Claims

1. A method for preparing an electrolyte-filled high charge per unit mass electrode for a high energy-density battery comprising two current collectors separated by an electrolyte composition, a separator and either:

a. two electrodes (an anode, a cathode) physically and electrically contacting said two current collectors, or

b. a cathode in contact only with a current collector respectively, the second current collector being in contact with said separator;

said method comprising the steps of:

i. preparing a mixture A comprising said electrolyte by mixing a metal salt with a solvent;

ii. mixing said mixture A with an active material to obtain a paste; and

iii. forming said electrode with a predetermined thickness.

2. The method according to claim 1, wherein said metal salt comprises (i) a cation selected from lithium, sodium, potassium, calcium, magnesium and zinc, and (ii) an anion selected from hexafluorophosphate (PF6), tetrafluoroborate (BF4), bis(trifluoromethanesulfonyl)imide (TFSI), bis(fluorosulfonyl)imide (FSI), dicyanamide (DCA), 4,5-dicyano-2-(trifluoromethyl)imidazolide (TDI), fluorosulfonyl-(trifluoromethanesulfonyl)imide (FTFSI), (difluoroethanesulfonyl)imide (DFTFSI), bis(oxalato)borate (BOB), difluoro(oxalato)borate (DFOB).

3. The method according to claim 1 or 2, wherein said solvent is selected from an aprotic organic solvent, a protic organic solvent, or a mixture thereof.

4. The method according to claim 3, wherein said aprotic organic solvent is selected from an ionic liquid, propylene carbonate, glyme, a salt concentrated in aqueous systems in solution.

5. The method according to claim 4, wherein said ionic liquid comprises (i) a cation selected from alkylimidazolium, or based on alkylpyrrolidinium, morpholinium, pyridinium, piperidinium, phosphonium, ammonium and (ii) an anion selected from hexafluorophosphate (PF6), tetrafluoroborate (BF4),

bis(trifluoromethanesulfonyl)imide (TFSI), bis(fluorosulfonyl)imide (FSI), dicyanamide (DCA), 4,5-dicyano-2-(trifluoromethyl)imidazolide (TDI), fluorosulfonyl-(trifluoromethanesulfonyl)imide (FTFSI), (difluoroethanesulfonyl)imide (DFTFSI), bis(oxalato)borate (BOB), difluoro(oxalato)borate (DFOB).

6. The method according to any one preceding claim wherein a binder is added to the two electrodes or a binder is added to the cathode.

7. The method according to claim 6, wherein said binder is selected from styrene butadiene latex copolymer (SBR), polyacrylonitrile (PAN), polyvinylidene fluoride (PVDF), polyvinylidene fluoride-co-hexafluoropropylene (PVdF-HFP), polyvinylidene fluoride-co-trichlorethylene, polymethyl methacrylate (PMMA), polyvinylpyrrolidone, polyvinyl acetate, polyethylene-co-vinyl acetate, polyethylene oxide, cellulose acetate, cellulose acetate butyrate, cellulose acetate priopionate, cyanoethyl pullulan, cyanoethyl polyvinyl alcohol, cyanoethyl cellulose, cyanoethyl saccharose, pullulan and carboxymethyl cellulose (CMC), polytetrafluoroethylene (PTFE) or a combination of at least two of them, as well as among polymers and their derivatives and/or composites such as polyaniline (PANI), poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) composites, polyaniline-polyacrylic polymer composites (PANI:PAA) containing a conducting/carboxyl polymer, polypyrrole-carboxymethycellulose PPy/CMC, a hydrogel-based polymer such as (2-acrylamido-2-methyl-1-propanesufonic acid-co-acrylonitrile) (PMAPS), an ionic liquid polymer.

