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RESEARCH PAPER

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The influence of conductive additives on the performance of a SiO/C composite anode in lithium-ion batteries

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Abstract: Two conductive additives, carbon black (Super-P) and vapor-grown carbon fibers (VGCFs), were used in 2.5 Ah Li ion batteries using $LiCoO₂$ as the cathode and a SiO/C composite as the anode. The electrode shape, peel strength, electrolyte absorption, electrolyte storage capacity, volume change and the electrochemical performance were investigated. Results indicate that both batteries have high energy densities (more than 600 Wh/L), but the batteries with VGCFs have the better low temperature performance, longer cycle life, higher rate capability and lower volume expansion, which is ascribed to the superior conductive network formed by the VGCFs.

Key Words: SiO/C anode; Lithium-ion battery; VGCF; Rate capabilities; Cycle life.

1 Introduction

SiO as a promising anode material displays a high theoretical capacity of 2043 mAh g^{-1} and a less volume expansion compared with $Si^{[1, 2]}$. However, it suffers from a poor electronic conductivity [3]. Conductive additive as a necessary ingredient has been widely used in the lithium ion battery system to enhance the conductivity of electrodes $[4-7]$, especially for the Si-based anode^[8,9]. Conductive additive wrapping around the active material grains could greatly reduce the contact resistance between grains, decrease inner resistance and improve the rate capability^[10]. At present, the commonly used conductive additives in commercial lithium ion batteries are acetylene black, Super-P, Ketjen Black, CNTs and vapor-grown carbonfibers (VGCFs) etc. In order to obtain a better conductivity, new conductive additives also have been developed. Recently, Qi et al.^[11] used S-doped carbon nanotubes (SCNTs) as a conductive additive for $LiFePO₄$ cathodes in lithium ion batteries, the SCNTs exhibit a higher specific surface area, higher conductivity and better hydrophily than the pristine CNTs, thus improving the electrochemical performance. Moreover, Ke et al.^[12] have found that when graphene is used as the conductive additive, the electrode thickness has a considerable effect on the high-rate performance of $LiFePO₄$ electrode. A composite electrically conductive network consisting of CNTs and Ag was also obtained to improve the conductivity of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O₂$ material, leading to a better rate capability and cycle performance^[13]. In addition, Lin et al.^[14] studied the surface nature of five carbon materials, and found that it plays an important role in charge/discharge performance of $Limn_2O_4$

and $LiNi_{0.5}Mn_{1.5}O₄$ cathodes for lithium ion batteries. Furthermore, 2 D reduced graphene oxide conductive additives to anode could improve the electrochemical performance, owing to the "plane-to-point" conducting network formed in the electrode^[15]. Carbon black (CB) and VGCFs have been used as cathode conductive additives to improve the electrochemical performance, because the fibrous VGCFs can bridge the isolated $Li_{1.18}Ni_{0.15}Co_{0.15}Mn_{0.52}O₂$ regions to construct an effective conductive network for electron $transport^[16]$. Besides, super-aligned carbon nanotube (SACNT) conductive additive has been uniformly introduced into LiCoO₂-Super P composite cathodes, the hybrid Super P-SACNT conductive network could improve the electrode performance^[17]. Novel graphene(GN) / carbon black(CB) binary conductive additive^[18] and grapheme nanosheet (GNS) conductive additive^[19] have been used to increase the electronic conductance and improve cathode rate performance. From the researches above, much attention is paid to the influence of conductive additives on the cathode. There is little report for the influence of VGCF conductive additives on the performance of SiO/C anode and the full battery.

In this paper, Super-P and VGCFs are chosen as conductive additives of SiO/C composite anode to fabricate 2.5 Ah Li ion batteries $(LiCoO₂$ as cathode) to investigate their physical properties and electrochemical performances of the batteries.

