Computational study on the stability of hybrid nanostructures of Silicon and Multi-walled carbon Nanotubes

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The importance and demand of lithium ion batteries as portable energy storage system is continuously growing, due to its increasing use in consumer and portable electronics such as cell phones, laptops, PDA's. They are also used in hybrid vehicles (HEV's) and plug-in electric vehicles (PEV's), replacing the internal combustion energy driven by traditional fossil fuels. Thus, continuous efforts are being directed towards reduction of the cost and weight of materials used in Li ion batteries. Traditional Li ion batteries still use graphite anodes that exhibit a capacity of ~372 mAh/g. Research in silicon based anodes has gained considerable importance due to its ability to provide ~4200 mAh/g Li alloying capacity. However silicon not being a good electrical conductor, requires a conductive matrix to maintain electrical conductivity throughout the anode. However, the colossal volumetric expansion associated with lithiation of silicon (~300 %) causes mechanical degradation of the active material as well as the conductive matrix, resulting in capacity fade even when cycled for only a few electrochemical cycles.

Carbon nanotubes (CNT) show excellent electrical conductivity as well as mechanical strength. Creating hybrid nanostructures of CNT and Si can be expected to alleviate some of the issues associated with the use of Si anode materials. Recent publications serve as a testimony of this concept [1, 2]. Experiments show that the core-shell hybrid structures of MWCNT/Si exhibit an initial capacity of ~2800 mAh/g. However, when deposited as a film, the anode performance deteriorates rapidly after first 7-8 cycles [1]. Investigation of the mechanisms responsible for the capacity fade will pave the way for further improvement in the performance of this particular novel anode configuration.

We utilize a thermodynamically consistent theoretical framework exploiting a finite element setting to model the Li intercalation induced deformation processes as well as failure of the hybrid anode configuration [3]. Delamination at the MWCNT/Si interface is modeled using a novel cohesive law [4]. Transport of Li inside the electrode is coupled with the mechanical stress field in a finite deformation setting. Possible mechanisms contributing to the capacity fade of the aforementioned anode configuration will be explored, using this framework. A detailed parametric study of the hybrid nanostructure will be presented. Based on the results, the opportunity and the impetus for interface control additives (ICA) will also be discussed.

References:

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