Towards Improved Cycling Performances of Silicon Electrodes after Electrochemical Pre-formatting

Florent Lepoivre[a,b], Dominique Larcher[c,d], Jean-Marie Tarascon[a,b,d]

[a] *Chimie du Solide et Energie, Collège de France, 11 Place Marcelin Berthelot, 75005 Paris, France*

[b] *Sorbonne Universités – UPMC, Université Paris VI, 4 Place Jussieu, 75005 Paris, France*

[c] *Laboratoire de Réactivité et Chimie des Solides (LRCS), UMR CNRS 7314,33 rue Saint Leu, 80039 Amiens, France*

[d] *Réseau sur le Stockage Electrochimique de l'Energie (RS2E), FR CNRS 3459, France*

email address of the presenting author: lepoivre@zoho.com

Li-ion batteries are the cornerstone of the mobility market due to their high performances out-ranging any other battery technology. Yet, they hardly meet the gluttonous demand of portable devices in term of autonomy, requiring development of higher capacity materials. Negative electrode materials such as alloys would allow for large gain in capacity if they were not hampered by intrinsic limitations[1]. For instance, silicon suffers from large volumetric expansion[2] altering the electrode microstructure hence limiting its cycling performances.

In our work, that swelling was minimized using Si nanoparticles and an environmentally friendly electrode preparation[3] enabling superior cycling retention of Si half-cells, yet at the price of capacity lower than the theoretical one. Two methodologies were developed to counter this capacity loss consisting in i) lithium plating/stripping during the 1st cycle or ii) constant load 1st discharge. The morphological and chemical changes induced by the aforementioned formatting steps were investigated by TGA analysis and TEM observations. It revealed the formation of a protective SEI layer along with the activation of extra silicon otherwise inactive, triggered by the sustained low potential applied to the Si electrode. Moreover, tremendous improvements of the cycling performances were observed with formatted electrodes, which exhibit capacities greater than 2100 mAh/$g_{of\ Si}$ for several hundreds of cycles without any capacity constraints.

We therefore demonstrate that such electrochemical formatting steps are a viable and well-controlled alternative to chemical and texturing routes previously developed for silicon anodes. Such results echo with the need for high energy density anode materials, emerging from the improvements on the positive side and the development of new technologies such as Li-Air, for which lithium metal is still mostly used.

References :

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