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LITHIUM AND MANGANESE RICH POSITIVE ACTIVE MATERIAL COMPOSITIONS

Abstract

A positive electrode active material may include a compound represented by the formula $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_x\text{N}_y\text{O}_{2-b}\text{F}_b$, wherein M is Co, Cr, or a combination thereof, N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof, $0 \leq a \leq 0.02$, $0 \leq b \leq 0.1$, $0 \leq c \leq 0.1$, $0 \leq x \leq 0.1$, and $0 \leq y \leq 0.04$. A positive electrode may contain the positive electrode active material. A battery may contain the positive electrode.

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Background/Summary

TECHNICAL FIELD

[0001] In at least one aspect, positive electrode active materials for lithium-ion batteries are provided.

BACKGROUND

[0002] Lithium and Manganese Rich (LMR) positive electrode active material has been considered a promising next-generation cathode material due to its high gravimetric energy density compared to currently used Nickel Cobalt Manganese (NCM) and Nickel Cobalt Aluminum (NCA) materials.

SUMMARY

[0003] In at least one aspect, a positive electrode active material is provided. The positive electrode active material includes a compound represented by the formula $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{N}_y\text{O}_{2-b}\text{F}_b$ (1), wherein M is Co, Cr, or a combination thereof, N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof, $0 \leq a \leq 0.02$, $0 \leq b \leq 0.1$, $0 \leq c \leq 0.01$, $0 \leq x \leq 0.1$, and $0 \leq y \leq 0.04$.

[0004] In another aspect, a positive electrode for a lithium-ion battery is provided. The positive electrode includes a positive electrode active material that includes a compound represented by the chemical formula $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{N}_y\text{O}_{2-b}\text{F}_b$, wherein M is Co, Cr, or a combination thereof, N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof, $0 \leq a \leq 0.02$, $0 \leq b \leq 0.1$, $0 \leq c \leq 0.01$, $0 \leq x \leq 0.1$, and $0 \leq y \leq 0.04$.

[0005] In another aspect, a rechargeable lithium-ion battery including at least one lithium-ion battery cell is provided. Each lithium-ion battery cell includes a positive electrode comprising a compound represented by the formula $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{N}_y\text{O}_{2-b}\text{F}_b$, a negative electrode including a negative electrode active material, and an electrolyte, wherein M is Co, Cr, or a combination thereof, N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof, $0 \leq a \leq 0.02$, $0 \leq b \leq 0.1$, $0 \leq c \leq 0.01$, $0 \leq x \leq 0.1$, $0 \leq y \leq 0.04$.

Description

BRIEF DESCRIPTION OF THE DRAWINGS

[0006] For a further understanding of the nature, objects, and advantages of the present disclosure, reference should be made to the following detailed description, read in conjunction with the following drawings, wherein like reference numerals denote like elements and wherein:

[0007] FIG. 1A is a schematic view, in cross-section, of a positive electrode that includes cathode active material on a single side of a current collector;

[0008] FIG. 1B is a schematic view, in cross-section, of a positive electrode that includes cathode active material on both sides of a current collector;

[0009] FIG. 2 is a schematic view, in cross-section, of a battery cell that includes the positive electrode of FIG. 1A; and

[0010] FIG. 3 is a schematic view, in cross-section, of a battery pack that includes the battery cells of FIG. 2.

DETAILED DESCRIPTION

[0011] Reference will now be made in detail to presently preferred compositions, embodiments and methods of the present invention, which constitute the best modes of practicing the invention presently known to the inventors. The figures are not necessarily to scale. However, it is to be understood that the disclosed embodiments are merely exemplary of the invention that may be embodied in various and alternative forms. Therefore, specific details disclosed herein are not to be interpreted as limiting, but merely as a representative basis for any aspect of the invention and/or as

