. Some non-limiting examples of homogenizers include stirring mixers, planetary stirring mixers, blenders and ultrasonicators.

[0095] There is no particular limitation to the stirring speed, except that it should be sufficient to enable good dispersion of the cathode active material in the reaction mixture to ensure the formation of the coating layer around each cathode active material particle. Similarly, there is no particular limitation to the time period of the coating reaction, except that such a time period should be sufficient to ensure all the cathode active material particles are coated with the phosphorus-containing compound.

[0096] In addition, there is no particular limitation to the temperature at which the coating reaction occurs, except that it should not be too high as to cause boiling of the solvent in the coating mixture, but at the same time sufficiently high to ensure the coating reaction can occur within a reasonable timeframe. In some embodiments, the coating reaction occurs at a temperature of from about 20 °C to about 95 °C, from about 25 °C to about 95 °C, from about 30 °C to about 95 °C, from about 35 °C to about 95 °C, from about 40 °C to about 95 °C, from about 45 °C to about 95 °C, from about 50 °C to about 95 °C, from about 55 °C to about 95 °C, from about 60 °C to about 95 °C, from about 65 °C to about 95 °C, from about 70 °C to about 95 °C, from about 75 °C to about 95 °C, from about 30 °C to about 75 °C, from about 35 °C to about 75 °C, from about 40 °C to about 75 °C, from about 45 °C to about 75 °C, from about 50 °C to about 75 °C, from about 55 °C to about 75 °C, from about 60 °C to about 75 °C, from about 30 °C to about 60 °C, from about 35 °C to about 60 °C, from about 40 °C to about 60 °C, from about 45 °C to about 60 °C, or from about 50 °C to about 60 °C.

[0097] In some embodiments, the coating reaction occurs at a temperature of less than 95 °C, less than 90 °C, less than 85 °C, less than 80 °C, less than 75 °C, less than 70 °C, less than 65 °C, less than 60 °C, less than 55 °C, less than 50 °C, less than 45 °C, or less than 40 °C. In some embodiments, the coating reaction occurs at a temperature of more than 20 °C, more than 25 °C, more than 30 °C, more than 35 °C, more than 40 °C, more than 45 °C, more than 50 °C, more than 55 °C, more than 60 °C, more than 65 °C, more than 70 °C, or more than 75 °C.

[0098] As described above, the chemical structure of a compound suitable for use as a phosphorus-containing compound can comprise one or two hydroxyl groups. Regardless of whether the compound has one or two hydroxyl groups, the proposed mechanism for the attachment of a molecule of the phosphorus-containing compound to the surface of a cathode active material particle would be similar. The bond between the oxygen and hydrogen atoms in a hydroxyl group of a phosphorus-containing compound molecule is highly polar in nature. As a result, the δ^- charged oxygen atom from the hydroxyl group (and the double-bonded oxygen atom) of the phosphorus-containing compound would be attracted to δ^+ charged species on the surface of the cathode active material particle, while the δ^+ charged hydrogen atom from the hydroxyl group would be attracted to the δ^- charged species on the surface of the same cathode active material particle. The combined effect of such attractive forces is a strong attachment of the phosphorus-containing compound molecule to the surface of the cathode active material particle. The attachment of phosphorus-containing compound molecules to the surface of the cathode active material particle is spontaneous in nature, and when more and more phosphorus-containing compound molecules are attached to the surface of the cathode active material particle, a coating layer of the phosphorus-containing compound is formed. The spontaneous phenomenon of the formation of the coating layer is referred to herein as self-assembly, and when the surface of a cathode active material particle is completely covered with a layer of phosphorus-containing compound molecules, the result is referred to herein as a self-assembled monolayer (SAM).

[0099] Meanwhile, the hydrophobic alkyl groups of the phosphorus-containing compound molecules, which are now attached to the cathode active material particle, would be oriented outwards relative to the surface of the cathode active material particle. The steric hindrance of the alkyl groups ensures that water cannot get close to the surface of the cathode active material particle and cause reaction and subsequent degradation of the cathode active material in the particle, while still allowing for transport of lithium ions. As a result, reaction of cathode active material with water can be suppressed, and the coated cathode active material can therefore be successfully used in a water-based electrode slurry without degradation of the cathode active material in water. As the coating layer of the present invention is self-assembling, apart from the coating mixture comprising the phosphorus-containing compound and the polar organic solvent, no additional chemicals or substances are required in order to coat the cathode active material particles with the coating layer. In certain embodiments, the process of coating the cathode active material particles is not a sol-gel process.

[00100] Figure 1 shows the amount of degradation of the coated cathode active materials of Examples 3 and 4 in water, normalized with respect to the amount of degradation of the uncoated cathode active material of Comparative Example 1 in water. As shown, compared to the uncoated cathode active material of Comparative Example 1, the amount of degradation of

the coated cathode active material of Example 3 was around 30% less, and the amount of degradation of the coated cathode active material of Example 4 was around 25% less. This shows that the coating of cathode active material using the phosphorus-containing compound is successful in reducing the degradation of cathode active materials due to reaction with water.

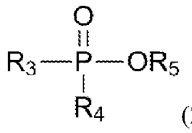
[00101] In some embodiments, through the presence of the coating surrounding each cathode active material particle, the degradation of cathode active material in water is inhibited by a percentage from about 1% to about 40%, from about 1% to about 38%, from about 1% to about 35%, from about 1% to about 32%, from about 1% to about 30%, from about 1% to about 29%, from about 1% to about 28%, from about 1% to about 27%, from about 1% to about 26%, from about 1% to about 25%, from about 1% to about 24%, from about 1% to about 23%, from about 1% to about 22%, from about 1% to about 21%, from about 1% to about 20%, from about 1% to about 19%, from about 1% to about 18%, from about 1% to about 17%, from about 1% to about 16%, from about 1% to about 15%, from about 5% to about 30%, from about 5% to about 29%, from about 5% to about 28%, from about 5% to about 27%, from about 5% to about 26%, from about 5% to about 25%, from about 5% to about 24%, from about 5% to about 23%, from about 5% to about 22%, from about 5% to about 21%, from about 5% to about 20%, from about 10% to about 30%, from about 10% to about 29%, from about 10% to about 28%, from about 10% to about 27%, from about 10% to about 26%, from about 10% to about 25%, from about 10% to about 24%, from about 10% to about 23%, from about 10% to about 22%, from about 10% to about 21%, from about 10% to about 20%, from about 15% to about 30%, from about 15% to about 29%, from about 15% to about 28%, from about 15% to about 27%, from about 15% to about 26%, from about 15% to about 25%, from about 20% to about 30%, from about 20% to about 29%, from about 20% to about 28%, from about 20% to about 27%, from about 20% to about 26%, or from about 20% to about 25%.

[00102] In some embodiments, through the presence of the coating surrounding each cathode active material particle, the degradation of cathode active material in water is inhibited by a percentage of less than 40%, less than 38%, less than 35%, less than 32%, 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%, or less than 10%. In some embodiments, through the presence of the coating surrounding each cathode active material particle, the degradation of cathode active material in water is inhibited by a percentage of more than 1%, more than 3%, more than 5%, more than 7%, 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%, more than 28%, or more than 29%.

[00103] In a phosphorus-containing compound molecule wherein the hydrocarbon chains in the molecule have a branched or ring configuration, steric hindrance from these large hydrocarbon chains means that the area occupied by each phosphorus-containing compound molecule would be much larger. As a result, fewer molecules would be able to attach to the surface of a cathode active material particle. Surface coverage of a cathode active material particle by such a phosphorus-containing compound would be poorer compared to that by a phosphorus-containing compound with only linear hydrocarbon chains, such as the phosphorus-containing compounds of the present invention.

[00104] It is desirable for the phosphorus-containing compound used in the present invention to contain both hydroxyl groups and linear hydrocarbon chains, since, for reasons elaborated below, the presence of both features is necessary to ensure good surface coverage of said phosphorus-containing compound around a cathode active material particle.

[00105] Imagine a phosphorus-containing compound with a chemical structure similar to that of the phosphorus-containing compounds of the present invention, except that no hydroxyl groups are present. Such a chemical structure can be represented by general formula (2) below,

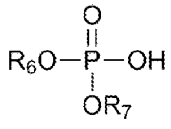


(2)

wherein R_3 and R_5 would each independently represent an alkyl group, while R_4 would represent an alkyl or alkoxy group. Due to the presence of three mutually sterically repulsive hydrocarbon chains, the area occupied by each phosphorus-containing compound molecule would be much larger. As a result, fewer molecules would be able to attach to the surface of a cathode active material particle, leading to poor surface coverage. Furthermore, the polarity of an O-hydrocarbon bond is smaller compared to the polarity of the O-H bond in a hydroxyl group. As a result, the attractive forces of a phosphorus-containing compound molecule represented by general formula (2) to the surface of a cathode active material particle would be weaker compared to those of a phosphorus-containing compound molecule of the present invention. If a phosphorus-containing compound represented by general formula (2) were used to coat a cathode active material instead of a phosphorus-containing compound of the present invention, formation of a monolayer would be much less efficient. There would also be a risk of the phosphorus-containing compound detaching in the water-based electrode slurry, leading to degradation of the cathode active material due to reaction with water. Therefore, it is desirable for the phosphorus-containing compound of the present invention to have hydroxyl groups.

[00106] On the other hand, imagine a phosphorus-containing compound with a chemical structure similar to that of the phosphorus-containing compound of the present invention, except that all the hydrocarbon chains are not directly connected to the phosphorus atom, but rather to

oxygens atoms that are bonded with the phosphorus atom. Such a compound would have a chemical structure represented by general formula (3) below,



(3)

wherein R_6 would represent an alkyl group, while R_7 would represent either a hydrogen atom or an alkyl group.

[00107] Figure 2 shows a Nyquist plot of the real and imaginary components of the impedance of a coin cell prepared according to Example 4 at different current frequencies, while Figure 3 shows a Nyquist plot of the real and imaginary components of the impedance of a coin cell prepared according to Comparative Example 14 at different current frequencies. The cathode active material in the coin cells of both examples were coated with a coating layer derived from a phosphorus-containing compound. The phosphorus-containing compound used in Example 4 is one disclosed by the present invention and has a chemical structure represented by general formula (1), while the phosphorus-containing compound used in Comparative Example 14 has a chemical structure represented by general formula (3). As shown, the diameter of the semicircle of the Nyquist plot in Figure 3 is greater than the diameter of the semicircle of the Nyquist plot in Figure 2, showing that the impedance of the coin cell prepared according to Comparative Example 14 is greater than that of the coin cell prepared according to Example 4. This suggests that batteries comprising a cathode active material coated with a coating layer derived from a phosphorus-containing compound represented by general formula (3) would have a higher impedance compared to batteries comprising a cathode active material coated with a coating layer derived from a phosphorus-containing compound represented by general formula (1). Accordingly, it is desirable for the phosphorus-containing compound of the present invention to have at least one hydrocarbon chain that is directly bonded to the phosphorus atom.