8. The method from any one preceding claim, wherein the material for the cathode is selected from

a. for a Li-ion battery: a lithium intercalating compound, selected from lithium iron phosphate, ($LiFePO_4$), lithium nickel-manganese-cobalt oxide, ($LiNi_xMn_yCo_zO_2$), doped lithium nickel-manganese-cobalt oxide ($LiNi_xMn_yCo_zO_2$), lithium cobalt oxide ($LiCoO_2$), doped lithium-cobalt oxide, lithium nickel oxide ($LiNiO_2$), doped lithium nickel oxide, lithium manganese oxide ($LiMn_2O_4$), doped lithium manganese oxide, lithium vanadium oxide, doped lithium vanadium oxide, lithium mixed metal oxides (LMNO), mixed transition metal oxides, mixed transition metals, doped lithium transition metal oxides, lithium vanadium phosphate, lithium manganese phosphate, lithium cobalt

phosphate, mixed lithium and metal phosphates, metal sulfides, and combinations thereof.

b. for Na-ion and K-ion batteries:

i. a metal oxide;

ii. layered NaMOX;

iii. one-dimensional tunnel oxides;

iv. fluorides;

v. sulfates;

vi. phosphates;

vii. pyrophosphates;

viii. fluorophosphates;

ix. mixed phosphates;

x. hexacyanometalates;

xi. cathodes without critical metal;

xii. Prussian white analogs

c. for Zn-ion and Mg-ion batteries:

i. transition metal oxides, MxV2O5 (M= Na, Ca, Zn, Mg, Ag, Li, etc.);

ii. a vanadate;

iii. layered and tunnel type vanadium-based compounds;

iv. polyanionic materials analogous to Prussian blue;

v. metal disulfides,

vi. NASICON compounds

vii. AxMM0 $(XO_4)_3$ (A: Li, Na, Mg, Zn, etc.; M: Mn, Ti, Fe, etc.; X:P, Si, S, etc.);

viii. organic materials such as quinones

d. for Mg-ion battery: layered sulfide/selenide

e. for Ca-ion battery:

i. 3D tunnel structures such as $CaMn_2O_4$ spinel,

ii. chevrel phases such as $CaMo_6X_8$ (X = S, Se, Te),

iii. layered transition metal oxides,

iv. Prussian blue analogs.

9. The method from any one preceding claim, wherein the material for the anode is an active material selected from

a. for a Li-ion battery:

i. a lithium-containing titanium composite oxide (LTO);

ii. metals (Me) such as Si, Sn, Li, Zn, Mg, Cd, Ce, Ni and Fe;

iii graphite, graphene, including particles of natural graphite, artificial graphite, meso-carbon microbeads (MCMB) and carbon (including soft carbon, hard carbon, carbon nanofibers and carbon nanotubes;

iv. silicon (Si), silicon/graphite composites, silicon combinations of germanium (Ge), tin (Sn), lead (Pb), antimony (Sb), bismuth (Bi), zinc (Zn), aluminum (Al), nickel (Ni), cobalt (Co), manganese (Mn), titanium (Ti), iron (Fe), and cadmium (Cd);

v. intermetallic alloys or compounds of Si, Ge, Sn, Pb, Sb, Bi, Zn, Al or Cd with other elements, said alloys or compounds being stoichiometric or non-stoichiometric;

vi. oxides, carbides, nitrides, sulfides, phosphides, selenides, and tellurides of Si, Ge, Sn, Pb, Sb, Bi, Zn, Al, Fe, Ni, Co, Ti, Mn or Cd, and mixtures or composites thereof;

vii. oxides (MeOx) of the metals (Me);

viii. and composites of metals (Me) with carbon;

ix. MXene materials, [M_XC where X = 2, 3, 4).

b. for Na-ion and K-ion batteries:

i. oxide, sulfides, selenides, phosphides and MOF-based materials and carbon-based materials.

ii. the carbon-based materials comprise expanded graphite, N-doped expanded graphite, carbon black, amorphous carbon, carbon microspheres, hard carbon, meso-powerful soft carbon, carbon nanotubes, graphene nanosheets, nitrogen-doped CNTs, N-doped graphene foam, N-doped porous nanofibers, microporous carbon and cube-shaped porous carbon.

