Recently, there has been increasing research interest in 3D carbon-based free-standing electrodes as they offer higher energy density and are resistant to corrosion. Unlike traditional electrodes, carbon-based free-standing electrodes are superior alternatives because they are light and metal-free, leading to higher battery energy density and better cycling stability. Free-standing electrodes include graphene foams/sponges, carbon cloth/carbon fibers, carbon nanotube paper/arrays/network, an the like. While these electrodes have good electrochemical performance, their conventional preparation methods involve specialized equipment and laborious methods, such as chemical vapor deposition (CVD), hydrothermal synthesis, ultrasonication, filtration, which are costly to implement for large-scale production.

Another method of preparing free-standing electrodes includes slurry coating on a single type of electrode of either lithium cobalt oxide, lithium titanium oxide or lithium iron phosphate (LFP), which is highly limited.

Accordingly, there is a need for a method of making a free-standing electrode that ameliorates one or more disadvantages mentioned above.

Summary

There is provided a hydrogel binder comprising an anionic polyacid, a cationic polyamine and a solvent, wherein the anionic polyacid and the cationic polyamine are derived from an acid-base reaction between a polyacid and a polyamine at a weight ratio in the range of 10:100 to 40:100.

In one aspect, there is provided a hydrogel binder comprising an anionic polyacid, a cationic polyamine and a solvent, wherein the anionic polyacid and the cationic polyamine are derived from an acid-base reaction between a polyacid and a polyamine at a weight ratio in the range of 20:100 to 40:100.

Advantageously, the weight ratio between the polyacid and the polyamine may result in a solid structure that allows an effective dispersion of materials, such as active electrode materials, single-wall nanotubes or conductive additives in the hydrogel binder.

There is provided a method of synthesizing a hydrogel binder comprising mixing a polyacid with a polyamine at a weight ratio in the range of 10:100 to 40:100 in a solvent.

In another aspect, there is provided a method of synthesizing a hydrogel binder comprising mixing a polyacid with a polyamine at a weight ratio in the range of 20:100 to 40:100 in a solvent.

There is provided a free-standing electrode comprising a hydrogel binder and carbon nanotubes, wherein the hydrogel binder comprises an anionic polyacid and a cationic polyamine, and wherein the anionic polyacid and the cationic polyamine are derived from an acid-base reaction between a polyacid and a polyamine at a weight ratio in the range of 10:100 to 40:100.

In another aspect, there is provided a free-standing electrode comprising a hydrogel binder and carbon nanotubes, wherein the hydrogel binder comprises an anionic polyacid and a cationic polyamine, wherein the anionic polyacid and the cationic polyamine are derived from an acid-base reaction between a polyacid and a polyamine at a weight ratio in the range of 20:100 to 40:100.

There is provided a method of preparing a free-standing electrode comprising the steps of: a) mixing a hydrogel binder and single-wall nanotubes in a mixing solvent to form a slurry, wherein the hydrogel binder comprises an anionic polyacid and a cationic polyamine, and wherein the anionic polyacid and the cationic polyamine are derived from an acid-base reaction between a polyacid and a polyamine at

a weight ratio in the range of 10:100 to 40:100 and a solvent; and b) casting and drying the slurry of step (a) on a substrate to form the free-standing electrode.

In another aspect, there is provided a method of preparing a free-standing electrode comprising the steps of: a) mixing a hydrogel binder and single-wall nanotubes in a mixing solvent to form a slurry, wherein the hydrogel binder comprises an anionic polyacid and a cationic polyamine, and wherein the anionic polyacid and the cationic polyamine are derived from an acid-base reaction between a polyacid and a polyamine at a weight ratio in the range of 20:100 to 40:100 and a solvent; and b) casting and drying the slurry of step (a) on a substrate to form the free-standing electrode.

In another aspect, there is provided a battery comprising the free-standing electrode as described herein, a sulfur source, a separator and an electrolyte.

In another aspect, there is provided a battery comprising at least one free-standing electrode as described herein, a separator and an electrolyte.

Definitions

The following words and terms used herein shall have the meaning indicated:

The term “polyacid” refers to a polymer comprising at least one acidic moiety, such as a carboxylic acid group, a phosphonic acid group, a sulfonic acid group. The polymer may be derived from monomers that comprise at least one of said acidic moiety.

The term “polyamine” refers to a polymer comprising at least one amino moiety. The polymer may be derived from monomers that comprise at least one amino moiety.

The term “transition metal” refers to an element selected from Groups 3 to 12 in the Periodic Table of Elements.

The term “free-standing” when used to refer electrodes means that the electrodes may function properly by themselves. Therefore, the electrodes do not contain additional metallic current collectors.