[00108] For the reasons explained above, it is preferable for the phosphorus-containing compound of the present invention to have hydroxyl groups as well as hydrocarbon chains that are directly bonded with the phosphorus atom. Considering the structure of the phosphorus containing compound, it is therefore desirable that the phosphorus-containing compound of the present invention contain one to two hydroxyl groups and one to two hydrocarbon chains that are directly bonded with the phosphorus atom (i.e., the chemical structure represented by general formula (1)).

[00109] In some embodiments, the thickness of the coating layer of the cathode active material particles is from about 0.1 nm to about 10 nm, from about 0.15 nm to about 10 nm, from about 0.2 nm to about 10 nm, from about 0.25 nm to about 10 nm, from about 0.3 nm to about 10 nm, from about 0.4 nm to about 10 nm, from about 0.5 nm to about 10 nm, from about

0.6 nm to about 10 nm, from about 0.8 nm to about 10 nm, from about 1 nm to about 10 nm, from about 1.2 nm to about 10 nm, from about 1.4 nm to about 10 nm, from about 1.6 nm to about 10 nm, from about 1.8 nm to about 10 nm, from about 2 nm to about 10 nm, from about 3 nm to about 10 nm, from about 4 nm to about 10 nm, from about 5 nm to about 10 nm, from about 0.25 nm to about 5 nm, from about 0.3 nm to about 5 nm, from about 0.4 nm to about 5 nm, from about 0.5 nm to about 5 nm, from about 0.6 nm to about 5 nm, from about 0.8 nm to about 5 nm, from about 1 nm to about 5 nm, from about 1.2 nm to about 5 nm, from about 1.4 nm to about 5 nm, from about 1.6 nm to about 5 nm, from about 1.8 nm to about 5 nm, from about 2 nm to about 5 nm, from about 0.5 nm to about 3 nm, from about 0.6 nm to about 3 nm, from about 0.8 nm to about 3 nm, from about 1 nm to about 3 nm, from about 1.2 nm to about 3 nm, from about 1.4 nm to about 3 nm, from about 1.6 nm to about 3 nm, from about 1.8 nm to about 3 nm, or from about 2 nm to about 3 nm.

[00110] In some embodiments, the thickness of the coating layer of the cathode active material particles is less than 10 nm, less than 8 nm, less than 6 nm, less than 5 nm, less than 4 nm, less than 3 nm, less than 2 nm, less than 1.8 nm, less than 1.6 nm, less than 1.4 nm, less than 1.2 nm, less than 1 nm, less than 0.8 nm, less than 0.6 nm, or less than 0.5 nm. In some embodiments, the thickness of the coating layer of the cathode active material particles is more than 0.1 nm, more than 0.15 nm, more than 0.2 nm, more than 0.25 nm, more than 0.3 nm, more than 0.4 nm, more than 0.5 nm, more than 0.6 nm, more than 0.8 nm, more than 1 nm, more than 1.2 nm, more than 1.4 nm, more than 1.6 nm, more than 1.8 nm, or more than 2 nm.

[00111] In some embodiments, the amount of phosphorus-containing compound coated per unit surface area of cathode active material is from about 0.1 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 0.2 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 0.25 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 0.5 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 0.75 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 1 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 1.5 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 2 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 2.5 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 4 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 6 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 8 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 10 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 12.5 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 15 $\mu mol/m^2$ to about 30 $\mu mol/m^2$, from about 0.1 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 0.2 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 0.25 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 0.5 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 0.75 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 1 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 1.5 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 2 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 2.5 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 4 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 6 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 8 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 10 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 12.5 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 15 $\mu mol/m^2$ to about 25 $\mu mol/m^2$, from about 0.1 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 0.2

$\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 0.25 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 0.5 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 0.75 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 1 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 1.5 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 2 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 2.5 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 4 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 6 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 8 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 10 $\mu mol/m^2$ to about 22.5 $\mu mol/m^2$, from about 0.5 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 0.75 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 1 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 1.5 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 2 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 2.5 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 4 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 6 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 8 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 10 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 12.5 $\mu mol/m^2$ to about 20 $\mu mol/m^2$, from about 1 $\mu mol/m^2$ to about 15 $\mu mol/m^2$, from about 1.5 $\mu mol/m^2$ to about 15 $\mu mol/m^2$, from about 2 $\mu mol/m^2$ to about 15 $\mu mol/m^2$, from about 2.5 $\mu mol/m^2$ to about 15 $\mu mol/m^2$, from about 4 $\mu mol/m^2$ to about 15 $\mu mol/m^2$, from about 6 $\mu mol/m^2$ to about 15 $\mu mol/m^2$, from about 8 $\mu mol/m^2$ to about 15 $\mu mol/m^2$, from about 10 $\mu mol/m^2$ to about 15 $\mu mol/m^2$, or from about 12.5 $\mu mol/m^2$ to about 15 $\mu mol/m^2$.

[00112] In some embodiments, the amount of phosphorus-containing compound coated per unit surface area of cathode active material is less than 30 $\mu mol/m^2$, less than 27.5 $\mu mol/m^2$, less than 25 $\mu mol/m^2$, less than 22.5 $\mu mol/m^2$, less than 20 $\mu mol/m^2$, less than 17.5 $\mu mol/m^2$, less than 15 $\mu mol/m^2$, less than 12.5 $\mu mol/m^2$, less than 10 $\mu mol/m^2$, less than 8 $\mu mol/m^2$, less than 6 $\mu mol/m^2$, less than 4 $\mu mol/m^2$, less than 2.5 $\mu mol/m^2$, less than 2 $\mu mol/m^2$, less than 1.5 $\mu mol/m^2$, less than 1 $\mu mol/m^2$, less than 0.75 $\mu mol/m^2$, less than 0.5 $\mu mol/m^2$, less than 0.25 $\mu mol/m^2$, or less than 0.2 $\mu mol/m^2$. In some embodiments, the amount of phosphorus-containing compound coated per unit surface area of cathode active material is more than 0.1 $\mu mol/m^2$, more than 0.2 $\mu mol/m^2$, more than 0.25 $\mu mol/m^2$, more than 0.5 $\mu mol/m^2$, more than 0.75 $\mu mol/m^2$, more than 1 $\mu mol/m^2$, more than 1.5 $\mu mol/m^2$, more than 2 $\mu mol/m^2$, more than 2.5 $\mu mol/m^2$, more than 4 $\mu mol/m^2$, more than 6 $\mu mol/m^2$, more than 8 $\mu mol/m^2$, more than 10 $\mu mol/m^2$, more than 12.5 $\mu mol/m^2$, more than 15 $\mu mol/m^2$, more than 17.5 $\mu mol/m^2$, more than 20 $\mu mol/m^2$, more than 22.5 $\mu mol/m^2$, more than 25 $\mu mol/m^2$, or more than 27.5 $\mu mol/m^2$.

[00113] Following completion of the coating reaction, the now-coated cathode active material is dispersed in the remaining reaction mixture, forming a product mixture. In some embodiments, the product mixture is filtered to separate the majority of the solvent from the coated cathode active material. In other embodiments, the product mixture is not filtered. In some embodiments, the product mixture is dried to remove the solvent using a dryer in order to obtain a coated cathode active material, and the drying can occur before, during or after the filtration, or without filtration. In other embodiments, drying is not conducted. Filtering and/or drying to separate the solvent from the product mixture may be beneficial in terms of logistics

by reducing the volume and mass required to transport or store a given amount of coated cathode active material. Apart from filtering and/or drying, no additional processing steps may be required following the coating reaction in order to obtain the coated cathode active material. In some embodiments, no calcining or sintering is required to obtain the coated cathode active material. In further embodiments, neither drying nor filtering is conducted, and the product mixture is directly transported, stored, or used in the production of an electrode slurry.

[00114] There is no particular limitation as to the dryer used to dry the product mixture, except that the dryer should be capable of removing the solvent from the product 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 a combination thereof. There are no particular limitations as to the conditions used to dry the product mixture, such as the temperature and pressure, except that such conditions should be sufficient to remove the solvent from the product mixture but do not cause the breakdown of the coating layer surrounding the cathode active material particles and/or the decomposition of the cathode active material. There is also no particular limitation as to the time used to dry the product mixture, except that the time period should be sufficiently long as to ensure the removal of the solvent from the product mixture.

[00115] Because of the coating layer derived from the phosphorus-containing compound, reaction of the coated cathode active material particles with water is suppressed. As a result, the coated cathode active material can be used in a water-based electrode slurry. Such a water-based slurry can then be used to produce cathodes for batteries and can also be stored and/or transported without risk of degradation of the cathode active material due to reaction with water. Accordingly, it is another aim of the invention to disclose a water-based electrode slurry that comprises the coated cathode active material disclosed above.

[00116] In some embodiments, the water-based electrode slurry comprises various electrode components suspended and/or dissolved in a solvent. In certain embodiments, the solvent is an aqueous solvent. In some embodiments, the electrode components comprise an electrode active material and a binder material. In some embodiments, the electrode active material is the coated cathode active material of the present invention. When an electrode slurry comprises a cathode active material, the slurry can also be known as a cathode slurry. Accordingly, a water-based electrode slurry comprising the coated cathode active material of the present invention can also be known as a water-based cathode slurry. In some embodiments, the electrode components further comprise a conductive agent. When the water-based cathode slurry comprises a conductive agent, the slurry would then comprise a coated cathode active material, a binder material, and a conductive agent as electrode components, all suspended and/or dissolved in a solvent.

[00117] There is no particular limitation to the binder material used, although the binder

material should have desirable properties as a binder and can be dispersed well in the aqueous solvent of the water-based electrode slurry to ensure an even, smooth coating. In some embodiments, the binder material comprises a polymer; such a polymer can be termed a binder polymer. In some embodiments, the binder polymer is a copolymer; such a copolymer can then be termed a binder copolymer. In other embodiments, the binder polymer is a homopolymer.

