Interface Optimization Principles of Si Anode for Li-ion Batteries

Judith Alvarado[a], Mahsa Sina[a], Hitoshi Shobukawa[a], Ying Shirley Meng[a*],

[a]*Department of NanoEngineering, University of California San Diego, La Jolla, CA 92093-0448, United States*

[*]shmeng@ucsd.edu

The high gravimetric capacity of lithiated silicon (Si) has motivated research as a potential anode for lithium ion batteries; however, major challenges remain for the implementation of Si in commercial devices. The large irreversibility associated with the formative cycles of Si anodes is primarily due to mechanical pulverization and chemical instability. The mechanical pulverization of the Si active material has largely been improved by adopting nanostructures and thin films which can accommodate the strain of full lithiation.[1-2] Conversely, the chemical instability due to the consumption of Li ions as the electrolyte is reduced to inactive side products,[4] forms the solid electrolyte interphase (SEI) leading to poor capacity retention and Coulombic efficiency. Therefore, a direct investigation of the SEI morphology and composition to improve Si electrodes has been carried out.

We have improved the performance of Si anode by optimizing electrode fabrication, incorporating electrolyte additives, and coating for Si nanoparticles. The optimized parameters improved the initial Coulombic efficiency to 88.8 % without the use of additives or coatings. A series of advanced characterization techniques was used to investigate the material and interface properties. Using atomic resolution scanning transmission electron microscopy and electron energy loss spectroscopy (EELS) demonstrates that a uniform dense SEI consisting of primarily inorganic compounds improves longterm cycling performace (Figure 1(a)). Anoxic X-ray photoelectron spectroscopy and time-of-flight secondary ion mass spectrometry depth profiling techniques were used to accurately characterize effect fluoroethylene carbonate (FEC) on amprphous Si thin films (Figure b,c)), giving evidence to fast reduction kinetics of FEC, leading to the improving silicon's conductivity via silicon native oxide etching and improved Li-ion transport through the SEI via a shuttling mechanism. The use of conformal coating to form an artificial SEI through molecular layer deposition can further improve both the mechanical and chemical instability of Si electrodes. The understanding of the SEI morphology and chemical composition will shed light on the design of future Si electrodes.

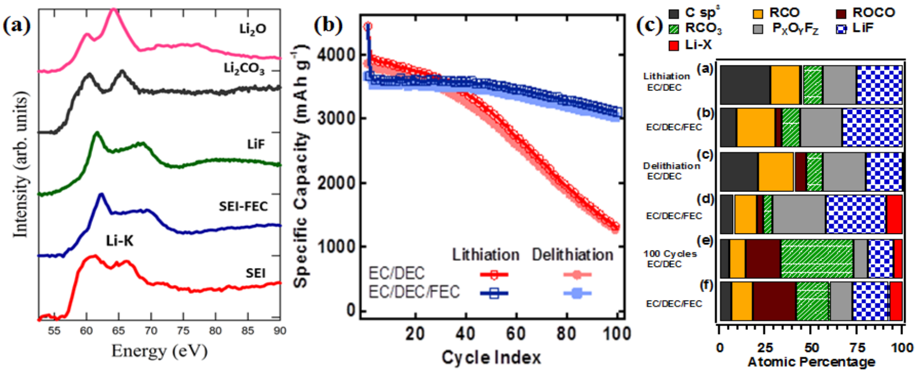

Figure 1(a) EELS spectra Li-K edge of cycled Si composite electrode (b) Specific capacity versus cycling of a-Si thin films at C/2 rate and (c) relative SEI composition after cycling.

[1] Xu, C.; Lindgren, F.; Philippe, B.; Gorgoi, M.; Björefors, F.; Edstrom, K.; Gustafsson, T. Chem. Mater. 27 (2015) 2591-2599.

[2] Liu, N.; Wu, H.; McDowell, M. T.; Yao, Y.; Wang, C.; Cui, Y. Nano Lett. 12 (2012) 3315–3321.

[3] Verma, P.; Maire, P.; Novák, P. Electrochim. Acta 55 (2010) 6332–6341.