Summary

The present disclosure relates to a cathode material comprising a layer of sulfur species dispersed within or thereon a porous matrix comprising a first conducting carbon material, a second conducting carbon material and a binder, wherein the second conducting carbon material is carbon fiber or carbon nanotube.

The present disclosure also relates to a cathode material comprising a layer of sulfur species dispersed within or thereon a porous matrix comprising a first conducting carbon material, a second conducting carbon material and a binder, wherein said porous matrix is interconnected with uniform pores.

In one aspect, the present disclosure relates to a cathode material comprising a layer of sulfur species dispersed within or thereon a porous matrix comprising reduced graphene oxide, carbon fiber and a binder, wherein the binder is a copolymer of acrylamide, lithium carboxylate and cyano group.

Advantageously, due to the way the cathode material is made, the cathode material may be interconnected with pores that are uniform in size and volume, large surface area and/or small ohmic resistance. The cathode material as described herein can be used in a lithium-sulfur battery and due to the large surface area, availability of electrochemically active sites for sulfur species, such as sulfur (S), lithium sulfide (Li_2S) and polysulfide (PS), can be increased, allowing both nucleation and binding to occur on the cathode surface, leading to higher specific capacities of the cathode material.

Further advantageously, the large surface area of the cathode material as described herein may lead to a decrease in the concentration of dissolved PS in bulk, reducing the undesired PS shuttling effect. Therefore, the capacity fading of the capacity fading of the electrode material may be inhibited.

The present disclosure also relates to a method for preparing a cathode material comprising the steps of:

a) coating a support with a slurry formed by mixing a mixture of a first conducting carbon material, a second conducting carbon material and a binder, wherein the second conducting carbon material is carbon fiber or carbon nanotube; and

b) adding a sulfur source in fluid state to the coated support to thereby obtain the cathode material.

The present disclosure also relates to a method for preparing a cathode material comprising the steps of:

a) coating a support with a slurry formed by mixing a mixture of a first conducting carbon material, a second conducting carbon material and a binder;

b) adding a sulfur source in fluid state to the coated support to thereby obtain the cathode material.

In another aspect, the present disclosure relates to a method for preparing a cathode material comprising the steps of:

a) coating a support with a slurry formed by mixing a mixture of reduced graphene oxide, carbon fiber and a binder, wherein the binder is a copolymer of acrylamide, lithium carboxylate and cyano group; and

b) adding a sulfur source in fluid state to the coated support to thereby obtain the cathode material.

Advantageously, by forming the coated support first and then adding the sulfur species onto the coated support, this results in the final cathode material that is interconnected with pores that are of uniform size and volume. The final cathode material may also have a large surface area as well as a smaller ohmic resistance. Therefore, the steps in the method have to be in this order whereby the cathode material is preformed, followed by dispersion of the sulfur source therein or thereon. This is in comparison to melt-diffusion methods in which the sulfur source is added to the support first, followed by the cathode material and the binder. Using melt-diffusion results in a cathode with inconsistent pore sizes.

Advantageously, the method as described herein by slurry coating may utilize inexpensive commercially available materials to develop high-performance lithium-PS batteries with large-surface-area cathodes as compared to conventional melt-diffusion method.

Further advantageously, the method as described herein is industrially scalable and environmentally friendly. In view that the cathode preparation method has a strong influence on the electrochemical

performance, different cathode preparation approaches should be considered for future designs of practical lithium-sulfur batteries.

The present disclosure also relates to a cathode material prepared by the method as described herein.

In another aspect, the present disclosure relates to an electrochemical cell comprising a cathode material as described herein and a liquid electrolyte.

In another aspect, the present disclosure relates to a lithium-sulfur battery comprising one or more electrochemical cells as described herein.

Advantageously, for the lithium-sulfur battery as described herein, high specific capacities between 1220 mAh g^{-1} and 1007 mAh g^{-1} can be achieved at charge rates of 0.2–2.0 C, with low capacity fade of lower than 0.14% per cycle over 200 cycles. At higher sulfur loading, a practical areal capacity of > 4 mAh g^{-1} can also be achieved. Remarkably, the cathode material as described herein may offer 48% higher specific capacity and 26% lower capacity fade than the sulfur cathode prepared by the conventional melt-diffused method due to differences in morphology, surface area and ohmic resistance of the cathodes.

Definitions

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

The term “interconnected” or “interconnectivity” as used here represents the characteristics of the cathode material that maintains unimpeded electronic pathways from current collector throughout the entire cathode structure during the charging and/or discharging operation of the sulfur-lithium battery, and therefore achieves good conductivity and low ohmic resistance of the cathode material.

The term “polysulfide” as used herein represents a class of chemical compounds comprising chains of sulfur atoms. The chain of sulfur atoms may have the general formal S_n^{2-} and may be a conjugate base to form a compound with metal ions, such as lithium or sodium.

The term “graphene” as used herein represents a two-dimensional allotrope of carbon in the form of a single layer of atoms with the carbon atoms arranged in a two-dimensional honeycomb lattice.