[00118] In certain embodiments, the binder polymer is styrene-butadiene rubber (SBR), carboxymethyl cellulose (CMC), polyacrylic acid (PAA), polyacrylonitrile (PAN), polyacrylamide (PAM), acrylic acid-acrylonitrile-acrylamide copolymer, latex, a salt of alginic acid, polyvinylidene fluoride (PVDF), poly(vinylidene fluoride)-hexafluoropropene (PVDF-HFP), polytetrafluoroethylene (PTFE), polystyrene, poly(vinyl alcohol) (PVA), poly(vinyl acetate), polyisoprene, polyaniline, polyethylene, polyimide, polyurethane, polyvinyl butyral, polyvinyl pyrrolidone (PVP), gelatin, chitosan, starch, agar-agar, xanthan gum, gum arabic, gellan gum, guar gum, gum karaya, tara gum, gum tragacanth, casein, amylose, pectin, PEDOT:PSS, carrageenans, or combinations thereof. In certain embodiments, the salt of alginic acid comprises a cation selected from the group consisting of Na, Li, K, Ca, NH_4, Mg, Al, and combinations thereof. In certain embodiments, the binder polymer is not styrene-butadiene rubber, carboxymethyl cellulose, polyacrylic acid, polyacrylonitrile, polyacrylamide, acrylic acid-acrylonitrile-acrylamide copolymer, latex, a salt of alginic acid, polyvinylidene fluoride, poly(vinylidene fluoride)-hexafluoropropene, polytetrafluoroethylene, polystyrene, poly(vinyl alcohol), poly(vinyl acetate), polyisoprene, polyaniline, polyethylene, polyimide, polyurethane, polyvinyl butyral, polyvinyl pyrrolidone, gelatin, chitosan, starch, agar-agar, xanthan gum, gum arabic, gellan gum, guar gum, gum karaya, tara gum, gum tragacanth, casein, amylose, pectin, PEDOT:PSS, or carrageenans. In certain embodiments, the binder polymer is not a fluorine-containing polymer such as PVDF, PVDF-HFP or PTFE.

[00119] In some embodiments, the binder polymer comprises one or more functional groups containing a halogen, O, N, S or a combination thereof. Some non-limiting examples of suitable functional groups include alkoxy, aryloxy, nitro, thiol, thioether, imine, cyano, amide, amino (primary, secondary or tertiary), carboxyl, epoxy, ketone, aldehyde, ester, hydroxyl, halo (fluoro, chloro, bromo, or iodo) and combinations thereof. In some embodiments, the functional group is or comprises carboxylic acid (*i.e.*, -COOH), carboxylic acid salt, sulfonic acid, sulfonic acid salt, phosphonic acid, phosphonic acid salt, phosphoric acid, phosphoric acid salt, nitrile, -CO_2CH_3, -$CONH_2$, -OCH_2CONH_2, or -NH_2.

[00120] In certain embodiments, the binder polymer is a copolymer with a composition as described below. The presence of hydrophilic functional groups in the binder copolymer enables the binder copolymer to be well dispersed within aqueous solvents such as water, thus providing good processibility when such copolymers are used in a water-based electrode slurry. Meanwhile, the presence of hydrophobic functional groups within the same copolymer allows

the presence of hydrophobic interactions of the copolymer with the coated cathode active material and the conductive agent, ensuring that all the various electrode components can be bound together. Combining both hydrophilic and hydrophilic effects, this means that all the various electrode components would be well bound together while still remaining dispersed in the solvent of the water-based electrode slurry. Electrode layers produced using such a slurry would then be smooth and homogeneous, and batteries comprising such electrodes would then have superb capacity and electrochemical performance.

[00121] In some embodiments, the binder copolymer comprises a structural unit (a) that is derived from an acid group-containing monomer, wherein the acid group is selected from the group consisting of carboxylic acid, sulfonic acid, phosphonic acid, phosphoric 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 an alkali metal forming the alkali metal cation include lithium, sodium, and potassium. In some embodiments, the salt of the acid comprises an ammonium cation.

[00122] 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 a combination 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, methacrolein, methacryloyl chloride, methacryloyl fluoride, methacryloyl bromide, or combinations thereof.

[00123] 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, allyl hydrogensulfate, vinyl hydrogensulfate, or a combination thereof.

[00124] 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 a combination thereof.

[00125] 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, vinyl hydrogen phosphate, allyl hydrogen phosphate, or a combination thereof, wherein (meth)acryloyloxy- means acryloyloxy- or methacryloyloxy-.

[00126] In some embodiments, the proportion of structural unit (a) within the binder copolymer is from about 15% to about 95%, from about 15% to about 90%, from about 15% to about 85%, from about 15% to about 80%, from about 15% to about 75%, from about 15% to about 70%, from about 15% to about 65%, from about 15% to about 60%, from about 15% to about 55%, from about 15% to about 50%, from about 20% to about 95%, from about 20% to about 90%, from about 20% to about 85%, from about 20% to about 80%, from about 20% to about 75%, from about 20% to about 70%, from about 20% to about 65%, from about 20% to about 60%, from about 20% to about 55%, from about 20% to about 50%, from about 25% to about 95%, from about 25% to about 90%, from about 25% to about 85%, from about 25% to about 80%, from about 25% to about 75%, from about 25% to about 70%, from about 25% to about 65%, from about 25% to about 60%, from about 25% to about 55%, from about 25% to about 50%, from about 30% to about 95%, from about 30% to about 90%, from about 30% to about 85%, from about 30% to about 80%, from about 30% to about 75%, from about 30% to about 70%, from about 30% to about 65%, from about 30% to about 60%, from about 35% to about 95%, from about 35% to about 90%, from about 35% to about 85%, from about 35% to about 80%, from about 35% to about 75%, from about 35% to about 70%, from about 35% to about 65%, from about 35% to about 60%, from about 40% to about 95%, from about 40% to

about 90%, from about 40% to about 85%, from about 40% to about 80%, from about 40% to about 75%, from about 40% to about 70%, from about 40% to about 65%, from about 40% to about 60%, from about 45% to about 95%, from about 45% to about 90%, from about 45% to about 85%, from about 45% to about 80%, from about 45% to about 75%, from about 45% to about 70%, from about 50% to about 95%, from about 50% to about 90%, from about 50% to about 85%, from about 50% to about 80%, from about 50% to about 75%, from about 50% to about 70%, from about 55% to about 95%, from about 55% to about 90%, from about 55% to about 85%, from about 55% to about 80%, from about 55% to about 75%, from about 60% to about 95%, from about 60% to about 90%, or from about 60% to about 85% by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00127] In some embodiments, the proportion of structural unit (a) within the binder copolymer is about 15%, about 20%, about 25%, about 30%, about 35%, about 40%, about 45%, about 50%, about 55%, about 60%, about 65%, about 70%, about 75%, about 80%, about 85%, about 90%, or about 95% by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00128] In some embodiments, the proportion of structural unit (a) within the binder copolymer is less than 95%, less than 90%, less than 85%, less than 80%, less than 75%, less than 70%, less than 65%, less than 60%, less than 55%, less than 50%, less than 45%, less than 40%, less than 35%, less than 30%, or less than 25% by mole, based on the total number of moles of monomeric units in the binder copolymer. In some embodiments, the proportion of structural unit (a) within the binder copolymer is more than 15%, more than 20%, more than 25%, more than 30%, more than 35%, more than 40%, more than 45%, more than 50%, more than 55%, more than 60%, more than 65%, more than 70%, more than 75%, more than 80%, or more than 85% by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00129] In some embodiments, the binder copolymer further comprises a structural unit (b) that is derived from a monomer selected from the group consisting of an amide group-containing monomer, a hydroxyl group-containing monomer and combinations thereof.

[00130] 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-dimethyl 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 a combination thereof.

[00131] In some embodiments, the hydroxyl group-containing monomer is an acrylate or methacrylate containing a C_1-C_{20} alkyl or C_5-C_{20} cycloalkyl with a hydroxyl group. In some embodiments, the hydroxyl group-containing monomer is 2-hydroxyethylacrylate, 2-hydroxyethyl methacrylate, 2-hydroxypropyl acrylate, 2-hydroxypropyl methacrylate, 2-hydroxybutyl methacrylate, 3-hydroxypropylacrylate, 3-hydroxypropylmethacrylate, 4-hydroxybutyl methacrylate, 5-hydroxypentylacrylate, 6-hydroxyhexyl methacrylate, 1,4-cyclohexanedimethanol monoacrylate, 1,4-cyclohexanedimethanol monomethacrylate, 3-chloro-2-hydroxypropyl methacrylate, diethylene glycol monoacrylate, diethylene glycol monomethacrylate, allyl alcohol, or a combination thereof.

[00132] In some embodiments, the proportion of structural unit (b) within the binder copolymer from about 5% to about 50%, from about 5% to about 45%, from about 5% to about 40%, from about 5% to about 35%, from about 5% to about 30%, from about 5% to about 25%, from about 10% to about 50%, from about 10% to about 45%, from about 10% to about 40%, from about 10% to about 35%, from about 10% to about 30%, from about 15% to about 50%, from about 15% to about 45%, from about 15% to about 40%, from about 20% to about 50%, from about 20% to about 45%, from about 20% to about 40%, or from about 25% to about 50%, by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00133] In some embodiments, the proportion of structural unit (b) within the binder copolymer is about 5%, about 10%, about 15%, about 20%, about 25%, about 30%, about 35%, about 40%, about 45%, or about 50% by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00134] In some embodiments, the proportion of structural unit (b) within the binder copolymer is 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 binder copolymer. In some embodiments, the proportion of structural unit (b) within the binder copolymer is more than 5%, more than 10%, more than 15%, more than 20%, more than 25%, more than 30%, more than 35%, or more than 40% by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00135] In some embodiments, the binder copolymer further comprises a structural unit (c) that is derived from a monomer selected from the group consisting of a nitrile group-containing monomer, an ester group-containing monomer, an epoxy group-containing monomer, a fluorine-containing monomer, and combinations thereof.

[00136] In some embodiments, the nitrile group-containing monomer is or comprises an α,β-ethylenically unsaturated nitrile monomer. In some embodiments, the nitrile group-containing monomer is acrylonitrile, α-halogenoacrylonitrile, α-alkylacrylonitrile, or a combination 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 a combination thereof.

[00137] In some embodiments, the ester group-containing monomer is C_1-C_{20} alkyl acrylate, C_1-C_{20} alkyl methacrylate, cycloalkyl acrylate, or a combination thereof. In some embodiments, the ester group-containing monomer is methyl acrylate, ethyl acrylate, n-propyl acrylate, isopropyl acrylate, n-butyl acrylate, sec-butyl acrylate, tert-butyl acrylate, pentyl acrylate, hexyl acrylate, heptyl acrylate, octyl acrylate, 3,3,5-trimethylhexyl acrylate, 2-ethylhexyl acrylate, nonyl acrylate, decyl acrylate, lauryl acrylate, n-tetradecyl acrylate, oxtadecyl acrylate, cyclohexyl acrylate, phenyl acrylate, methoxymethyl acrylate, methoxyethyl acrylate, ethoxymethyl acrylate, ethoxyethyl acrylate, perfluorooctyl acrylate, stearyl acrylate, or a combination thereof. In some embodiments, the ester group-containing monomer is cyclohexyl acrylate, cyclohexyl methacrylate, isobornyl acrylate, isobornyl methacrylate, 3,3,5-trimethylcyclohexylacrylate, or combinations thereof. In some embodiments, the ester group-containing monomer is methyl methacrylate, ethyl methacrylate, n-propyl methacrylate, isopropyl methacrylate, n-butyl methacrylate, sec-butyl methacrylate, tert-butyl methacrylate, isobutyl methacrylate, n-pentyl methacrylate, isopentyl methacrylate, hexyl methacrylate, heptyl methacrylate, octyl methacrylate, 2-ethylhexyl methacrylate, nonyl methacrylate, decyl methacrylate, lauryl methacrylate, n-tetradecyl methacrylate, stearyl methacrylate, 2,2,2-trifluoroethyl methacrylate, phenyl methacrylate, benzyl methacrylate, or a combination thereof.