a representative basis for teaching one skilled in the art to variously employ the present invention. [0012] Except in the examples, or where otherwise expressly indicated, all numerical quantities in this description indicating amounts of material or conditions of reaction and/or use are to be understood as modified by the word “about” in describing the broadest scope of the invention. Practice within the numerical limits stated is generally preferred. Also, unless expressly stated to the contrary, when a given chemical structure includes a substituent on a chemical moiety (e.g., on an aryl, alkyl, etc.) that substituent is imputed to a more general chemical structure encompassing the given structure; percent, “parts of,” and ratio values are by weight; the term “polymer” includes “oligomer,” “copolymer,” “terpolymer,” and the like; molecular weights provided for any polymers refer to weight average molecular weight unless otherwise indicated; the description of a group or class of materials as suitable or preferred for a given purpose in connection with the invention implies that mixtures of any two or more of the members of the group or class are equally suitable or preferred; description of constituents in chemical terms refers to the constituents at the time of addition to any combination specified in the description, and does not necessarily preclude chemical interactions among the constituents of a mixture once mixed; the first definition of an acronym or other abbreviation applies to all subsequent uses herein of the same abbreviation and applies mutatis mutandis to normal grammatical variations of the initially defined abbreviation; and, unless expressly stated to the contrary, measurement of a property is determined by the same technique as previously or later referenced for the same property.

[0013] It should be noted that, as used in the specification and the appended claims, the singular form “a,” “an,” and “the” comprise plural referents unless the context clearly indicates otherwise. For example, reference to a component in the singular is intended to comprise a plurality of components.

[0014] As used herein, the term “about” means that the amount or value in question may be the specific value designated or some other value in its neighborhood. Generally, the term “about” denoting a certain value is intended to denote a range within +1-5% of the value. As one example, the phrase “about 100” denotes a range of 100 ± 5 , i.e. the range from 95 to 105. Generally, when the term “about” is used, it can be expected that similar results or effects according to the invention can be obtained within a range of +1-5% of the indicated value. As used herein, the term “and/or” means that either all or only one of the elements of said group may be present. For example, “A and/or B” shall mean “only A, or only B, or both A and B.” In the case of “only A,” the term also covers the possibility that B is absent, i.e. “only A, but not B.”

[0015] It is also to be understood that this invention is not limited to the specific embodiments and methods described below, as specific components and/or conditions may, of course, vary. Furthermore, the terminology used herein is used only for the purpose of describing particular embodiments of the present invention and is not intended to be limiting in any way.

[0016] The term “comprising” is synonymous with “including,” “having,” “containing,” or “characterized by.” These terms are inclusive and open-ended and do not exclude additional, unrecited elements or method steps. The phrase “consisting of” excludes any element, step, or ingredient not specified in the claim. When this phrase appears in a clause of the body of a claim, rather than immediately following the preamble, it limits only the element set forth in that clause; other elements are not excluded from the claim as a whole. The phrase “consisting essentially of” limits the scope of a claim to the specified materials or steps, plus those that do not materially affect the basic and novel characteristic(s) of the claimed subject matter. The phrase “composed of” means “including” or “consisting of.” Typically, this phrase is used to denote that an object is formed from a material. With respect to the terms “comprising,” “consisting of,” and “consisting essentially of,” where one of these three terms is used herein, the presently disclosed and claimed subject matter can include the use of either of the other two terms. The term “one or more” means “at least one” and the term “at least one” means “one or more.” The terms “one or more” and “at least one” include “plurality” and “multiple” as a subset. In a refinement, “one or more” includes

“two or more.” The term “substantially,” “generally,” or “about” may be used herein to describe disclosed or claimed embodiments. The term “substantially” may modify a value or relative characteristic disclosed or claimed in the present disclosure. In such instances, “substantially” may signify that the value or relative characteristic it modifies is within $\pm 0\%$, 0.1% , 0.5% , 1% , 2% , 3% , 4% , 5% or 10% of the value or relative characteristic.

[0017] It should also be appreciated that integer ranges explicitly include all intervening integers. For example, the integer range 1-10 explicitly includes 1, 2, 3, 4, 5, 6, 7, 8, 9, and 10. Similarly, the range 1 to 100 includes 1, 2, 3, 4 . . . 97, 98, 99, 100. Similarly, when any range is called for, intervening numbers that are increments of the difference between the upper limit and the lower limit divided by 10 can be taken as alternative upper or lower limits. For example, if the range is 1.1. to 2.1, the following numbers 1.2, 1.3, 1.4, 1.5, 1.6, 1.7, 1.8, 1.9, and 2.0 can be selected as lower or upper limits.

[0018] When referring to a numeral quantity, in a refinement, the term “less than” includes a lower non-included limit that is 5 percent of the number indicated after “less than.” For example, “less than 20” includes a lower non-included limit of 1. Therefore, this refinement of “less than 20” includes a range between 1 and 20. In another refinement, the term “less than” includes a lower non-included limit that is, in increasing order of preference, 20 percent, 10 percent, 5 percent, or 1 percent of the number indicated after “less than.”

