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Prelithiated Si nanoparticles-carbon nanotubes composite anodes for Li-ion batteries

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Abstract

Freestanding flexible Si nanoparticles-multi-walled carbon nanotubes (SiNPs-MWNTs) composite paper anodes for Li-ion batteries (LIBs) have been prepared using a combination of ultra-sonication and pressure filtration. No conductive additive, binder or metal current collector is used. The SiNPs-MWNTs composite electrode material achieves first cycle specific discharge and charge capacities of 2298 and 1492 mAh/g, respectively. To address the first cycle irreversibility, stabilized Li metal powder (SLMP) has been utilized to pre-lithiate the composite anodes. As a result, the first cycle irreversible capacity loss is reduced from 806 to 28 mAh/g and the first cycle coulombic efficiency is increased from 65% to 98%. The relationship between different SLMP loadings and cell performance has been established to understand the pre-lithiation process of SLMP and to optimize the construction of Si-based cells. A cell containing the pre-lithiated anode is able to deliver charge capacity over 800 mAh/g without undergoing the initial discharge process, which enables the exploration of novel cathode materials.

It was also found out the SiNPs-MWNTs electrode with 3:2 Si/MWNT ratio exhibits the optimal balance between the high capacity of SiNPs and the high electrical conductivity and structural stabilization quality of MWNTs, leading to a high rate capability, high specific capacity, and cycle life surpassing the conventional slurry-cast SiNPs electrode using binder and Cu current collector. The reversible capacity is 1866 mAh/g (based on the total composite weight, the same below) at current density of 100 mA/g. After 100 cycles, the electrode retains capacity of 1170 mAh/g at 100 mA/g and 750 mAh/g at 500 mA/g. The superior performance is believed to be due to the cooperative or even synergistic effect achieved by the optimal combination. Furthermore, the freestanding feature of our electrode eliminates the non-active mass, which is promising for enhanced capacity and energy density of Li-ion cells.