2 Experimental

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2.1 Reagents and equipment

Cathode material (Beijing Easpring), anode material (Battery grade SiO/C, Si – 5%), separator – Ube PP/PE/PP 3 layers 16m and other materials are listed as follows:

(1) Cathode material: $LiCoO₂$ (Beijing Easpring, BET: 0.223 m² g⁻¹; TD: 5.846 g cm⁻³; particle size analysis (PSA): *D*10=3.557 μm, *D*50=8.091 μm, *D*90=14.827 μm, PH=10.67);

(2) Anode material: SiO/C composite (BET: 2.993 m² g⁻¹; TD: 2.27 g cm⁻³; PSA: *D*₁₀=8.586 μm, *D*₅₀=20.267 μm, D_{90} =43.164 μ m);

(3) Anode conductive: Super-P, VGCFs;

(4) Binder: carboxymethyl cellulose (CMC), polymerized styrene butadiene rubber (SBR);

(5) Separator (Japan Ube);

2.2 Preparation of electrode

The cathode was composed of 95 wt.% $LiCoO₂$, 2 wt.% conductive additive (Super-P), and 3 wt.% binder (polyvinylidene difluoride). $LiCoO₂$ and conductive additives were dried at 80 °C for 4 h. The solvent is N-methyl-2-pyrrolidone (NMP) with a solid-to-liquid mass ratio of 1: 3. Then the slurry was vigorously stirred for 5 h, the viscosity of which for cathode material is 3000-4000 cPa·s. The slurry was kept for 5 min under vacuum condition prior to coating on Al foil with a length of 18 m. The obtained electrode was dried continuously at four zones of 60, 80, 100 and 120 °C. The following steps are shown in Fig. 1a. The anode was composed of 90 wt.% SiO/C, 2 wt.% conductive additive (Super-P or VGCFs), 3 wt.% CMC and 5 wt.% SBR. SiO/C composite and conductive additives were dried at 150 °C for 4 h before using. The solvent is deionized water and the solid-to-liquid mass ratio is 1: 2.5. Then the slurry was vigorously stirred for 5 h. The viscosity of the slurry for anode material is 1800-2500 cPa·s. The slurry was kept for 10 min under vacuum condition prior to coating on a Cu foil with a length of 16 m. The obtained electrode was continuously dried at four temperature zones of 50, 70, 90 and 105 $^{\circ}$ C. The following steps are shown in Fig. 1a, which is similar to the cathode preparation process.

2.3 Production of batteries

As shown in Fig. 1b, the polymer cells were assembled (without sealing and electrolyte injection) with the obtained cathode, anode and the separator.

After assembling, the polymer cells were dried and weighed for the second time, and then the electrolyte was injected into the cells one by one in the injection machine. The amount of the electrolyte in one cell is 6.8g (nominal capacity (2720 mAh) \times liquid injection coefficient (2.5) = 6.8 g). Next, the sealed cells were stored in the aging room for 24 h after weighing and recording. After aging, the polymer cells were pre-charged, dried, second sealed, tailored, folded and regularized in turn. Finally, they would be put into aging room again.

Fig. 1 A flow chart for (a) electrode preparation and (b) battery assembling.

2.4 Material characterizations

The particle size and surface morphology of the samples were carried out on a QUANTA 6000 scanning electron microscope of FEI Holland. Galvanostatic charge/discharge was tested in a BTS-5V20A battery testing device of Kinte Guangzhou.

3 Results and discussion

3.1 Morphological characterization of SiO/C materials with different conductive additives

Fig. 2a shows the SEM image of the SiO/C material electrode using Super-P as its conductive additive. The white spots on the graphite are conductive additive Super-P carbon particles. Similarly, they can also be observed in SEM image (Fig. 2b) when VGCFs were used as the conductive additive in the electrode.

According to the SEM images, it can be seen that the coating electrode using different conductive additives differs in morphology. For Super-P with pretty small particle sizes, it cannot contact well with the electrode material particles when the gap is large. It could lead to the increase of the contact resistance. However, VGCFs could contact well with the electrode material particles with large gap due to their long fibrous structure. And fibrous VGCFs can bridge the SiO/C material, constructing an effective conductive network for electron transport. Therefore, the resistance of the battery decreased for the VGCF added sample, resulting in an improvement of the battery performance.

3.2 Peeling strength analysis of the SiO/C material electrode with different kinds of conductive additives

In order to measure the adhesion degree between the SiO/C material and current collector when Super-P and VGCF were used as the conductive additives, the electrode peeling

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