[0019] The term “positive electrode” means a battery cell electrode from which current flows out when the lithium-ion battery cell or battery is discharged. Sometimes a “positive electrode” is referred to as a “cathode.” The term “negative electrode” means a battery cell electrode to which current flows in when the lithium-ion battery cell is discharged. Sometimes a “negative electrode” is referred to as an “anode.” The term “cell” or “battery cell” means an electrochemical cell made of at least one positive electrode, at least one negative electrode, an electrolyte, and a separator membrane. The term “battery” or “battery pack” means an electric storage device made of at least one battery cell. In a refinement, “battery” or “battery pack” is an electric storage device made of a plurality of battery cells. The term “specific capacity” means the capacity per unit mass of the anode active. Specific capacity has units of milliamp hours/gram (mAh/g).

[0020] There are intrinsic issues with current LMR material compositions such as voltage decay during cycling, rate capability, cycle performance, and volumetric energy density. Accordingly, there is a need for optimized LMR material compositions for positive electrode active materials for lithium-ion batteries with increased rate capability, cell performance, and volumetric energy density.

[0021] The present disclosure provides compositions for Lithium Manganese Rich (LMR) cathodes used in lithium-ion batteries. The performance issues inherent to conventional LMR compositions may include voltage decay during cycling, decreased rate capability over successive cycles, and poor cycle performance. These issues can be mitigated by optimizing the composition of the LMR.

[0022] In one embodiment of this disclosure, the LMR composition is optimized to have lower Li content (Li.sub.2MnO.sub.3) than conventional compositions. This modification better the cycle performance, power performance, and rate capability by enhancing the voltage decay, and electronic and ionic conductivity of the LMR. However, it is recognized that lower Li content typically results in lower capacity than conventional LMR.

[0023] To address this potential reduction in capacity, the LMR composition is further modified to incorporate increased Nickel (Ni) with optimized Cobalt (Co) or Chromium (Cr) content, with Cobalt being the primary focus. This modified composition series increases capacity from Ni and Co by controlling the average oxidation state of the Ni ion. Optimized Co content also assists in enhancing electronic and ionic conductivity.

[0024] In one embodiment, the general formula of the new LMR composition is given as $\text{Li.sub.(1.1+a)Mn.sub.(0.51+c)Ni.sub.(0.38-x)M.sub.x-yN.sub.yO.sub.(2-b)F.sub.b}$, where M can be Co, Cr, or a combination thereof, and N is W.sup.+6 , Ta.sup.+5 , V.sup.+5 or a combination thereof. The

average oxidation state of the Ni ion is controlled to be 2.0. The average oxidation state of the Mn ion is controlled between 3.7 and 4.0 by controlling the N(Hexa and Pentavalent cation) and F (Fluorine) doping content, where “y” is again a value between 0 and 0.04, where “b” is again a value between 0 and 0.1.

[0025] In the LMR composition $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{N}_y\text{O}_{2-b}\text{F}_b$ F doping $\text{Li}_{1.1}$ in the formula represents an optimized lithium content that promotes cycling, mitigates voltage decay, and bolsters power performance. The configuration also allows for the attainment of closer to maximum actual capacity by manipulating the amounts of Ni^{2+} and Co^{3+} ions within the composition.

[0026] Optimal Co^{3+} content within this formula also serves to enhance electronic conductivity and power capability. In this composition, the Co^{3+} content is further restricted to between 0 and 0.06, a constraint that helps balance performance metrics such as capacity and power against production. This formula also facilitates a larger slope region at 1st charge, achieved through the heightened incorporation of Mn^{3+} . This is made possible by Hexa and Pentavalent cation doping and F⁻ doping, which effectively lowers the oxidation state of Mn, resulting in an increase in the Mn^{3+} ion presence.

[0027] Higher Initial Coulombic Efficiency (ICE), leading to increased discharge capacity, is another notable advantage. This increased ICE is tied to better structural stability after the initial charge, which can be attributed to strong Metal-Fluoride (Me-F) bonding in the composition. Additionally, the elevated Mn^{3+} content further augments electronic conductivity. With respect to Mn^{3+} content and ICE, this adaptation shows enhancements compared to compositions previously known.