The tern ”reduced graphene oxide” is one form of graphene oxide that is processed by chemical, thermal and other methods in order to reduce the oxygen content.

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.

As used herein, the term "about", in the context of concentrations of components of the formulations, 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 and brevity and should not be construed as an inflexible limitation on the scope of the disclosed ranges. Accordingly, the description of a range should be considered to have specifically disclosed all the possible sub-ranges as well as individual numerical values within that range. For example, description of a range such as from 1 to 6 should be considered to have specifically disclosed sub-ranges such as

Claims

1. A cathode material comprising a layer of sulfur species dispersed within or thereon a porous matrix comprising reduced graphene oxide, carbon fiber and a binder,
wherein the binder is a copolymer of acrylamide, lithium carboxylate and cyano group.

2. The cathode material according to claim 1, wherein said porous matrix is interconnected with uniform pores.

3. The cathode material according to claim 1 or 2, wherein the sulfur species is a polysulfide or elemental sulfur.

4. The cathode material according to claim 3, where the polysulfide has a formula of Li_2S_n, wherein $2<n\leq8$.

5. The cathode material according to any one of the preceding claims, wherein the cathode material has a sulfur content in the range of 30 wt% to 80 wt% based on the total weight of the cathode material.

6. The cathode material according to any one of the preceding claims, wherein the reduced graphene oxide is doped with nitrogen, oxygen, sulfur, boron, phosphorus or their mixtures thereof.

7. The cathode material according to any one of the preceding claims, wherein the amount of the reduced graphene oxide is in the range of 20 wt% to 60 wt% based on the total weight of the cathode material.

8. The cathode material according to any one of the preceding claims, wherein the amount of the binder is in the range of 5 wt% to 15 wt% based on the total weight of the cathode material.

9. The cathode material according to any one of the preceding claims, wherein the carbon fiber has a diameter in the range of 0.1 nm to 100 μm.

10. The cathode material according to any one of the preceding claims, wherein the carbon fiber is vapor grown carbon fiber (VGCF).

11. The cathode material according to any one of the preceding claims, wherein the amount of the carbon fiber is in the range of 5 wt% to 35 wt% based on the total weight of the cathode material.

12. The cathode material according to any one of the preceding claims, wherein the cathode material has a sulfur loading density in the range of 1.3 mg cm^{-2} to 15 mg cm^{-2}.

13. The cathode material according to any one of the preceding claims, wherein the cathode material has a surface area in the range of 200 m^2/g to 900 m^2/g.

14. The cathode material according to any one of the preceding claims, wherein the cathode material has a pore volume in the range of 0.25 cm^3/g to 3 cm^3/g.

15. The cathode material according to any one of the preceding claims, wherein the cathode material has a pore size distrbution of mesopore size in the range of 2.0 nm to 50 nm and macropore size larger than 50 nm.

16. A method for preparing a cathode material comprising the steps of:

a) coating a support with a slurry formed by mixing a mixture of reduced graphene oxide, carbon fiber and a binder, wherein the binder is a copolymer of acrylamide, lithium carboxylate and cyano group; and

b) adding a sulfur source in fluid state to the coated support to thereby obtain the cathode material.

17. The method according to claim 16, further comprising, before said coating step (a), the step of (a1) stirring said mixture in a solvent overnight with a solid content in the range of 3 wt% to 10 wt%.

18. The method according to claim 17, wherein the solvent is water or water mixture with polar organic solvents.

19. The method according to any one of claims 16 to 18, wherein the reduced graphene oxide has a concentration in the range of 60 wt% to 90 wt% based on the total weight of solid content in the slurry.

20. The method according to any one of claims 16 to 19, wherein the carbon fiber has a concentration in the range of 5 wt% to 50 wt% based on the total weight of solid content.

21. The method according to any one of claims 16 to 20, wherein the binder has a concentration in the range of 5 wt% to 20 wt% based on the total weight of solid content.

22. The method according to any one of claims 16 to 21, further comprising, after said coating step (a), the step of (a2) drying the coated support at a temperature in the range of 40 ^{o}C to 80 ^{o}C for more than 2 hours.

23. The method according to any one of claims 16 to 22, comprising the step of preparing a polysulfide (PS) solution as the sulfur source in fluid state by stirring a mixture of sulfur (S) and lithium sulfide (Li_2S).

24. The method according to claim 23, wherein the mixture is stirred at a temperature in the range of 40 °C to 60 °C overnight in a glovebox.

25. The method according to claim 23 or 24, wherein the mixture has a S/Li_2S mass ratio in the range of 2:1 to 5:1.

26. The method according to any one of claims 16 to 22, comprising the step of obtaining said sulfur source in fluid state by heating elemental sulfur solid at a temperature in the range of 160^{o}C to 190^{o}C.

27. The method according to claim 26, wherein duration of the heating step is in the range of 5 minutes to 40 minutes.

28. An electrochemical cell comprising a cathode material according to any one of claims 1 to 15 and a liquid electrolyte.

29. A lithium-sulfur battery comprising one or more electrochemical cells according to claim 28.