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Interface Analysis of Si-based Anode in Li-ion Batteries through Electrochemical Impedance Spectroscopy and equivalent electrical circuit analysis

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Due to its high capacity compared to graphite, silicon has attracted attention for li-ion battery technology as a promising material for negative electrodes. It is abundant and non-toxic. However, this material is well known to undergo large volume changes upon lithiation and delithiation (up to 320 %). This phenomenon causes particle cracking, instability of the solid electrolyte interphase (SEI) at the interface between solid Si and liquid electrolyte, and finally leads to electrode delamination and battery performance loss. To overcome these problems, several strategies have been proposed such as using submicronic particles (< 150 nm) to mitigate the volume changes and protection of the silicon material with a carbon layer to stabilize the active surface in contact with electrolyte. Combining both strategies, Si@C core-shell nanoparticles synthesized in one step process have recently been proposed as a promising anode material [1]. Specifically, the Si-based nanoparticles are synthesized in a two stage laser pyrolysis reactor, which yields carbon coated silicon nanoparticles in a single step. This approach mitigates material oxidation because there is no air exposure between the synthesis of the core and shell. Additionally, the nanometric size of the particles prevents material pulverization upon cycling. The synthesis technique also allows control of the core crystallinity, and both highly crystalline and amorphous silicon cores have been synthesized. Moreover, the shell thickness can be tuned by changing the flow of carbon precursor.

To optimize the design of composite electrodes based on such active materials, an in-depth understanding of their performance and chemical/mechanical degradation processes remains critical. In this work, the analysis of the electrochemical performances of such Si-based electrodes was performed using several electrochemical techniques to compare crystalline or amorphous Si nanoparticles without shell as well as crystalline Si nanoparticles coated by a thin or a thick layer of carbon (Si@C). The EIS study, carried out at various states of lithiation of Si material, allows tracking the evolution of several critical parameters (for example SEI resistance and charge transfer resistance) of the equivalent electrical circuit describing the electrode electrical behavior. Figure 1a shows typical impedance spectra while Figure 1b shows the evolution of the charge transfer resistance for the different coated and non-coated materials. A very different behavior is observed as a function of the interface material and carbon thickness. The modification of the interface between electrolyte and Si or Si@C materials is also visible on measured equilibrium potentials and power capabilities. This presentation will be devoted to the analysis of the impact of interfaces between electrolyte and Si or Si@C materials on electrochemical performances.

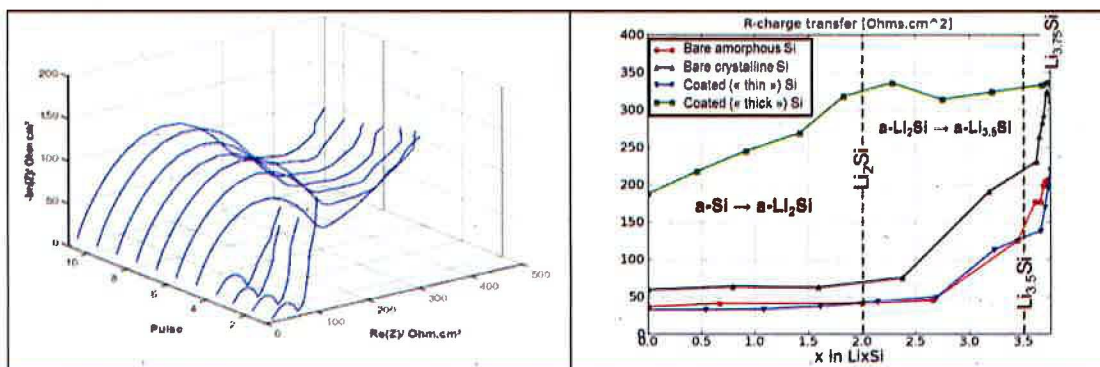


Figure 1: a) example of Impedance spectra during the lithiation in a non-coated crystalline silicon electrode and b) charge transfer resistance calculated from spectra fitting for different materials (i.e bare crystalline or amorphous material and Si coated Si with a C shell and two different C content

[1] J. Sourice, A. et al, ACS Appl. Mater. Interfaces 7 (2015) 6637.