[0028] In another embodiment the general formula of the new LMR composition is represented as $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{N}_y\text{O}_{2-b}\text{F}_b$, where M can be Co or Cr or a combination thereof, and N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof. Here, “x” is a variable value between 0 and 0.1, defining the proportion of M in the composition. The average oxidation state of the Ni ion is controlled to be 2, while the average oxidation state of the Manganese (Mn) ion is controlled between 3.7 and 4.0 by controlling the Hexa and Pentavalent cation doping and fluorine (F) doping content, “b,” which lies between 0 and 0.1.

[0029] The LMR composition $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{N}_y\text{O}_{2-b}\text{F}_b$, coupled with F⁻ doping, has several advantages. $\text{Li}_{1.1}$ is found to be an optimized Li content for achieving good cycle, voltage decay, and power performance. Maximum actual capacity is realized in the $\text{Li}_{1.1}$ composition by controlling the contents of Ni^{3+} and Co^{3+} . Furthermore, optimized Co^{3+} content betters electronic conductivity, leading to better power capability.

[0030] Restricted Co^{3+} content, between 0 and 0.1, is advantageous in controlling performance meaning capacity and power. A larger slope region is at 1st charge obtainable with a higher incorporation of Mn^{3+} as Hexa and Pentavalent cation doping and F⁻ doping can lower the Mn oxidation state. Higher Initial Coulombic Efficiency (ICE) can lead to higher discharge capacity, owing to better structural stability after the first charge due to strong Metal-Fluoride (Me-F) bonding. Moreover, higher Mn^{3+} enhances electronic conductivity. Higher Mn^{3+} content and higher ICE results in an overall increase in performance.

[0031] Referring to FIG. 1, a schematic of a positive electrode that includes a positive electrode active material is provided. Positive electrode **10** includes positive electrode active material layer **12** of positive electrode active material disposed over and typically contacting positive electrode current collector **14**. Typically, the positive electrode current collector **14** is a metal plate or metal foil composed of a metal such as aluminum, copper, platinum, zinc, titanium, and the like. Currently, aluminum is most commonly used for the positive electrode current collector. The positive electrode active material is represented by formula 1:

$\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{O}_{2-b}\text{F}_b$ (1) [0032]
wherein: [0033] M is Co, Cr, or a combination thereof; [0034] N is W^{+6} , Ta^{+5} , V^{+5} or combination of there of; [0035] $0 \leq a \leq 0.02$; [0036] $0 \leq b \leq 0.01$; [0037] $0 \leq c \leq 0.1$; [0038] $0 \leq x \leq 0.1$; and [0039] $0 \leq y \leq 0.04$.

[0040] A specific active electrode composition can be $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{O}_{2-b}\text{F}_b$, where M can be Co, Cr, or a combination thereof, and N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof. The average oxidation state of the Ni ion is controlled to be 2. The average oxidation state of the Mn ion is controlled between 3.7 and 4.0 by controlling the HExa and Pentavalent cation doping and F doping content, where “b” is again a value between 0 and 0.1.

[0041] Another LMR composition is represented as $\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_{x-y}\text{O}_{2-b}\text{F}_b$, where M can be Co or Cr or a combination thereof and N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof, and N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof. Here, “x” is a variable value between 0 and 0.1, defining the proportion of M in the composition. The average oxidation state of the Ni ion is controlled to be 2.0, while the average oxidation state of the Manganese (Mn) ion is controlled between 3.7 and 4.0 by controlling the Hexa and Pentavalent cation doping and fluorine (F) doping content, “b,” which lies between 0 and 0.1.

[0042] With reference to FIG. 2, a schematic of a rechargeable lithium-ion battery cell incorporating the positive electrode of FIG. 1 is provided. Battery cell 20 includes the positive electrode 10 as described above, negative electrode 22, and separator 24 interposed between the positive electrode and the negative electrode. The negative electrode 22 includes a negative electrode current collector 26 and a negative active material layer 28 disposed over and typically contacting the negative electrode current collector 26. Typically, the negative electrode current collector 26 is a metal plate or metal foil composed of a metal such as aluminum, copper, platinum, zinc, titanium, and the like. Currently, copper is most commonly used for the negative electrode current collector 26. The battery cell 20 is immersed in electrolyte 30 which is enclosed by battery cell case 32. The electrolyte 30 imbibes into the separator 24. In other words, the separator 24 includes the electrolyte thereby allowing lithium ions to move between the negative and positive electrodes. The electrolyte 30 includes a non-aqueous organic solvent and a lithium salt. The non-aqueous organic solvent serves as a medium for transmitting ions taking part in the electrochemical reaction of a battery. Advantageously, the battery cell 20 can have a specific capacity of greater than 150 mAh/g.