[00138] In some embodiments, the epoxy group-containing monomer is vinyl glycidyl ether, allyl glycidyl ether, allyl 2,3-epoxypropyl ether, butenyl glycidyl ether, butadiene monoepoxide, chloroprene monoepoxide, 3,4-epoxy-1-butene, 4,5-epoxy-2-pentene, 3,4-epoxy-1-vinylcyclohexane, 1,2-epoxy-4-vinylcyclohexane, 3,4-epoxy cyclohexylethylene, epoxy-4-vinylcyclohexene, 1,2-epoxy-5,9-cyclododecadiene, or a combination thereof.

[00139] In some embodiments, the epoxy group-containing monomer is 3,4-epoxy-1-butene, 1,2-epoxy-5-hexene, 1,2-epoxy-9-decene, glycidyl acrylate, glycidyl methacrylate, glycidyl crotonate, glycidyl 2,4-dimethyl pentenoate, glycidyl 4-hexenoate, glycidyl 4-heptenoate, glycidyl 5-methyl-4-heptenoate, glycidyl sorbate, glycidyl linoleate, glycidyl oleate, glycidyl 3-butenoate, glycidyl 3-pentenoate, glycidyl-4-methyl-3-pentenoate, or a combination thereof.

[00140] In some embodiments, the fluorine-containing monomer is an acrylate or methacrylate (or a combination thereof) that contains a C_1-C_{20} alkyl group and at least one fluorine atom. In some embodiments, the fluorine-containing monomer is perfluoro alkyl acrylate such as perfluoro dodecyl acrylate, perfluoro n-octyl acrylate, perfluoro n-butyl acrylate, perfluoro hexylethyl acrylate and perfluoro octylethyl acrylate; perfluoro alkyl methacrylate such as perfluoro dodecyl methacrylate, perfluoro n-octyl methacrylate, perfluoro n-butyl methacrylate, perfluoro hexylethyl methacrylate and perfluoro octylethyl methacrylate; perfluoro oxyalkyl acrylate such as perfluoro dodecyloxyethyl acrylate and perfluoro decyloxyethyl acrylate; perfluoro oxyalkyl methacrylate such as perfluoro dodecyloxyethyl methacrylate and perfluoro decyloxyethyl methacrylate; or a combination thereof. In some embodiments, the fluorine-containing monomer is a carboxylate containing at least one C_1-C_{20} alkyl group and at least one fluorine atom; wherein the carboxylate is selected from the group consisting of crotonate, malate, fumarate, itaconate, and combinations thereof. In some embodiments, the fluorine-containing monomer is vinyl fluoride, trifluoroethylene, trifluorochloroethylene, fluoroalkyl vinyl ether, perfluoroalkyl vinyl ether, hexafluoropropylene, 2,3,3,3-tetrafluoropropene, vinylidene fluoride, tetrafluoroethylene, 2-fluoro acrylate, or a combination thereof.

[00141] In some embodiments, the proportion of structural unit (c) within the binder copolymer is from about 5% to about 80%, from about 5% to about 75%, from about 5% to about 70%, from about 5% to about 65%, from about 5% to about 60%, from about 5% to about 55%, from about 5% to about 50%, from about 5% to about 45%, from about 5% to about 40%, from about 5% to about 35%, from about 5% to about 30%, from about 5% to about 25%, from about 10% to about 80%, from about 10% to about 75%, from about 10% to about 70%, from about 10% to about 65%, from about 10% to about 60%, from about 10% to about 55%, from about 10% to about 50%, from about 10% to about 45%, from about 10% to about 40%, from about 15% to about 80%, from about 15% to about 75%, from about 15% to about 70%, from about 15% to about 65%, from about 15% to about 60%, from about 15% to about 55%, from about 15% to about 50%, from about 15% to about 45%, from about 15% to about 40%, from about 20% to about 80%, from about 20% to about 75%, from about 20% to about 70%, from about 20% to about 65%, from about 20% to about 60%, from about 20% to about 55%, from about 20% to about 50%, from about 25% to about 80%, from about 25% to about 75%, from about 25% to about 70%, from about 25% to about 65%, from about 25% to about 60%, from about 25% to about 55%, from about 25% to about 50%, from about 30% to about 80%, from about 30% to about 75%, from about 30% to about 70%, from about 30% to about 65%, from about 30% to about 60%, from about 35% to about 80%, from about 35% to about 75%, from about 35% to about 70%, from about 35% to about 65%, from about 35% to about 60%, from about 40% to about 80%, from about 40% to about 75%, from about 40% to about 70%, from

about 45% to about 80%, from about 45% to about 75%, from about 45% to about 70%, from about 50% to about 80%, from about 50% to about 75%, or from about 50% to about 70% by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00142] In some embodiments, the proportion of structural unit (c) within the binder copolymer is about 5%, about 10%, about 15%, about 20%, about 25%, about 30%, about 35%, about 40%, about 45%, about 50%, about 55%, about 60%, about 65%, about 70%, about 75%, or about 80% by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00143] In some embodiments, the proportion of structural unit (c) within the binder copolymer is less than 80%, less than 75%, less than 70%, less than 65%, less than 60%, 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 binder copolymer. In some embodiments, the proportion of structural unit (c) within the binder copolymer is more than 5%, more than 10%, more than 15%, more than 20%, more than 25%, more than 30%, more than 35%, more than 40%, more than 45%, more than 50%, more than 55%, or more than 60% by mole, based on the total number of moles of monomeric units in the binder copolymer.

[00144] In certain embodiments, the binder copolymer may additionally comprise a structural unit derived from an olefin. Any hydrocarbon that has at least one carbon-carbon double bond may be used as an olefin. In some embodiments, the olefin includes a C_2-C_{20} aliphatic compound, a C_8-C_{20} aromatic compound, a 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 a combination thereof. In some embodiments, the binder copolymer does not comprise a structural unit derived from an olefin. In some embodiments, the binder copolymer does not comprise a structural unit derived from 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 or cyclooctene.

[00145] A conjugated diene group-containing monomer constitutes an olefin. In some embodiments, a conjugated diene group-containing monomer includes 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 side chain conjugated hexadienes; and combinations thereof. In some embodiments, the copolymer does not comprise a structural unit derived from C_4-C_{40} dienes, aliphatic conjugated dienes, substituted linear conjugated pentadienes or substituted side chain conjugated hexadienes.

[00146] In certain embodiments, the binder copolymer may additionally comprise a structural unit derived from an aromatic vinyl group-containing monomer. In some embodiments, the aromatic vinyl group-containing monomer is styrene, α-methylstyrene, vinyltoluene, divinylbenzene, or a combination thereof. In some embodiments, the binder copolymer does not comprise a structural unit derived from an aromatic vinyl group-containing monomer. In some embodiments, the binder copolymer does not comprise a structural unit derived from styrene, α-methylstyrene, vinyltoluene or divinylbenzene.

[00147] In some embodiments, the binder copolymer may be first dissolved, suspended or dispersed in a solvent to form a binder composition before being added to the water-based electrode slurry. In some embodiments, the solid content of the binder composition is from about 1% to about 99%, from about 5% to about 99%, from about 10% to about 99%, from about 15% to about 99%, from about 20% to about 99%, from about 30% to about 99%, from about 40% to about 99%, from about 50% to about 99%, from about 60% to about 99%, from about 70% to about 99%, from about 80% to about 99%, from about 1% to about 95%, from about 5% to about 95%, from about 10% to about 95%, from about 15% to about 95%, from about 20% to about 95%, from about 30% to about 95%, from about 40% to about 95%, from about 50% to about 95%, from about 60% to about 95%, from about 70% to about 95%, from about 1% to about 90%, from about 5% to about 90%, from about 10% to about 90%, from about 15% to about 90%, from about 20% to about 90%, from about 30% to about 90%, from about 40% to about 90%, from about 50% to about 90%, from about 60% to about 90%, from about 70% to about 90%, from about 1% to about 80%, from about 5% to about 80%, from about 10% to about 80%, from about 15% to about 80%, from about 20% to about 80%, from about 30% to about 80%, from about 40% to about 80%, from about 50% to about 80%, from about 60% to about 80%, from about 1% to about 70%, from about 5% to about 70%, from about 10% to about 70%, from about 15% to about 70%, from about 20% to about 70%, from about 30% to about 70%, from about 40% to about 70%, from about 50% to about 70%, from about 1% to about 60%, from about 5% to about 60%, from about 10% to about 60%, from about 15% to about 60%, from about 20% to about 60%, from about 30% to about 60%, from about 40% to about 60%, from about 1% to about 50%, from about 5% to about 50%, from about 10% to about 50%, from about 15% to about 50%, from about 20% to about 50%, from about 30% to about 50%, from about 1% to about 40%, from about 5% to about 40%, from about 10% to

about 40%, from about 15% to about 40%, from about 20% to about 40%, from about 1% to about 30%, from about 5% to about 30%, from about 10% to about 30%, from about 15% to about 30%, from about 1% to about 25%, from about 5% to about 25%, from about 10% to about 25%, from about 1% to about 20%, from about 5% to about 20%, from about 10% to about 20%, from about 1% to about 15%, from about 3% to about 15%, from about 5% to about 15%, from about 8% to about 15%, from about 10% to about 15%, from about 1% to about 12%, from about 3% to about 12%, from about 5% to about 12%, from about 1% to about 10%, from about 3% to about 10%, from about 5% to about 10%, or from about 1% to about 5% by weight, based on the total weight of the binder composition.

[00148] In some embodiments, the solid content of the binder composition is less than 99%, less than 95%, less than 90%, less than 80%, less than 70%, less than 60%, less than 50%, less than 40%, less than 30%, less than 25%, less than 20%, less than 15%, less than 12%, less than 10%, less than 8%, or less than 5% by weight, based on the total weight of the binder composition. In some embodiments, the solid content of the binder composition is more than 1%, more than 2%, more than 3%, more than 5%, more than 8%, more than 10%, more than 12%, more than 15%, more than 20%, more than 25%, more than 30%, more than 40%, more than 50%, more than 60%, more than 70%, more than 80%, more than 90%, or more than 95% by weight, based on the total weight of the binder composition.

