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Analysis of Si-based Anode in Li-ion Batteries Combining Electrochemical Characterization and Multi-Scale Modeling

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Due to its high capacity compared to graphite, silicon has attracted attention among li-ion batteries technology as a promising material for negative electrodes. It is abundant, not toxic. However, this material is well known to be subject to large volume changes upon lithation and delithation (up to 320 %). This phenomenon causes particles cracking, instability of the passivation layer appearing at the interface of solid Si and liquid electrolytes (SEI) and finally leads to electrode delamination, affecting the cycle performance of such batteries in particular long time performances. To overcome these problems, several strategies have been proposed such as using submicronic particles (i.e.< 150 nm) to mitigate the volume changes and/or protection of the silicon material with a carbon layer [reference]. Combining both strategies Si@C core-shell nanoparticles synthesized in one step process have recently been proposed as a promising anode material [1]. More specifically, the Si-based nanoparticles are synthesized in a two stages laser pyrolysis reactor, which allows obtaining carbon coated silicon nanoparticles in one single step, mitigating oxidation because there is no air exposure between the synthesis of the core and shell as well as particle degradation due to nanometric size. The synthesis technique also allows controlling organization of the core, both crystalline and amorphous silicon cores have been synthesized. Moreover, the shell thickness can be varied 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, multi-scale modeling is developed based on the analysis of the electrochemical performances of electrodes composed of Si-based nanoparticles (with and without shell). Several electrochemical techniques such as GITT (Galvanostatic Intermittent Titration Technique), galvanostatic cycling and Electrochemical Impedance Spectroscopy (EIS) were used to characterize the battery performance. These data provide physical parameters to feed a mathematical model (Newman) of the Si electrode based on the porous-electrode theory [2]. This Newman model combines description at the nanoparticle scale up to the composite electrode scale. Besides, the EIS study, carried out at various cell states of lithiation of Si material, allows tracking the evolution of several critical parameters (for example SEI resistance, charge transfer resistance at the interface) of the mathematical model (figure 1). Figure 1b shows the evolution of the exchange current density with SOC for coated and non-coated materials together with the expected theoretical evolution with constant kinetic rate evidencing the need for an accurate modelling. This presentation will be devoted to our first results concerning such a modelling and application to the multiscale behavior of coated as well as non-coated silicon as active anode materials.

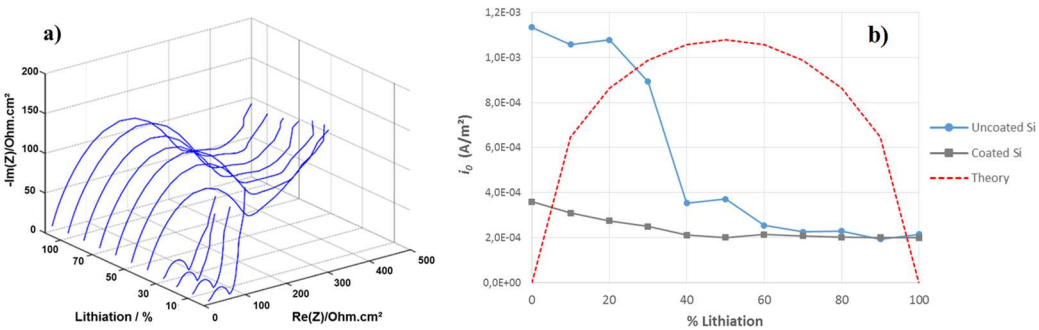

Figure 1:a) Impedance spectra during the lithiation of a non-coated crystalline silicon electrode and b) exchange current density (i_0) calculated from spectra fitting for different materials (i.e bare and coated Si) compared to theoretic evolution of i_0 according to the Butler-Volmer equation and constant kinetic rate

[1] J. Sourice, A. *et al*, ACS Appl. Mater. Interfaces 7 (2015) 6637.
[2] J. Newman, W. Tiedemann, AIChE J. 21 (1975) 25.