[0043] With reference to FIG. 3, a schematic of a rechargeable lithium-ion battery incorporating the positive electrode of FIG. 1 and the battery cells of FIG. 2 is provided. Rechargeable lithium-ion battery 40 includes at least one battery cell of the design in FIG. 2. Typically, the rechargeable lithium-ion battery 40 includes at least one battery cell 20 of the design of FIG. 2. Each of the lithium-ion battery cells 20' includes the positive electrode 10 which includes the compound represented by formula 1, the negative electrode 22 which includes a negative active material, and the electrolyte 30, where i is an integer label for each battery cell. The label i runs from 1 to nmax, where nmax is the total number of battery cells in the rechargeable lithium-ion battery 40. The electrolyte 30 includes a non-aqueous organic solvent and a lithium salt. The non-aqueous organic solvent serves as a medium for transmitting ions taking part in the electrochemical reaction of a battery. The plurality of battery cells can be wired in series, in parallel, or a combination thereof. The voltage output from the battery 40 is provided across terminals 42 and 44. Advantageously, the rechargeable lithium-ion battery 40 can have a specific capacity of greater than 150 mAh/g for each battery cell therein.

[0044] Referring to FIGS. 2 and 3, the separator 24 physically separates the negative electrode 22 from the positive electrode 10 thereby preventing shorting while allowing the transport of lithium

ions for charging and discharging. Therefore, the separator **24** can be composed of any material suitable for this purpose. Examples of suitable materials from which the separator **24** can be composed include but are not limited to, polytetrafluoroethylene (e.g., TEFLON®), glass fiber, polyester, polyethylene, polypropylene, and combinations thereof. The separator **24** can be in the form of either a woven or non-woven fabric. The separator **24** can be in the form of a non-woven fabric or a woven fabric. For example, a polyolefin-based polymer separator such as polyethylene and/or polypropylene is typically used for a lithium-ion battery. In order to ensure heat resistance or mechanical strength, a coated separator includes a coating of ceramic or a polymer material may be used.

[0045] Referring to FIGS. **2** and **3**, the electrolyte **30** includes a lithium salt dissolved in the non-aqueous organic solvent. Therefore, the electrolyte **30** includes lithium ions that can intercalate into the positive electrode active material during discharging and into the anode active material during charging. Examples of lithium salts include but are not limited to LiPF_6 , LiBF_4 , LiSbF_6 , LiAsF_6 , $\text{LiC}_4\text{F}_9\text{SO}_3$, LiClO_4 , LiAlO_2 , LiAlCl_4 , LiCl , LiI , $\text{LiB}(\text{C}_2\text{O}_4)_2$, and combinations thereof. In a refinement, the electrolyte includes the lithium salt in an amount from about 0.1 M to about 2.0 M.

