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Si and Si@C Nanoparticles for Lithium-Ion Batteries Anodes: Electrode/Electrolyte Interface Evolution

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Abstract Text:

Due to increasing demand in energy storage, much attention has been paid to Si as an anode material in Li-Ion batteries because of its theoretical capacity (3579 mAh/g in the Li_{15}Si_4 alloy vs 372 mAh/g for graphitic carbon). However, silicon suffers from several drawbacks, including rapid pulverization and SEI ripening, limiting its use. Nanostructuration and protection of silicon with a carbon coating are proven methods to improve the behavior of silicon-based anodes [1]. Using the laser pyrolysis method, the synthesis of silicon-carbon core-shell nanoparticles was achieved in a continuous way, without intermediate manipulations between the synthesis of the core from silane precursor and the shell from ethylene [2].

The influence of the carbon coating on electrochemical performances was studied in coin cells in operando conditions by using electrochemical impedance spectroscopy (EIS) as well as post mortem analysis of the anode by using X-Ray Photon electron Spectroscopy (XPS), Scanning Electron Microscopy (SEM) and High Resolution Transmission Electron Microscopy (HRTEM). Special attention was paid to the first cycle because of its major importance in the formation and growth of the SEI and the long term behavior of the battery. By comparing measurements on Si and Si@C materials, EIS clearly demonstrates the beneficial effect of the carbon shell in the SEI stabilization. The stability of the the SEI resistance shows the protective effect of the the carbon shell while the SEI resistance is strongly modified and increases during lithiation. Such behavior can be related to the evolution of the chemical composition determined by XPS at different potentials during lithiation and delithiation.

![Graph showing Evolution of SEI resistance (R_{SEI}) as a function of potential during the first cycle](image-url)