Preparation and electrochemical performance of Si/graphite/disordered carbon composite anode for lithium-ion batteries

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Introduction

The commercial use of silicon anode materials has been hindered by the drastic volume change during lithium insertion and extraction, which inevitably introduces cracking or crumbling of anode materials and thus causes loss in capacity during cycling. To improve the stability of silicon-based anodes, silicon/carbon composite anodes attract great interest because of the good electrical conductivity and stress-buffer nature of carbon. Various methods have been employed for preparing Si/C composite anodes. Here we proposed and prepared a optimized structure of the Si/graphite/disorded carbon composites to improve the cycle performance using ultra-fine grinding and pyrolysis method. In this work, the influences of graphite substrate, proportion of silicon carbon, and pyrolysis temperature on electrochemical performance of Si/graphite/disorded carbon composite were systematically reported. Owing to the simplicity of this method, it is believed that this method is easy to scale up and will lead to an industrial route for the mass production of silicon-based anode materials.

Experimental

Nano-silicon particles(99%, 50-70 nm), sucrose, and graphite powders were used to prepare the Si/carbon/graphite composites. The nano-silicon particles were first dispersed using ultra-fine grinding for 1 hour with ethanol as dispersing medium. And then, the sucrose and graphite powders were added in the nano-silicon slurry. The nano-silicon particles, sucrose and graphite powders were mixed using ultra-fine grinding for 2 hours. The precursor was obtained after spray drying of the mixture slurry. The spray-dried precursor was calcined at 650 ℃ for 10 hours in a furnace with a flowing argon atmosphere, and then cooled naturally to ambient temperature. The final substance was the Si/graphite/disorded carbon composites.

Results and discussion

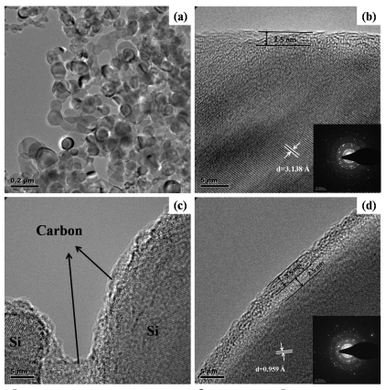

Fig.1 The TEM pictures of nano-silicon particles(a,b) and Si/graphite/disorded carbon composite (c,d)

Fig.1 shows the TEM pictures of nano-silicon particles and Si/graphite/disorded carbon composites. A uniform carbon was coated on the surface of the nano-silicon particles for the Si/graphite/disorded carbon composites by the decomposition of sucrose. And also, there was conductive carbon network between the Si/graphite/disorded carbon composites particles.

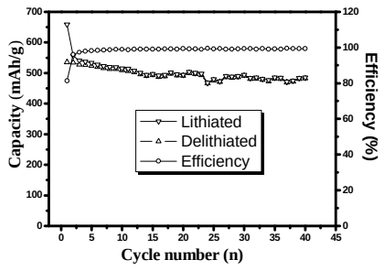

Fig.2 Cycling behavior of the electrode for Si/graphite/disorded carbon composite

Fig.2 gives the cycling behavior of the electrode for Si/graphite/disorded carbon composites. The delithiation capacity of the Si/graphite/disorded carbon composites was 535 mAh/g with 81.3% coulombic efficiency in the 1st cycle, and the delithiation capacity was 491 mAh/g with 91.7% capacity retention rate after 20th cycling.

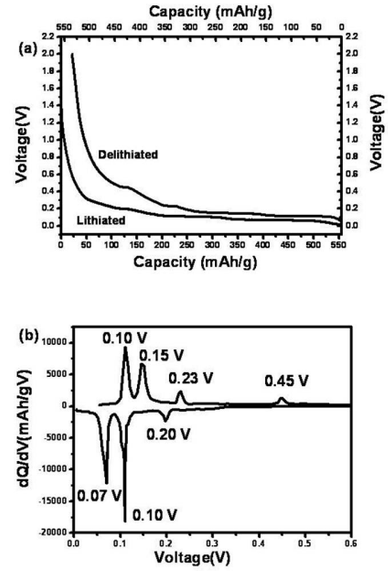

Fig.3 The capacity-voltage curve(a) and differential capacity curve(b) for Si/graphite/disorded carbon composite

Fig.3 shows the capacity-voltage curve and differential capacity curve. There was a unique 0.45 V platform of silicon material on the voltage-capacity curve with the Si/graphite/disorded carbon composites. The increase of platform during the late stage in lithium intercalation process could benefit the security issues by avoiding the formation of lithium dendrites.

Conclusions

A new type of pyrolytic-carbon coated Si/graphite composites was developed. The prepared composites exhibited excellent electrochemical performance.

Acknowledgements

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