iii. the oxides comprise MnO_{2-} nanoflowers, NiO nanosheets, porous SnO, porous SnO_2 nanotubes, porous 3D Fe_3O_4–C, porous CuO-RGO, MnO-CNT doped with ultrasmall nitrogen, CuS microflowers, SnS_2-RGO, Co_3S_4-PANI, ZnS-RGO, NiS-RGO, Co_3S_4-PANI, MoS_2-C, conductive WS2-carbon nanosheets doped with nitrogen, Sb_3Se_3-RGO nanorods, $MoSe_2$-carbon fiber, the multishell Sn_4P_3 nanostructures, Sn_4P_3–C nanospheres, Se_4P_4, CoP nanoparticles, FeP nanorods matrices on carbon cloth, MoP-C, CUP_2 -C, hollow NiO/Ni graphene, CoSe/C structured in shell-yellow doped with nitrogen

iv. Na metal

c. for Mg-ion and Zn-ion batteries: graphite, polynano crystalline graphite, expanded graphite, hard carbon/carbon black, hard-soft composite carbon, hard carbon microspheres, activated charcoal, a multi-layer F-doped graphene, nitrogen-doped carbon microsphere, hierarchically porous N-doped carbon, phosphorous and oxygen dual-doped graphene, nitrogen- and oxygen-doped carbon nanofiber, tire rubber derived hard carbon, porous carbon nanofiber paper, polycristalline soft carbon, nitrogen-doped natural carbon nanofibers, nitrogen/oxygen double-doped hard carbon, $K_2Ti_4O_9$ and $K_2Ti_4O_9$.

d. for Zn-ion batteries:

i. zinc metal, zinc alloys

ii. graphite and carbonaceous materials

e. for Ca-ion battery:

i. calcium-metal alloys

ii. tin metal

iii. graphite and carbonaceous materials.

10. The method according to any one of the preceding claims, wherein a conducting material is added to the cathode before the mixing of step (ii).

11. A method according to any one of the preceding claims, wherein said electrolyte comprises an additive.

12. The method according to claim 10, wherein said conducting material is selected from carbon black consisting of acetylene black, carbon black, ketjen black, canal black, furnace black, lamp black or thermal black; graphite, such as natural graphite or artificial graphite and a mixture thereof, or a combination of at least two of them; a conducting material comprising conducting fibers, such as carbon fibers or metal fibers; a metal powder, such as a fluorocarbon, aluminum or nickel powder; conducting metal monocrystalline filaments, such as zinc oxide, or potassium titanate; titanium dioxide; a polyphenylene derivative.

13. The method according to any one preceding claim, wherein step iii of forming the electrode is selected from the following techniques: the paste rolling technique, the 3D printing technique for paste, an extrusion technique and a jet milling technique.

14. The method from any one preceding claim, wherein the percentage by mass of electrolyte: active material is comprised in the range [15: 85] and preferably in the range [30: 75], or even more preferably in the range [40: 60].

15. An electrolyte-filled high charge per unit mass electrode for a high energy-density battery obtained by the method defined in any one of claims 1 to 13.

16. A high energy-density battery comprising at least one electrolyte-filled electrode prepared in accordance with the method defined in any one of claims 1 to 13, a separator and two current collectors, wherein:

a. when the battery comprises two electrodes, said current collectors are respectively connected to said electrodes (i.e. cathode, anode), and said electrodes consist of:

i. an anode electrode prepared in accordance with the method defined in any one of claims 1 to 13, and a cathode electrode, or

ii. a cathode electrode prepared in accordance with the method defined in any one of claims 1 to 13, and an anode electrode, or

iii. a cathode electrode and an anode electrode both prepared in accordance with the method defined in any one of claims 1 to 13, or

b. when the battery comprises a cathode electrode only, said current collectors are respectively connected to said cathode and to the separator, where said cathode electrode is prepared in accordance with the method according to one of claims 1 to 13.

17. A high energy-density battery according to claim 16 characterized in that the cathode electrolyte is different from the anode electrolyte.