[00149] In some embodiments, the water-based electrode slurry comprises an aqueous solvent. In certain 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, de-ionized water (DI water), D_2O, and combinations thereof.

[00150] In some embodiments, the aqueous solvent can further comprise a minor component in addition to water. In some embodiments, the volume ratio of water to the minor component is from about 51:49 to about 99:1. Any water-miscible or volatile solvents can be used as the minor component of the aqueous solvent. Some non-limiting examples of the minor component include alcohols, lower aliphatic ketones, lower alkyl acetates, and combinations thereof. The addition of alcohol can improve the processibility of the polymerization process. 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 the minor component include 1,4-dioxane, diethyl ether, tetrahydrofuran (THF), chloroform, dichloromethane, acetonitrile, dimethyl sulfoxide, nitromethane, propylene carbonate, ethylene carbonate, dimethyl carbonate,

pyridine, acetaldehyde, acetic acid, propanoic acid, butyric acid, furfuryl alcohol, diethanolamine, dimethylacetamide (DMAc), and dimethylformamide (DMF). In some embodiments, no minor component is present in the aqueous solvent.

[00151] In some embodiments, the water-based electrode slurry may additionally comprise a conductive agent. The conductive agent enhances the electrically-conducting properties of an electrode. Therefore, it may be advantageous for the water-based electrode slurry to comprise a conductive agent. 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, 0-dimensional KS6, vapor grown carbon fibers (VGCF), mesoporous carbon, and combinations thereof. In certain embodiments, the conductive agent does not comprise a carbonaceous material.

[00152] 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 plays two roles simultaneously, not only as a conductive agent but also as a binder. In certain embodiments, the electrode layer comprises two components, the electrode active material and conductive polymer. In other embodiments, the electrode layer comprises the electrode active material, conductive agent and conductive polymer. In certain embodiments, the conductive polymer is an additive and the electrode layer comprises the electrode active material, conductive agent, binder material, and conductive polymer. In other embodiments, the conductive agent does not comprise a conductive polymer.

[00153] In some embodiments, the proportion of coated cathode active material in the solid portion of the water-based electrode slurry is from about 60% to about 99%, from about 70% to about 99%, from about 75% to about 99%, from about 80% to about 99%, from about 85% to about 99%, from about 90% to about 99%, from about 60% to about 95%, from about 65% to about 95%, from about 70% to about 95%, from about 75% to about 95%, from about 80% to about 95%, from about 85% to about 95%, from about 60% to about 90%, from about 65% to about 90%, from about 70% to about 90%, from about 75% to about 90%, from about 80% to about 90%, from about 60% to about 85%, from about 65% to about 85%, from about 70% to about 85%, from about 75% to about 85%, from about 60% to about 80%, from about 65% to about 80%, or from about 70% to about 80% by weight, based on the total weight of the solid portion of the electrode slurry.

[00154] In some embodiments, the proportion of coated cathode active material in the

solid portion of the water-based electrode slurry is less than 99%, less than 95%, less than 90%, less than 85%, less than 80%, less than 75%, less than 70%, or less than 65% by weight, based on the total weight of the solid portion of the electrode slurry. In some embodiments, the proportion of coated cathode active material in the solid portion of the water-based electrode slurry is more than 60%, more than 65%, more than 70%, more than 75%, more than 80%, more than 85%, more than 90%, or more than 95% by weight, based on the total weight of the solid portion of the electrode slurry.

[00155] In some embodiments, the proportion of binder polymer in the solid portion of the water-based electrode slurry is from about 1% to about 20%, from about 2% to about 20%, from about 3% to about 20%, from about 4% to about 20%, from about 5% to about 20%, from about 6% to about 20%, from about 7% to about 20%, from about 8% to about 20%, from about 9% to about 20%, from about 10% to about 20%, from about 11% to about 20%, from about 12% to about 20%, from about 13% to about 20%, from about 14% to about 20%, from about 15% to about 20%, from about 1% to about 15%, from about 2% to about 15%, from about 3% to about 15%, from about 4% to about 15%, from about 5% to about 15%, from about 6% to about 15%, from about 7% to about 15%, from about 8% to about 15%, from about 9% to about 15%, from about 10% to about 15%, 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 1% to about 5%, from about 2% to about 5%, or from about 3% to about 5% by weight, based on the total weight of the solid portion of the electrode slurry.

[00156] In some embodiments, the proportion of binder polymer in the solid portion of the water-based electrode slurry is 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%, less than 8%, less than 7%, less than 6%, or less than 5% by weight, based on the total weight of the solid portion of the electrode slurry. In some embodiments, the proportion of binder polymer in the solid portion of the water-based electrode slurry 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%, more than 9%, more than 10%, more than 11%, more than 12%, more than 13%, more than 14%, or more than 15% by weight, based on the total weight of the solid portion of the electrode slurry.

[00157] In some embodiments, the proportion of conductive agent in the solid portion of the water-based electrode slurry is from about 1% to about 20%, from about 2% to about 20%, from about 3% to about 20%, from about 4% to about 20%, from about 5% to about 20%, from about 6% to about 20%, from about 7% to about 20%, from about 8% to about 20%, from about 9% to about 20%, from about 10% to about 20%, from about 11% to about 20%, from about 12% to about 20%, from about 13% to about 20%, from about 14% to about 20%, from about 15% to about 20%, from about 1% to about 15%, from about 2% to about 15%, from about 3%

to about 15%, from about 4% to about 15%, from about 5% to about 15%, from about 6% to about 15%, from about 7% to about 15%, from about 8% to about 15%, from about 9% to about 15%, from about 10% to about 15%, 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 1% to about 5%, from about 2% to about 5%, or from about 3% to about 5% by weight, based on the total weight of the solid portion of the electrode slurry.

[00158] In some embodiments, the proportion of conductive agent in the solid portion of the water-based electrode slurry is 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%, less than 8%, less than 7%, less than 6%, or less than 5% by weight, based on the total weight of the solid portion of the electrode slurry. In some embodiments, the proportion of conductive agent in the solid portion of the water-based electrode slurry 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%, more than 9%, more than 10%, more than 11%, more than 12%, more than 13%, more than 14%, or more than 15% by weight, based on the total weight of the solid portion of the electrode slurry.

[00159] In some embodiments, the solid content of the water-based electrode slurry is from about 40% to about 80%, from about 40% to about 75%, from about 40% to about 70%, from about 40% to about 65%, from about 40% to about 60%, from about 40% to about 55%, from about 45% to about 80%, from about 45% to about 75%, from about 45% to about 70%, from about 45% to about 65%, from about 45% to about 60%, from about 50% to about 80%, from about 50% to about 75%, from about 50% to about 70%, from about 50% to about 65%, from about 55% to about 80%, from about 55% to about 75%, from about 55% to about 70%, from about 60% to about 80%, from about 60% to about 75%, from about 65% to about 80%, from about 65% to about 75%, or from about 70% to about 80% by weight, based on the total weight of the electrode slurry.

[00160] In some embodiments, the solid content of the water-based electrode slurry is less than 80%, less than 75%, less than 70%, less than 65%, less than 60%, less than 55%, or less than 50% by weight, based on the total weight of the electrode slurry. In some embodiments, the solid content of the water-based electrode slurry is more than 40%, more than 45%, more than 50%, more than 55%, more than 60%, more than 65%, or more than 70% by weight, based on the total weight of the electrode slurry.

[00161] In some embodiments, the pH of the water-based electrode slurry is from about 8 to about 14, from about 8 to about 13.5, from about 8 to about 13, from about 8 to about 12.5, from about 8 to about 12, from about 8 to about 11.5, from about 8 to about 11, from about 8 to about 10.5, from about 8 to about 10, from about 8.5 to about 14, from about 8.5 to about 13.5,

from about 8.5 to about 13, from about 8.5 to about 12.5, from about 8.5 to about 12, from about 8.5 to about 11.5, from about 8.5 to about 11, from about 8.5 to about 10.5, from about 9 to about 14, from about 9 to about 13.5, from about 9 to about 13, from about 9 to about 12.5, from about 9 to about 12, from about 9 to about 11.5, from about 9 to about 11, from about 9.5 to about 14, from about 9.5 to about 13.5, from about 9.5 to about 13, from about 9.5 to about 12.5, from about 9.5 to about 12, from about 9.5 to about 11.5, from about 10 to about 14, from about 10 to about 13.5, from about 10 to about 13, from about 10 to about 12.5, from about 10 to about 12, from about 10.5 to about 14, from about 10.5 to about 13.5, from about 10.5 to about 13, from about 10.5 to about 12.5, from about 11 to about 14, from about 11 to about 13.5, from about 11 to about 13, from about 11.5 to about 14, from about 11.5 to about 13.5, or from about 12 to about 14.

[00162] In certain embodiments, the pH of the water-based electrode slurry is less than 14, less than 13.5, less than 13, less than 12.5, less than 12, less than 11.5, less than 11, less than 10.5, less than 10, less than 9.5, less than 9, or less than 8.5. In some embodiments, the pH of the water-based electrode slurry is more than 8, more than 8.5, more than 9, more than 9.5, more than 10, more than 10.5, more than 11, more than 11.5, more than 12, more than 12.5, more than 13, or more than 13.5.

[00163] There is no particular limitation on the method used to produce the water-based electrode slurry from the various electrode components, except that all electrode components should be added into a homogenizer and mixed to form the water-based electrode slurry. In some embodiments, all the materials used to produce the water-based electrode slurry are added into the homogenizer in a single batch. In other embodiments, each electrode component of the water-based electrode slurry can be added to the homogenizer in one or more batches, and each batch may comprise more than one electrode material component. The homogenizer may be equipped with a temperature control system and the temperature of the water-based electrode slurry can be controlled by the temperature control system. Any homogenizer that can reduce or eliminate particle aggregation and/or promote homogeneous distribution of electrode components in the electrode slurry can be used herein. Homogeneous distribution plays an important role in fabricating batteries with good battery performance. In some embodiments, the homogenizer is a planetary stirring mixer, a stirring mixer, a blender, or an ultrasonicator.

[00164] There are no particular limitations to the conditions used to form a water-based electrode slurry, except such conditions should be sufficient to produce a homogenous slurry with good dispersion of the electrode components within the solvent of the slurry. There is no particular limitation on the time taken in homogenization to produce the water-based electrode

slurry, except that the time period should be sufficient to ensure homogeneous distribution of the various electrode components in the solvent of the electrode slurry. There is no particular limitation to the temperature used in homogenization to form the water-based electrode slurry, except that the temperature used should allow the homogenization to occur smoothly and the electrode slurry to be processed easily. There is no particular limitation to the stirring speed used in homogenization to form the water-based electrode slurry, except that the stirring speed should be sufficient to ensure homogeneous distribution of the various electrode components in the solvent of the electrode slurry.

