Facile approach to silicon based anode material from SiO for lithium ion batteries

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In recent years, researchers focus on silicon based anode materials due to their high lithium storage capacity. However, the main problem of these materials is the pulverization and loss of electronic conductivity resulted from huge volumetric change during lithiation/delithiation.

To solve this problem, various nanostructured Si and composite materials have been developed. 2, 4 They show a significant improvement in electrochemical performance. However, their complicated preparation technologies may cause high cost and be difficult for large scale production. Comparing to silicon, the volume expansion of SiO upon electrochemical lithiation is certainly suppressed due to the formation of lithium oxide and lithium silicate, acting as buffer layers. Therefore, silicon monoxide (SiO) appears particularly promising for its long cycle life and low cost. 5, 6 Nevertheless, it should be realized that the expansion rate of SiO is still close to 200% for its full lithiation.

In this work, the influence of environmentally friendly aqueous binders and carbon coating on the electrochemical performance of SiO powder anodes has been investigated in detail. Aqueous binders contain sodium alginate (Alg), polyacrylic acid (PAA), polyvinyl alcohol (PVA). In comparison with SiO, the volume expansion of SiO upon electrochemical lithiation is certainly suppressed due to the formation of lithium oxide and lithium silicate, acting as buffer layers. Therefore, silicon monoxide (SiO) appears particularly promising for its long cycle life and low cost. Nevertheless, it should be realized that the expansion rate of SiO is still close to 200% for its full lithiation.

In this work, the influence of environmentally friendly aqueous binders and carbon coating on the electrochemical performance of SiO powder anodes has been investigated in detail. Aqueous binders contain sodium alginate (Alg), styrene butadiene rubber/sodium carboxymethyl cellulose (SCMC, 3:5 by weight), polyacrylic acid (PAA), polyvinyl alcohol (PVA). In order to further increase the reversible capacity of SiO based anode composite, part of oxygen in SiO was reduced by Mg powder. The SiO/Si composite was obtained via a partial reduction reaction between silicon monoxide (SiO) and Mg, using high-energy mechanical milling process and chemical vapor deposition.

Figure 1 illustrates the cycling performance of the SiO electrodes containing four different binders at a current density of 100 mA g⁻¹ for the initial nine cycles and at 300 mA g⁻¹ for the following cycles. The reversible capacity of the SiO electrode using PVA binder was 1001.7 mAh g⁻¹ for the initial cycle. The fast capacity fade indicates that PVA is not an effective binder for SiO electrode. In comparison with PVA, PAA, SCMC and Alg as binders are more effective to stabilize the electrochemical performance of SiO electrodes. Particularly, the SiO-Alg electrode displayed the highest reversible capacity of 924.4 mAh g⁻¹ after 50 cycles. Compared to PAA and SCMC, it appears that sodium alginate is a more promising binder for SiO electrode.

As shown in Figure 2, carbon coating significantly improves the cycling performance. The capacity retention up to 100 cycles is 88.5% versus the 10th cycle at 300 mA g⁻¹ for the SiO/C-Alg electrode. Moreover, the advantage of carbon coating on SiO is more remarkable for the electrode using SCMC binder. After 150 cycles, faster capacity fade takes place for SiO/C- Alg, not for SiO/C-SCMC. Therefore, SCMC binder is robust enough to maintain mechanical and conducting stability of the SiO/C electrode.

Figure 3 shows the cycling performance of SiO/C and SiO/Si/C composite. The SCMC binder is suitable for the carbon coating anode composite, so the SiO/Si/C composite electrode was prepared using SCMC binder. In comparison with SiO/C, the SiO/Si/C composite is increased from ca.1030 mAh g⁻¹ to ca.1250 mAh g⁻¹. The higher capacity can be attributed to the enhanced ratio of silicon atom to oxygen atom. Meanwhile, the SiO/Si/C composite shows excellent cycling stability with 90.6% capacity retention at the 100th cycle versus the 2nd cycle.

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