

First principles investigation on the lithiation behavior of nanostructured silicon-based alloys and composites

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Nanostructured silicon-based alloys and composites have received great attention as a possible replacement of the conventional carbon-based anodes due to their higher lithium storage capacity. In many cases, however very little is known about their electrochemical properties and microstructure evolution during lithiation and delithiation, despite their importance for overcoming many technical hurdles faced in practical use. Using first principles-based atomistic modeling, we have explored the lithiation and delithiation behavior in various Si-based nanostructures and nanocomposites including nanowires, Si-C composites, and Si suboxides.

This talk will present our recent progress, particularly focusing on addressing (1) the lithiation and delithiation mechanisms of Si near the surface and interface, with comparisons to those in bulk Si, and (2) how the surface and interface affects the performance of Si-based nanomaterials as Li-ion battery anodes, such as charging rate and capacity retention. Our study shows that by alloying Si with transition metal (M) elements, the theoretical capacity is compromised slightly, but in return the Si-M network may help stabilize the lithiated host matrix and therefore contributes to the improved cycling performance. Similarly, the presence of surfaces and interfaces has been also found to alter the lithiation behavior considerably; for instance, the Li mobility along the surface or interface tends to be significantly enhanced by several factors. We will also discuss how the Li mobility and distribution are determined by alloy composition and local atomic environment. The improved understanding may offer important guidance for the rational design of nanostructured Si-based alloys and composites in order to maximize their capacity retention and rate capability.