[0046] Still referring to FIGS. **2** and **3**, the electrolyte includes a non-aqueous organic solvent and a lithium salt. Advantageously, the non-aqueous organic solvent serves as a medium for transmitting ions, and in particular, lithium ions participate in the electrochemical reaction of a battery. Suitable non-aqueous organic solvents include carbonate-based solvents, ester-based solvents, ether-based solvents, ketone-based solvents, alcohol-based solvents, aprotic solvents, and combinations thereof. Examples of carbonate-based solvents include but are not limited to dimethyl carbonate, diethyl carbonate, dipropyl carbonate, methylpropyl carbonate, ethylpropyl carbonate, methylethyl carbonate, ethylene carbonate, propylene carbonate, butylene carbonate, and combinations thereof. Examples of ester-based solvents include but are not limited to methyl acetate, ethyl acetate, n-propyl acetate, methylpropionate, ethylpropionate, γ -butyrolactone, decanolide, valerolactone, mevalonolactone, caprolactone, and combinations thereof. Examples of ether-based solvents include but are not limited to dibutyl ether, tetraglyme, diglyme, dimethoxyethane, 2-methyltetrahydrofuran, tetrahydrofuran, and the like, and the ketone-based solvent may include cyclohexanone, and the like. Examples of alcohol-based solvents include but are not limited to methanol, ethyl alcohol, n-propyl alcohol, isopropyl alcohol, and the like. Examples of the aprotic solvent include but are not limited to nitriles such as $\text{R}-\text{CN}$ (where R is a C_{2-20} linear, branched, or cyclic hydrocarbon that may include a double bond, an aromatic ring, or an ether bond), amides such as dimethylformamide, dioxolanes such as 1,3-dioxolane, sulfolanes, and the like. Advantageously, the non-aqueous organic solvent can be used singularly. In other variations, mixtures of the non-aqueous organic solvent can be used. Such mixtures are typically formulated to optimize battery performance. In a refinement, a carbonate-based solvent is prepared by mixing a cyclic carbonate and a linear carbonate. In a variation, the electrolyte **30** can further include vinylene carbonate or an ethylene carbonate-based compound to increase battery cycle life.

[0047] Referring to FIGS. **1**, **2**, and **3**, the negative electrode and the positive electrode can be fabricated by methods known to those skilled in the art of lithium-ion batteries. Typically, an active material (e.g., the positive or negative active material) is mixed with a conductive material, and a binder in a solvent (e.g., N-methylpyrrolidone) is mixed into an active material composition and coated on the current collector. The electrode manufacturing method is well known and thus is not described in detail in the present specification. The solvent includes N-methylpyrrolidone and the like but is not limited thereto.

[0048] Referring to FIGS. **1**, **2**, and **3**, the positive electrode active material layer **12** includes the positive electrode active material represented by formula 1, a binder, and a conductive material. The binder can increase the binding properties of positive electrode active material particles with one another and with the positive electrode current collector **14**. Examples of suitable binders

include but are not limited to polyvinyl alcohol, carboxylmethyl cellulose, hydroxypropyl cellulose, diacetyl cellulose, polyvinylchloride, carboxylated polyvinylchloride, polyvinylfluoride, an ethylene oxide-containing polymer, polyvinylpyrrolidone, polyurethane, polytetrafluoroethylene, polyvinylidene fluoride, polyethylene, polypropylene, a styrene-butadiene rubber, an acrylate styrene-butadiene rubber, an epoxy resin, nylon, and the like, and combinations thereof. The conductive material provides the positive electrode **10** with electrical conductivity. Examples of suitable electrically conductive materials include but are not limited to natural graphite, artificial graphite, carbon black, acetylene black, ketjen black, carbon fibers, copper, metal powders, metal fibers, and combinations thereof. Examples of metal powders and metal fibers are composed of including nickel, aluminum, silver, and the like.

[0049] Referring to FIGS. **1**, **2**, and **3**, the negative active material layer **26** includes a negative active material, a binder, and optionally a conductive material. The negative active materials used herein can be those negative materials known to one skilled in the art of lithium-ion batteries. Negative active materials include but are not limited to, carbon-based negative active materials, silicon-based negative active materials, and combinations thereof. A suitable carbon-based negative active material may include graphite and graphene. A suitable silicon-based negative active material may include at least one selected from silicon, silicon oxide, silicon oxide coated with conductive carbon on the surface, and silicon (Si) coated with conductive carbon on the surface. For example, silicon oxide can be described by the formula SiO_z where z is from 0.09 to 1.1. Mixtures of carbon-based negative active materials or silicon-based negative active materials can also be used for the negative active material.

[0050] The negative electrode binder increases the binding properties of negative active material particles with one another and with a current collector. The binder can be a non-aqueous binder, an aqueous binder, or a combination thereof. Examples of non-aqueous binders may be polyvinylchloride, carboxylated polyvinylchloride, polyvinylfluoride, an ethylene oxide-containing polymer, polyvinylpyrrolidone, polyurethane, polytetrafluoroethylene, polyvinylidene fluoride, polyethylene, polypropylene, polyamideimide, polyimide, or a combination thereof. Aqueous binders can be rubber-based binders or polymer resin binders. Examples of rubber-based binders include but are not limited to styrene-butadiene rubbers, acrylated styrene-butadiene rubbers, acrylonitrile-butadiene rubbers, acrylic rubbers, butyl rubbers, fluorine rubbers, and combinations thereof. Examples of polymer resin binders include but are not limited to polyethylene, polypropylene, ethylenepropylene copolymer, polyethyleneoxide, polyvinylpyrrolidone, epichlorohydrin, polyphosphazene, polyacrylonitrile, polystyrene, ethylenepropylenediene copolymer, polyvinylpyridine, chlorosulfonated polyethylene, latex, a polyester resin, an acrylic resin, a phenolic resin, an epoxy resin, polyvinyl alcohol, and combinations thereof.

