Fabrication and Characterization of Silicon Nanoparticles Embedded in SiOx Matrix for Li-Ion Batteries

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Introduction

Silicon is an attractive negative active material due to its high gravimetric and volumetric capacity density of 4200 mAh/g and 9800 mAh/L, respectively. In spite of this advantage, Si-based anodes show numerous problems that prevent the material from being adopted in commercial Li-ion batteries.[1]

In this study, we evaluate electrochemical characteristics of nanoparticles in an anode fabricated through plasma evaporation process that shows high initial capacity and long cycle life.

Experimental

Silicon sub-oxide nanoparticles (SiOx NPs) were fabricated for active anode materials using thermal plasma evaporation. The SiOx NPs were then granulated by the water granulation method, which will be referred to as SiOx NPG. The physical properties of the synthesized material were investigated using X-ray diffraction (XRD), field emission scanning electron microscopy (FE-SEM), and X-ray fluorescence (XRF).

Coin half- and full cells were made in order to evaluate the electrochemical properties including initial efficiency, cycle life, etc. The electrodes were composed of 80wt% of SiOx NP, 10wt% of graphite, 2% of conductive agent and 8wt% of binder. Li(NiCoMn)O2 was used as the positive active material in the counter electrode of coin full cells. Current density was around 2.9mA/cm2. 1.5 M LiPF6 in the mixture of EC (ethylene carbonate), DEC (dethyl carbonate), FEC (fluoroethylene carbonate) with a ratio of 5:70:25 by volume was used as electrolyte.

Results and Discussion

The XRD patterns of SiOx NP and SiOx NPG are shown in Fig. 1. Both exhibit similar XRD patterns with a main peak at 28°, indicating that silicon phase was well crystallized during the plasma process. The crystallite size of each material was calculated using the Scherrer equation,.

Fig. 2 compares the morphologies of the SiOx NP and SiOx NPG powders observed using the FE-SEM. An XRF analysis has been developed to determine the oxygen ratio (x) as in SiOx where x is $1.0 \le x \le 1.5$.

The SiOx NP half cells show the initial charge capacity and coulombic efficiency of 1260 mAh/g and 69%, respectively, whereas the SiOx NPG half cells exhibit a much higher capacity (1400mAh/g) and coulombic efficiency (82%) at 0.2C as in Fig. 3. Fig. 4 shows the capacity variation with cycling of the SiOx NP and SiOx NPG cells at 0.5C. The latter offers a much better cycle performance with the reversible capacity of 459 mAh/g after 50 cycles.

Conclusions

The water granulated SiOx NPG electrode showed the higher initial capacity and initial coulombic efficiency. It also exhibited a good cycle performance. However, to use this material for xEVs and IT devices, surface treatments such as carbon coating would be needed.

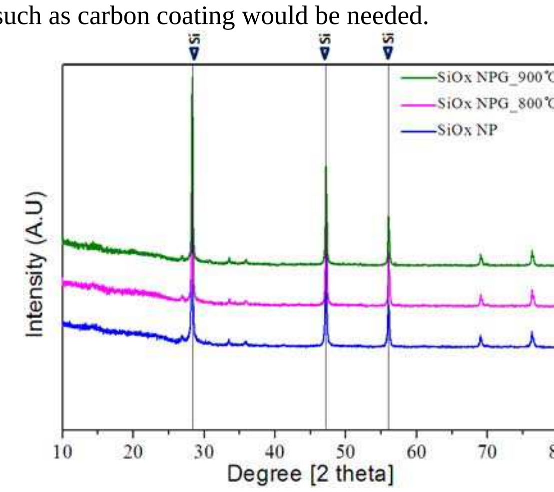

Fig. 1. XRD patterns of the as-fabricated SiOx nanoparicles [SiOx NP] and water granulated particles [SiOx NPG]

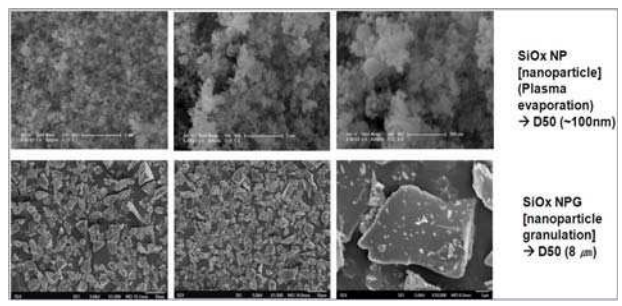

Fig. 2. SEM images of the SiOx NP and SiOx NPG; before and after granulation.

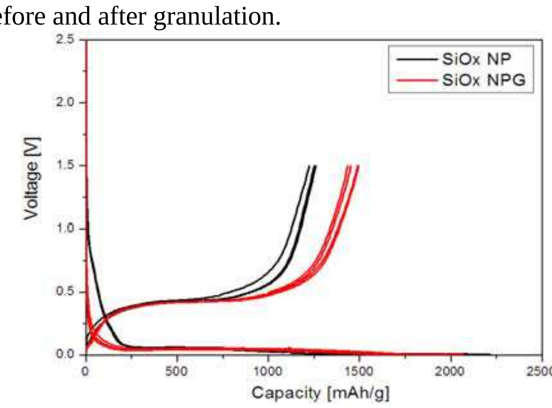

Fig. 3. Initial charge-discharge curves of SiOx NP and SiOx NPG.

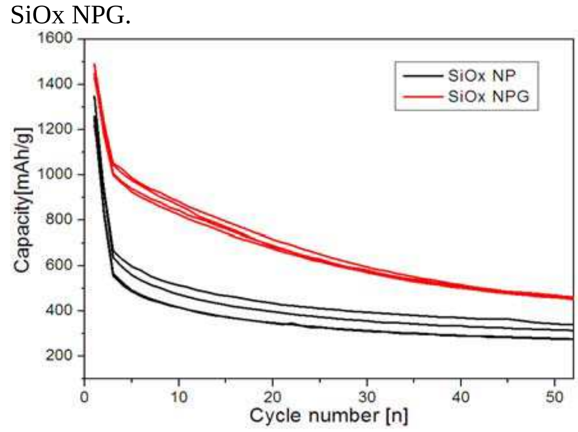

Fig. 4. Comparison of cycle performances of SiOx NP and SiOx NPG.

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References

1 A. Guerfi, P. Charest, M. Dontigny, J. Trottier, M. Lagacé, P. Hovington, A. Vijh, and K. Zaghib, "SiOx–graphite as negative for high energy Li-ion batteries," Journal of Power Sources 196 (13), 5667-5673 (2011).