[00165] Due to the coating of the cathode active material, degradation of the cathode active material through dissolution of lithium into the aqueous solvent of the water-based electrode slurry is suppressed. In some embodiments, the concentration of lithium ions (Li^+) in the water-based electrode slurry is from about 0.05 M to about 1.25 M, from about 0.1 M to about 1.25 M, from about 0.15 M to about 1.25 M, from about 0.2 M to about 1.25 M, from about 0.25 M to about 1.25 M, from about 0.3 M to about 1.25 M, from about 0.35 M to about 1.25 M, from about 0.4 M to about 1.25 M, from about 0.45 M to about 1.25 M, from about 0.5 M to about 1.25 M, from about 0.55 M to about 1.25 M, from about 0.6 M to about 1.25 M, from about 0.65 M to about 1.25 M, from about 0.7 M to about 1.25 M, from about 0.75 M to about 1.25 M, from about 0.05 M to about 1 M, from about 0.1 M to about 1 M, from about 0.15 M to about 1 M, from about 0.2 M to about 1 M, from about 0.25 M to about 1 M, from about 0.3 M to about 1 M, from about 0.35 M to about 1 M, from about 0.4 M to about 1 M, from about 0.45 M to about 1 M, from about 0.5 M to about 1 M, from about 0.55 M to about 1 M, from about 0.6 M to about 1 M, from about 0.05 M to about 0.75 M, from about 0.1 M to about 0.75 M, from about 0.15 M to about 0.75 M, from about 0.2 M to about 0.75 M, from about 0.25 M to about 0.75 M, from about 0.3 M to about 0.75 M, from about 0.35 M to about 0.75 M, from about 0.4 M to about 0.75 M, from about 0.45 M to about 0.75 M, from about 0.5 M to about 0.75 M, from about 0.05 M to about 0.5 M, from about 0.1 M to about 0.5 M, from about 0.15 M to about 0.5 M, from about 0.2 M to about 0.5 M, from about 0.25 M to about 0.5 M, or from about 0.3 M to about 0.5 M.

[00166] In some embodiments, the concentration of lithium ions (Li^+) in the water-based electrode slurry is less than 1.25 M, less than 1.2 M, less than 1.15 M, less than 1.1 M, less than 1.05 M, less than 1 M, less than 0.95 M, less than 0.9 M, less than 0.85 M, less than 0.8 M, less than 0.75 M, less than 0.7 M, less than 0.65 M, less than 0.6 M, less than 0.55 M, less than 0.5 M, less than 0.45 M, less than 0.4 M, less than 0.35 M, less than 0.3 M, or less than 0.25 M. In some embodiments, the concentration of lithium ions (Li^+) in the water-based electrode slurry is more than 0.05 M, more than 0.1 M, more than 0.15 M, more than 0.2 M, more than 0.25 M,

more than 0.3 M, more than 0.35 M, more than 0.4 M, more than 0.45 M, more than 0.5 M, more than 0.55 M, more than 0.6 M, more than 0.65 M, more than 0.7 M, more than 0.75 M, more than 0.8 M, more than 0.85 M, more than 0.9 M, more than 0.95 M, more than 1 M, or more than 1.05 M.

[00167] In some embodiments, after homogenization of the water-based electrode slurry, the water-based electrode slurry can be coated onto one side or both sides of a current collector to form a coated electrode film. There is no particular limitation to the equipment and the conditions used in coating the electrode slurry to form a coated electrode film, except that a homogeneous, flat and smooth coated electrode film should be formed. In certain embodiments, the coating process is performed using a doctor blade coater, a slot-die coater, a transfer coater, a spray coater, a roll coater, a gravure coater, a dip coater, or a curtain coater. In some embodiments, the water-based electrode slurry is applied or calendered directly onto a current collector. In other embodiments, the water-based electrode slurry is first applied or calendered onto a release film to form a free-standing electrode layer. The free-standing electrode layer is then combined with a current collector and pressed to form the coated electrode film on the current collector.

[00168] 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, 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, 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 more than three layers.

[00169] In some embodiments, a conductive layer can be coated on a current collector to improve its current conductivity. In some embodiments, the conductive layer is positioned between the current collector and the electrode layer. 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.

[00170] In some embodiments, the conductive layer has a thickness from about 0.5 μm to about 5.0 μm. The 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.

[00171] 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.

[00172] 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 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.

[00173] In some embodiments, following the coating of the water-based electrode slurry onto the current collector to form a coated electrode film, the coated electrode film is dried to form an electrode. Any equipment that can dry the coated electrode film in order to affix the coated electrode film to the current collector can be used herein. Some non-limiting examples of equipment that can be used to dry the coated electrode 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. In some embodiments, the drying oven includes one or more heating sections that are individually temperature-controlled, wherein each heating section comprises one or more individually temperature-controlled heating elements.

[00174] There are no particular limitations to the conditions used for drying, except that the drying conditions should be sufficient to ensure that the electrode layer adheres strongly to the current collector. However, drying the coated electrode film at temperatures above 150 °C may result in undesirable deformation of the electrode, thus affecting the performance of the electrode. The drying time can be regulated, for example by controlling the conveyor length and speed when a conveyor drying oven is used to dry the coated electrode film on the current collector. Such a drying time should be optimized with respect to the other drying conditions such as drying temperature, in order to ensure that the aqueous solvent is removed from the coated electrode film.

[00175] In some embodiments, the electrode is compressed mechanically following drying in order to increase the density of the electrode. In some embodiments, the compressed coated electrode film on the current collector is designated as an electrode layer. In some embodiments, when the coated electrode film comprises a cathode active material, such as the coated cathode active material of the present invention, the electrode layer is specifically a cathode electrode layer.

[00176] In some embodiments, the proportion of coated cathode active material in the electrode layer of the electrode is from about 60% to about 99%, from about 70% to about 99%, from about 75% to about 99%, from about 80% to about 99%, from about 85% to about 99%, from about 90% to about 99%, from about 60% to about 95%, from about 65% to about 95%, from about 70% to about 95%, from about 75% to about 95%, from about 80% to about 95%, from about 85% to about 95%, from about 60% to about 90%, from about 65% to about 90%, from about 70% to about 90%, from about 75% to about 90%, from about 80% to about 90%, from about 60% to about 85%, from about 65% to about 85%, from about 70% to about 85%,

from about 75% to about 85%, from about 60% to about 80%, from about 65% to about 80%, or from about 70% to about 80% by weight, based on the total weight of the electrode layer.

[00177] In some embodiments, the proportion of coated cathode active material in the electrode layer of the electrode is less than 99%, less than 95%, less than 90%, less than 85%, less than 80%, less than 75%, less than 70%, or less than 65% by weight, based on the total weight of the electrode layer. In some embodiments, the proportion of coated cathode active material in the electrode layer of the electrode is more than 60%, more than 65%, more than 70%, more than 75%, more than 80%, more than 85%, more than 90%, or more than 95% by weight, based on the total weight of the electrode layer.

[00178] In some embodiments, the proportion of binder polymer in the electrode layer of the electrode is from about 1% to about 20%, from about 2% to about 20%, from about 3% to about 20%, from about 4% to about 20%, from about 5% to about 20%, from about 6% to about 20%, from about 7% to about 20%, from about 8% to about 20%, from about 9% to about 20%, from about 10% to about 20%, from about 11% to about 20%, from about 12% to about 20%, from about 13% to about 20%, from about 14% to about 20%, from about 15% to about 20%, from about 1% to about 15%, from about 2% to about 15%, from about 3% to about 15%, from about 4% to about 15%, from about 5% to about 15%, from about 6% to about 15%, from about 7% to about 15%, from about 8% to about 15%, from about 9% to about 15%, from about 10% to about 15%, 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 1% to about 5%, from about 2% to about 5%, or from about 3% to about 5% by weight, based on the total weight of the electrode layer.

[00179] In some embodiments, the proportion of binder polymer in the electrode layer of the electrode is 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%, less than 8%, less than 7%, or less than 6% by weight, based on the total weight of the electrode layer. In some embodiments, the proportion of binder polymer in the electrode layer of the electrode 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%, more than 9%, more than 10%, more than 11%, more than 12%, more than 13%, more than 14%, or more than 15% by weight, based on the total weight of the electrode layer.

[00180] In some embodiments, the proportion of conductive agent in the electrode layer of the electrode is from about 1% to about 20%, from about 2% to about 20%, from about 3% to about 20%, from about 4% to about 20%, from about 5% to about 20%, from about 6% to about 20%, from about 7% to about 20%, from about 8% to about 20%, from about 9% to about 20%, from about 10% to about 20%, from about 11% to about 20%, from about 12% to about 20%,

from about 13% to about 20%, from about 14% to about 20%, from about 15% to about 20%, from about 1% to about 15%, from about 2% to about 15%, from about 3% to about 15%, from about 4% to about 15%, from about 5% to about 15%, from about 6% to about 15%, from about 7% to about 15%, from about 8% to about 15%, from about 9% to about 15%, from about 10% to about 15%, 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 1% to about 5%, from about 2% to about 5%, or from about 3% to about 5% by weight, based on the total weight of the electrode layer.

[00181] In some embodiments, the proportion of conductive agent in the electrode layer of the electrode is 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%, less than 8%, less than 7%, or less than 6% by weight, based on the total weight of the electrode layer. In some embodiments, the proportion of conductive agent in the electrode layer of the electrode 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%, more than 9%, more than 10%, more than 11%, more than 12%, more than 13%, more than 14%, or more than 15% by weight, based on the total weight of the electrode layer.

[00182] In certain embodiments, the thickness of the electrode layer on the current collector is from about 5 μm to about 90 μm, from about 5 μm to about 50 μm, from about 5 μm to about 25 μm, from about 10 μm to about 90 μm, from about 10 μm to about 50 μm, from about 10 μm to about 30 μm, from about 15 μm to about 90 μm, from about 20 μm to about 90 μm, from about 25 μm to about 90 μm, from about 25 μm to about 80 μm, from about 25 μm to about 70 μm, from about 25 μm to about 50 μm, from about 30 μm to about 90 μm, or from about 30 μm to about 80 μm. In some embodiments, the thickness of the cathode electrode layer on the current collector is more than 5 μm, more than 10 μm, more than 15 μm, more than 20 μm, more than 25 μm, more than 30 μm, more than 40 μm, more than 50 μm, more than 60 μm, more than 70 μm, or more than 80 μm. In some embodiments, the thickness of the cathode electrode layer on the current collector is less than 90 μm, less than 80 μm, less than 70 μm, less than 60 μm, less than 50 μm, less than 40 μm, less than 30 μm, less than 25 μm, less than 20 μm, less than 15 μm, or less than 10 μm.