[0051] While exemplary embodiments are described above, it is not intended that these embodiments describe all possible forms of the invention. Rather, the words used in the specification are words of description rather than limitation, and it is understood that various changes may be made without departing from the spirit and scope of the invention. Additionally, the features of various implementing embodiments may be combined to form further embodiments of the invention.

Claims

1. A positive electrode active material for lithium-ion batteries, comprising a compound represented by the general formula 1:

$\text{Li}_{(1.1+a)}\text{Mn}_{(0.51+c)}\text{Ni}_{(0.38-x)}\text{M}_{x-y}\text{O}_{(2-b)}\text{F}_b$ (1) wherein: M is Co, Cr, or a combination thereof; N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof; $0 \leq a \leq 0.02$; $0 \leq c \leq 0.01$; $0 \leq x \leq 0.1$; $0 \leq y \leq 0.04$; and $0 \leq b \leq 0.1$.

2. The positive electrode active material of claim 1, wherein $a=0$ and $c=0.01$.

3. The positive electrode active material of claim 1, wherein $a=0.1$ and $c=0$.
4. The positive electrode active material of claim 1, wherein the average oxidation state of Mn is controlled between 3.7 and 4.0.
5. A positive electrode for a lithium-ion battery, the positive electrode comprising a positive electrode active material comprising a compound represented by chemical formula 1:
$$\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_x\text{N}_y\text{O}_{2-b}\text{F}_b \quad (1)$$
 wherein: M is Co, Cr, or a combination thereof; N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof; $0 \leq a \leq 0.02$; $0 \leq c \leq 0.01$; $0 \leq x \leq 0.1$; $0 \leq y \leq 0.04$; and $0 \leq b \leq 0.1$.
6. The positive electrode of claim 5, wherein $a=0$ and $c=0.01$.
7. The positive electrode of claim 5, wherein $a=0.1$ and $c=0$.
8. The positive electrode claim 5, wherein the average oxidation state of Mn is between 3.7 and 4.0.
9. A rechargeable lithium-ion battery comprising at least one lithium-ion battery cell, each lithium-ion battery cell including a positive electrode comprising a positive electrode active material as represented by formula 1.
10. A rechargeable lithium-ion battery comprising at least one lithium-ion battery cell, each lithium-ion battery cell including: a positive electrode comprising a positive electrode active material as represented by formula 1:
$$\text{Li}_{1.1+a}\text{Mn}_{0.51+c}\text{Ni}_{0.38-x}\text{M}_x\text{N}_y\text{O}_{2-b}\text{F}_b \quad (1)$$
 wherein: M is Co, Cr, or a combination thereof; N is W^{+6} , Ta^{+5} , V^{+5} or a combination thereof; $0 \leq a \leq 0.02$; $0 \leq c \leq 0.01$; $0 \leq x \leq 0.1$; $0 \leq y \leq 0.04$; $0 \leq b \leq 0.1$; a negative electrode including a negative active material; and an electrolyte.
11. The rechargeable lithium-ion battery of claim 10, wherein the positive electrode active material $a=0$ and $c=0.01$.
12. The rechargeable lithium-ion battery of claim 10, wherein the positive electrode active material $a=0.1$ and $c=0$.
13. The rechargeable lithium-ion battery of claim 10, wherein the positive electrode active material the average oxidation state of Mn is between 3.7 and 4.0.
14. The rechargeable lithium-ion battery of claim 10, wherein the at least one lithium-ion battery cell is a plurality of battery cells.
15. The rechargeable lithium-ion battery of claim 10, wherein each battery cell further includes a separator interposed between the positive electrode and the negative electrode.
16. The rechargeable lithium-ion battery of claim 10, wherein each battery cell has a specific capacity greater than 150 mAh/g.
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