

SEI CHARACTERIZATION AND FAILURE MECHANISM OF SI ELECTRODES IN FULL LI-ION CELLS

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Silicon-based electrodes are very attractive negative electrodes for lithium-ion batteries (LiB) compared to graphite due to their very high specific capacities (3572mAh g⁻¹ vs 372mAh g⁻¹). However, a high cycling irreversibility appears on cycling due to 300 % volume expansion of the silicon particles, leading to cracks and decrepitation, creating new surfaces of silicon bound to react with the electrolyte. Although the interface between Si particles and the electrolyte is playing a major role in the electrochemical performance, it has rarely been characterized in depth and the failure mechanism of silicon-based electrodes has been studied only in a half-cell configuration [1].

This work takes part in the on-going European project Baccara (BAttery and superCapacitor ChARAacterization and testing) (<http://project-baccara.eu/>). Cells (nano-Si vs NMC) are cycled in carbonate electrolyte with a limited lithiation capacity of 1200mAh g⁻¹ of silicon at a rate of one lithium in 2h in delithiation and lithiation of the silicon electrode (Figure 1a).

Classical and advanced techniques (NMR, XPS, TEM-EELS, FIB-TOFSIMS) are used to investigate and characterize silicon surface, Si/electrolyte interface, lithiated/non lithiated phases and explore and analyse their evolution upon aging/cycling of the full cell. Surface investigations show that a continuous SEI layer is formed during cycling, mainly composed of inorganic species such as LiF and organic species. The formation of the inorganic part of the SEI occurs during the early stage of cycling and this layer was found to not increase over the course of cycling (Figure 1b). Although the organic species produced by the degradation of organic solvents seem to be lithiated during the first part of the cycling, carbonate species found in the outer part of the SEI for an extended cycling are clearly not lithiated.

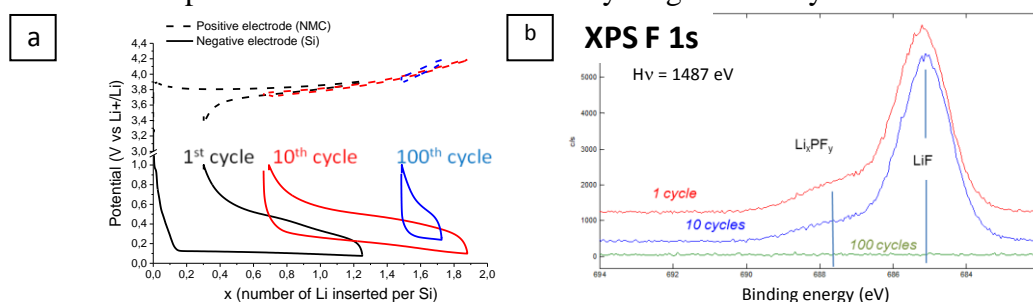


Figure 1 : a) Electrochemical behavior of negative and positive electrodes versus x, the lithium content at different cycles and b) F 1s XPS spectra of Si electrodes after 1st, 10th and 100th (end of lithiation)

References

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