The term “metalloid” as used in this disclosure refers to an element selected from the group consisting of boron, silicon, germanium, arsenic, antimony, tellurium, or combinations thereof.

The word “substantially” does not exclude “completely” e.g. a composition which is “substantially free” from Y may be completely free from Y. Where necessary, the word “substantially” may be omitted from the definition of the invention.

Unless specified otherwise, the terms "comprising" and "comprise", and grammatical variants thereof, are intended to represent "open" or "inclusive" language such that they include recited elements but also permit inclusion of additional, unrecited elements.

The term "about" as used herein typically means +/- 5 % of the stated value, more typically +/- 4 % of the stated value, more typically +/- 3 % of the stated value, more typically, +/- 2 % of the stated value, even more typically +/- 1 % of the stated value, and even more typically +/- 0.5 % of the stated value.

Throughout this disclosure, certain embodiments may be disclosed in a range format. It should be understood that the description in range format is merely for convenience

Claims

1. A hydrogel binder comprising an anionic polyacid, a cationic polyamine and a solvent, wherein the anionic polyacid and the cationic polyamine are derived from an acid-base reaction between a polyacid and a polyamine at a weight ratio in the range of 20:100 to 40:100.

2. The hydrogel binder of claim 1, wherein the weight ratio of the polyacid and the polyamine is 24:100.

3. The hydrogel binder of claim 1, wherein the polyacid is alginic acid or a salt thereof, the polyamine is polyethyleneimine or a salt thereof and the solvent is an aqueous medium.

4. A method of synthesizing a hydrogel binder comprising mixing a polyacid with a polyamine at a weight ratio in the range of 20:100 to 40:100 in a solvent.

5. The method of claim 4, wherein the weight ratio of the polyacid and the polyamine is 24:100.

6. The method of claim 4 or 5, wherein the polyacid is alginic acid or a salt thereof, the polyamine is polyethyleneimine or a salt thereof and the solvent is an aqueous medium.

7. A free-standing electrode comprising a hydrogel binder and carbon nanotubes, wherein the hydrogel binder comprises an anionic polyacid and a cationic polyamine, wherein the anionic polyacid and the cationic polyamine are derived from an acid-base reaction between a polyacid and a polyamine at a weight ratio in the range of 20:100 to 40:100.

8. The free-standing electrode of claim 7, wherein the weight ratio of the polyacid and the polyamine is 24:100.

9. The free-standing electrode of claim 7 or 8, wherein the polyacid is alginic acid or a salt thereof, and the polyamine is polyethyleneimine or a salt thereof.

10. The free-standing electrode of any one of claims 7 to 9, further comprising a conductive additive selected from the group consisting of graphene, graphene oxide, reduced graphene oxide, carbon fiber, carbon black and combinations thereof.

11. The free-standing electrode of any one of claims 7 to 10, further comprising an active electrode material selected from the group consisting of transition metal oxides, phosphate salts of lithium and a transition metal, metalloids and combinations thereof.

12. A method of preparing a free-standing electrode comprising the steps of:

a) mixing a hydrogel binder and carbon nanotubes in a mixing solvent to form a slurry, wherein the hydrogel binder comprises an anionic polyacid and a cationic polyamine, and wherein the anionic polyacid and the cationic polyamine are derived from

an acid-base reaction between a polyacid and a polyamine in a weight ratio in the range of 20:100 to 40:100 and a solvent; and

b) casting and drying the slurry of step (a) on a substrate to form the free-standing electrode.

13. The method of claim 12, wherein in step (a), the weight ratio of the polyacid and the polyamine is 24:100.

14. The method of claim 12 or 13, wherein in step (a), the polyacid is alginic acid or a salt thereof, the polyamine is polyethyleneimine or a salt thereof and the solvent is an aqueous medium.

15. The method of any one of claims 12 to 14, wherein in step (a), the mixing solvent is a mixture of N-methyl-2-pyrrolidone and water at a volume ratio of 1:1.

16. The method of any one of claims 12 to 15, wherein the mixing step (a) further comprises mixing the slurry with a conductive additive selected from the group consisting of graphene, graphene oxide, reduced graphene oxide, carbon fiber, carbon black and combinations thereof.

17. The method of any one of claims 12 to 16, wherein the mixing step (a) further comprises mixing the slurry with an active electrode material selected from the group consisting of transition metal oxides, phosphate salts of lithium and a transition metal, metalloids and combinations thereof.

18. The method of any one of claims 12 to 17, wherein the mixing step (a) is undertaken by ball milling and/or stirring.

19. A battery comprising the free-standing electrode of any one of claims 7 to 10, a sulfur source, a separator and an electrolyte.

20. A battery comprising at least one free-standing electrode of claim 11, a separator and an electrolyte.