[00183] In some embodiments, the surface density of the electrode layer on the current collector is from about 1 mg/cm^2 to about 50 mg/cm^2, from about 2.5 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 1 mg/cm^2 to about 30 mg/cm^2, from about 2.5 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 2.5 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 2.5 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. In some embodiments, the surface density of the cathode electrode layer on the current collector is less than 50 mg/cm^2, less than 40 mg/cm^2, less than 30 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 2.5 mg/cm^2. In some embodiments, the surface density of the cathode electrode layer on the current collector is more than 1 mg/cm^2, more than 2.5 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 30 mg/cm^2, or more than 40 mg/cm^2.

[00184] In some embodiments, the density of the electrode layer on the current collector 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 3.5 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. In some embodiments, the density of the cathode electrode layer on the current collector is less than 7.5 g/cm^3, less than 6.5 g/cm^3, less than 5.5 g/cm^3, less than 4.5 g/cm^3, less than 3.5 g/cm^3, less than 2.5 g/cm^3, less than 2 g/cm^3, or less than 1.5 g/cm^3. In some embodiments, the density of the cathode electrode layer on the current collector 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 3.5 g/cm^3, more than 4.5 g/cm^3, or more than 5.5 g/cm^3.

[00185] In addition, an electrode prepared via a water-based electrode slurry comprising the coated cathode active material of the present invention 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 as this prevents delamination or separation of the electrode, which would greatly impact the mechanical stability of the electrodes and the cyclability of the battery. Therefore, the electrodes should have sufficient peeling strength to withstand the rigors of battery manufacture.

[00186] In some embodiments, the peeling strength between the current collector and the

electrode layer of the electrode is in the range of 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.

[00187] In some embodiments, the peeling strength between the current collector and the electrode layer of the electrode 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. In some embodiments, the peeling strength between the current collector and the electrode layer of the electrode is less than 8.0 N/cm, less than 7.5 N/cm, less than 7.0 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.

[00188] By coating the cathode active material with a phosphorus-containing compound, degradation of the cathode active material due to reaction with water can be suppressed. As a result, the coated cathode active material of the present invention can be successfully used in a water-based electrode slurry. Batteries comprising electrodes produced using a water-based electrode slurry comprising coated cathode active material of the present invention have improved electrochemical performance, because performance losses due to cathode active material degradation as a result of reaction with water in the water-based electrode slurry can be avoided through the coating of the cathode active material using one or more phosphorus-containing compounds disclosed. Furthermore, water-based electrode slurries comprising the coated cathode active material of the present invention can be stored or transported without concern of cathode active material degradation due to reaction with water, thereby increasing logistical flexibility in production of electrodes.

[00189] 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 described in each example should not be construed as necessary features of the invention.

EXAMPLES

[00190] 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 (N). The mean roughness depth (R_z) of the current collector was 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.

[00191] The amount of degradation of cathode active material, whether coated or uncoated, was determined by measuring the amount of lithium dissolved into water from a cathode active material sample. 2.0 g of a cathode active material sample was added to 18.0 g of DI water, then stirred at a speed of 250 rpm at room temperature for 3 hours. The mixture was filtered using 200 μm nylon mesh. The filtrate of the mixture was then analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES) to determine the amount of lithium present in the filtrate. The percentage of lithium dissolved in the filtrate that originated from the cathode active material sample was then determined. This dissolved lithium percentage was normalized with respect to the percentage of lithium dissolved in the filtrate of a baseline test. The baseline test was performed by adding 2.0 g of an uncoated cathode active material sample to 18.0 g of DI water, then stirring at a speed of 250 rpm at room temperature for 3 hours and filtering the mixture with 200 μm nylon mesh.

[00192] The amount of phosphorus-containing compound coated onto the cathode active material is measured by the amount of phosphorus present in a coated cathode active material sample. 0.2 g of a coated cathode active material was digested with acid, then analyzed by ICP-OES to determine the amount of phosphorus present in the coated cathode active material sample. When the specific surface area of the cathode active material and the molecular mass of the phosphorus-containing compound are known, the amount of phosphorus-containing compound coated per unit surface area of cathode active material in terms of mol/m^2 could then be calculated.

Example 1

A) Preparation of Coated Cathode Active Material

[00193] The coating reaction was conducted in an argon-filled glovebox. 0.45 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was a methyl group and R_2 was a hydroxyl group, was added to a conical flask containing 180 g of anhydrous ethanol. The mixture was stirred at 300 rpm at room temperature until all the phosphorus-containing compound was dissolved in the ethanol to form a coating mixture.

[00194] 75 g of NMC811 with a specific surface area of 0.75 m^2/g (obtained from Shandong Tianjiao New Energy Co., Ltd, China) was added into the coating mixture, then the resultant reaction mixture was stirred at 450 rpm at room temperature for 6 hours to carry out the coating reaction.

[00195] Following the coating reaction, the product mixture was filtered and washed with additional anhydrous ethanol, then dried in a vacuum oven at 80 °C for 24 hours to obtain a coated cathode active material. The amount of phosphorus-containing compound coated per unit surface area of cathode active material was 21.9 $\mu mol/m^2$.

B) Preparation of Binder Material

[00196] 7.45 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.

[00197] 16.77 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.

[00198] 7.19 g of acrylamide was dissolved in 10 g of DI water to form an acrylamide solution. Thereafter, 17.19 g of 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.

[00199] 35.95 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.

[00200] 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. 3.015 g of APS solution and 1.5075 g of 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.

[00201] After the complete reaction, the temperature of the fifth suspension was lowered to 25 °C. 3.72 g of NaOH was dissolved in 400 g of DI water. Thereafter, 403.72 g of sodium

hydroxide solution was added dropwise into the fifth suspension to adjust pH to 7.3 to form the binder material. The binder material was filtered using 200 μm nylon mesh. The solid content of the binder material was 8.88 wt.%.

C) Preparation of Positive Electrode

[00202] A first mixture was prepared by dispersing 3 g of conductive agent (SuperP; obtained from Timcal Ltd, Bodio, Switzerland) and 25 g of the binder material described above in 18.5 g of deionized water while stirring with an overhead stirrer (R20, IKA). After the addition, the first mixture was further stirred for about 30 minutes at 25 °C at a speed of 1,200 rpm.

[00203] Thereafter, a second mixture was prepared by adding 69 g of the coated cathode active material described above to the first mixture at 25 °C while stirring with an overhead stirrer. Then, the second mixture was degassed under a pressure of about 10 kPa for 1 hour. Then, the second mixture was further stirred for about 60 minutes at 25 °C at a speed of 1,200 rpm to form a homogenized cathode slurry.

[00204] The homogenized cathode slurry was coated onto one side of a current collector, an aluminum foil of thickness 16 μm, using a doctor blade coater with a gap width of 120 μm. The coated slurry of 80 μm on the aluminum foil was dried to form a cathode electrode layer using an electrically heated oven at 80 °C for about 120 minutes. The electrode was then pressed to decrease the thickness of the cathode electrode layer to 34 μm. The surface density of the cathode electrode layer on the current collector was 5 mg/cm^2.

D) Assembling of Coin Cell

[00205] The electrochemical performance of the cathode prepared above was tested in CR2032 coin-type Li cells assembled in an argon-filled glove box. The coated cathode sheet was cut into disc-form positive electrodes for coin-type cell assembly. A lithium metal foil having a thickness of 500 μm was used as a counter electrode. The cathode and counter electrode plates were kept apart by separators. 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 then 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.

[00206] 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.

F) Electrochemical Measurements

[00207] The coin cells were analyzed in a constant current mode using a multi-channel battery tester (BTS-4008-5V10mA, obtained from Neware Electronics Co. Ltd, China). An initial cycle at C/2 was completed and the discharge capacity was recorded. Then, the coin cells were further repeatedly charged and discharged at a rate of C/2. The charging/discharging cycling tests of the batteries were performed between 3.0 and 4.3 V at a current density of C/2 at 25 °C to obtain the capacity retention after 50 cycles. The electrochemical performance of the coin cells of Example 1 is shown in Table 1 below.

Example 2

[00208] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.64 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was a butyl group and R_2 was a hydroxyl group, and 255 g of anhydrous ethanol were used to form the coating mixture. The amount of phosphorus-containing compound coated per unit surface area of cathode active material particles was 4.1 $\mu mol/m^2$.

Example 3

[00209] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.90 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was an octyl group and R_2 was a hydroxyl group, and 360 g of anhydrous ethanol were used to form the coating mixture. The amount of phosphorus-containing compound coated per unit surface area of cathode active material was 2.2 $\mu mol/m^2$.

[00210] Four more iterations of Example 3 were prepared with minor changes to the composition of the binder material. The binder material of each iteration was prepared according to the method described in Example 1, except that the amount of each reactant used was changed as described below.

[00211] In the first iteration, 27.27 g of sodium hydroxide was added in the preparation of the first suspension, 52.48 g of acrylic acid was added in the preparation of the second suspension, 8.63 g of acrylamide was added in the preparation of the third suspension, and 8.59 g of acrylonitrile was added in the preparation of the fourth suspension.

[00212] In the second iteration, 18.37 g of sodium hydroxide was added in the preparation of the first suspension, 36.44 g of acrylic acid was added in the preparation of the second suspension, 23.73 g of acrylamide was added in the preparation of the third suspension, and 9.12 g of acrylonitrile was added in the preparation of the fourth suspension.

[00213] In the third iteration, 3.00 g of sodium hydroxide was added in the preparation of

the first suspension, 8.75 g of acrylic acid was added in the preparation of the second suspension, 7.19 g of acrylamide was added in the preparation of the third suspension, and 41.86 g of acrylonitrile was added in the preparation of the fourth suspension.

[00214] In the fourth iteration, 11.50 g of sodium hydroxide was added in the preparation of the first suspension, 24.06 g of acrylic acid was added in the preparation of the second suspension, 9.07 g of acrylamide was added in the preparation of the third suspension, and 29.18 g of acrylonitrile was added in the preparation of the fourth suspension.

Example 4

[00215] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 1.56 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was an octadecyl group and R_2 was a hydroxyl group, and 620 g of anhydrous ethanol were used to form the coating mixture. The amount of phosphorus-containing compound coated per unit surface area of cathode active material was 0.94 μmol/m^2.

Example 5

[00216] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.58 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was a methyl group and R_2 was an ethoxy group, and 230 g of anhydrous ethanol were used to form the coating mixture.

Example 6

[00217] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, except 0.44 g of a phosphorus-containing compound represented by general formula (1), wherein both R_1 and R_2 were methyl groups, and 175 g of anhydrous ethanol were used to form the coating mixture.

Example 7

[00218] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.24 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was a methyl group and R_2 was a hydroxyl group, and 95 g of anhydrous ethanol was used to form the coating mixture, and 75 g of $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO; obtained from Chengdu Xingneng New Materials Co. Ltd, China) was used instead of NMC811.

Example 8

[00219] A positive electrode was prepared with the method described in Example 7, except that when preparing the coated cathode active material, 0.44 g of a phosphorus-

containing compound represented by general formula (1), wherein R_1 was an octyl group and R_2 was a hydroxyl group, and 193 g of anhydrous ethanol was used to form the coating mixture.

Example 9

[00220] A positive electrode was prepared with the method described in Example 7, except that when preparing the coated cathode active material, 0.83 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was an octadecyl group and R_2 was a hydroxyl group, and 330 g of anhydrous ethanol was used to form the coating mixture.

Example 10

[00221] A positive electrode was prepared with the method described in Example 1, except that the coated cathode active material was prepared using 75 g of a core-shell cathode active material comprising NMC532 as the core and $Li_{0.95}Ni_{0.53}Mn_{0.29}Co_{0.15}Al_{0.03}O_2$ as the shell instead of NMC811. The cathode active material has a particle size D50 of about 35 μm, and a shell thickness of about 3 μm.

Example 11

[00222] A positive electrode was prepared with the method described in Example 10, except that when preparing the coated cathode active material, 0.91 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was an octyl group and R_2 was a hydroxyl group, and 366 g of anhydrous ethanol was used to form the coating mixture.

Example 12

[00223] A positive electrode was prepared with the method described in Example 10, except that when preparing the coated cathode active material, 1.58 g of a phosphorus-containing compound represented by general formula (1), wherein R_1 was an octadecyl group and R_2 was a hydroxyl group, and 630 g of anhydrous ethanol was used to form the coating mixture.

Example 13

[00224] A positive electrode was prepared with the method described in Example 3, except that the coated cathode active material was prepared using 75 g of NMC532 (obtained from Shandong Tianjiao New Energy Co., Ltd, China) instead of NMC811. The specific surface area of the NMC532 was 0.38 m^2/g and the amount of phosphorus-containing compound coated per unit surface area of cathode active material was 3.0 $\mu mol/m^2$.

Example 14

[00225] A positive electrode was prepared with the method described in Example 3,

except that the coated cathode active material was prepared using 75 g of NMC622 (obtained from Shandong Tianjiao New Energy Co., Ltd, China) instead of NMC811. The specific surface area of the NMC622 was 0.44 m^2/g and the amount of phosphorus-containing compound coated per unit surface area of cathode active material was 2.5 $\mu mol/m^2$.

Example 15

[00226] A positive electrode was prepared with the method described in Example 3, except that the coated cathode active material was prepared using 75 g of $LiNi_{0.88}Co_{0.1}Al_{0.02}O_2$ (NCA; obtained from Shandong Tianjiao New Energy Co., Ltd, China) instead of NMC811. The specific surface area of the NCA was 0.75 m^2/g and the amount of phosphorus-containing compound coated per unit surface area of cathode active material was 2.1 $\mu mol/m^2$.

Assembling of Coin Cells of Examples 2-15

[00227] The coin cells of Examples 2-15 were assembled in the same manner as Example 1.

Electrochemical Measurements of Example 2

[00228] Electrochemical measurements were taken by the same method described in Example 1. The electrochemical performance of the coin cell of Example 2 was measured and is shown in Table 1 below.

Electrochemical Measurements of Example 3

[00229] Electrochemical measurements were taken by the same method described in Example 1. The electrochemical performance of the coin cell of Example 3 was measured and is shown in Table 1 below. The electrochemical performances of coin cells of the various iterations of the binder material were also measured, and it was found that these coin cells had an electrochemical performance similar to that of the coin cell of Example 3.

[00230] Furthermore, the amount of degradation of the coated cathode active material of Example 3 was also measured.

Electrochemical Measurements of Example 4

[00231] Electrochemical measurements were taken by the same method described in Example 1. The electrochemical performance of the coin cell of Example 4 was measured and is shown in Table 1 below.

[00232] Electrochemical impedance spectroscopy (EIS) was also conducted on another coin cell of Example 4 using a CHI660E spectroscope (Beijing Chinese Science Days Technology Co. Ltd., China). The coin cell was first subjected to an initial charge/discharge cycle at C/20. Following initial cycling, the coin cell was connected to the spectroscope and the impedance of the coin cell was obtained for current frequencies between 0.01 and 10,000 Hz,

with an initial voltage of 3.3V and a voltage amplitude of 5 mV. The real and imaginary components of the resultant impedance data was then plotted on a Nyquist plot. The Nyquist plot of the coin cell of Example 4 is shown in Figure 2.

[00233] Furthermore, the amount of degradation of the coated cathode active material of Example 4 was also measured.

Electrochemical Measurements of Examples 5-15

[00234] Electrochemical measurements were taken by the same method described in Example 1. The electrochemical performance of the coin cells of Examples 5-15 were measured and is shown in Table 1 below.

Comparative Example 1

[00235] A positive electrode was prepared in the same manner as Example 1, except that 69 g of uncoated NMC811 was used instead of the coated cathode active material of Example 1.

Comparative Example 2

[00236] A positive electrode was prepared in the same manner as Example 1, except that 69 g of uncoated NMC532 was used instead of the coated cathode active material of Example 1.

Comparative Example 3

[00237] A positive electrode was prepared in the same manner as Example 1, except that 69 g of uncoated NMC622 was used instead of the coated cathode active material of Example 1.

Comparative Example 4

[00238] A positive electrode was prepared in the same manner as Example 1, except that 69 g of uncoated NCA was used instead of coated cathode active material of Example 1.

Comparative Example 5

[00239] A positive electrode was prepared in the same manner as Example 1, except that 69 g of uncoated LNMO was used instead of the coated cathode active material of Example 1.

Comparative Example 6

[00240] A positive electrode was prepared in the same manner as Example 1, except that 69 g of uncoated core-shell cathode active material comprising NMC532 as the core and $Li_{0.95}Ni_{0.53}Mn_{0.29}Co_{0.15}Al_{0.03}O_2$ as the shell was used instead of the coated cathode active material of Example 1. The cathode active material has a particle size D50 of about 35 μm, and a shell thickness of about 3 μm.

Comparative Example 7

[00241] A positive electrode was prepared with the method described in Example 1,

except that when preparing the coated cathode active material, 1.43 g of mono-2-ethylhexyl (2-ethylhexyl)phosphonate and 570 g of anhydrous ethanol were used to form the coating mixture.

Comparative Example 8

[00242] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 1.31 g of oleic acid and 525 g of anhydrous ethanol were used to form the coating mixture.

Comparative Example 9

[00243] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 1.04 g of melamine polyphosphate and 415 g of anhydrous ethanol were used to form the coating mixture.

Comparative Example 10

[00244] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.58 g of dimethyl methylphosphonate and 230 g of anhydrous ethanol were used to form the coating mixture.

Comparative Example 11

[00245] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.74 g of phenyl phosphonic acid and 295 g of anhydrous ethanol were used to form the coating mixture.

Comparative Example 12

[00246] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.76 g of cyclohexyl phosphonic acid and 305 g of anhydrous ethanol were used to form the coating mixture.

Comparative Example 13

[00247] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.63 g of potassium dihydrogen phosphate and 255 g of anhydrous ethanol were used to form the coating mixture.

Comparative Example 14

[00248] A positive electrode was prepared with the method described in Example 1, except that when preparing the coated cathode active material, 0.98 g of dibutyl phosphate and 390 g of anhydrous ethanol were used to form the coating mixture.

Assembling of Coin Cells of Comparative Examples 1-13

[00249] The coin cells of Comparative Examples 1-13 were assembled in the same

manner as Example 1.

Electrochemical Measurements of Comparative Example 1

[00250] Electrochemical measurements were taken by the same method described in Example 1. The electrochemical performance of the coin cell of Comparative Example 1 was measured and is shown in Table 2 below.

[00251] Furthermore, the amount of degradation of the cathode active material of Comparative Example 1 was also measured.

Electrochemical Measurements of Comparative Examples 2-13

[00252] Electrochemical measurements were taken by the same method described in Example 1. The electrochemical performance of the coin cells of Comparative Examples 2-13 were measured and is shown in Table 2 below.

Electrochemical Measurements of Comparative Example 14

[00253] EIS was conducted on a coin cell of Comparative Example 14 by the same method described in Example 4. The real and imaginary components of the resultant impedance data was then plotted on a Nyquist plot. The Nyquist plot of the coin cell of Comparative Example 14 is shown in Figure 3.

[00254] 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.

Table 1

	Cathode active material	Phosphorus-containing compound		0.5C initial discharging capacity (mAh/g)	Capacity retention after 50 cycles (%)
		R_1*	R_2*		
Example 1	NMC811	C_1	OH	168.6	90.9
Example 2	NMC811	C_4	OH	178.8	91.8
Example 3	NMC811	C_8	OH	176.3	92.1
Example 4	NMC811	C_{18}	OH	170.9	89.1
Example 5	NMC811	C_1	C_2O	168.8	87.6
Example 6	NMC811	C_1	C_1	168.8	86.6
Example 7	LNMO	C_1	OH	115.6	80.3
Example 8	LNMO	C_8	OH	116.9	81.3
Example 9	LNMO	C_{18}	OH	117.1	81.1
Example 10	Core-Shell	C_1	OH	173.2	92.2
Example 11	Core-Shell	C_8	OH	172.9	91.2
Example 12	Core-Shell	C_{18}	OH	172.4	89.0
Example 13	NMC532	C_8	OH	149.6	97.2
Example 14	NMC622	C_8	OH	152.3	92.4
Example 15	NCA	C_8	OH	178.7	93.0

*C_n refers to a straight-chain alkyl group with carbon chain length n. C_nO then refers to a straight-chain alkoxy group with carbon chain length n. OH represents the hydroxyl group.

Table 2

	Cathode active material	Cathode active material coating material	0.5C initial discharging capacity (mAh/g)	Capacity retention after 50 cycles (%)
Comparative Example 1	NMC811	—	155.9	74.3
Comparative Example 2	NMC532	—	133.7	70.2
Comparative Example 3	NMC622	—	141.3	70.8
Comparative Example 4	NCA	—	158.2	71.6
Comparative Example 5	LNMO	—	106.6	68.7
Comparative Example 6	Core-Shell	—	146.0	69.5
Comparative Example 7	NMC811	mono-2-ethylhexyl (2-ethylhexyl)phosphonate	158.5	74.9
Comparative Example 8	NMC811	oleic acid	156.4	72.1
Comparative Example 9	NMC811	melamine polyphosphate	129.2	77.3
Comparative Example 10	NMC811	dimethyl methylphosphonate	159.7	81.8
Comparative Example 11	NMC811	phenyl phosphonic acid	158.1	77.2
Comparative Example 12	NMC811	cyclohexyl phosphonic acid	156.1	76.7
Comparative Example 13	NMC811	potassium dihydrogen phosphate	158.0	78.6