Nano-Composites as Anode Materials for Li-Ion Batteries

Subproject JP8.1: *Atomistic modeling of nanoparticular Silicon dispersed in SiCN(O)- and SiCO-based matrices*
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Project description:
We use density functional theory (DFT) calculations and classical molecular-dynamics (MD) simulations to study lithiation of Si/SiOC and Si/SiCN composites on the atomic level.

During the *first period* we have successfully generated low-energy model geometries of amorphous Li-Si alloys with varying Li concentration (*J. Phys. Chem. C*, 2013, **117** (37), pp 18796–18803) and of unlithiated amorphous SiOC ceramics. Figure 1.A summarizes our results for Li-Si alloys, showing in particular the existence of various two-phase regions that appear during Li insertion and extraction. These two-phase regions are believed to contribute to internal degradation by means of crack formation during cycling of Si anodes. Figure 1.B shows a low-energy model for a carbon-rich SiOC ceramic. Calculated formation energies and radial distribution functions agree reasonably with experimental measurements (not shown).

During the *second period*, our investigations are currently focused on the following aspects:

- DFT calculations on Li diffusivities in Li-Si alloys as function of Li concentration.
- DFT calculations on thermodynamics and kinetics of interfaces between c-Si and a-Li-Si as to reveal the origin of anisotropic swelling of Si nanowire anodes.
- DFT calculations on Li storage sites and diffusion coefficients in SiOC.
- DFT structure modelling of carbon-rich SiCN; Li storage sites and diffusivities.
- Development of reactive bond-order potentials for the above systems.
- Classical MD simulations to assess mechanical behavior of (lithiated) Si/SiOC and Si/SiCN composites.
- Extension of simulations to Li-Sn systems.

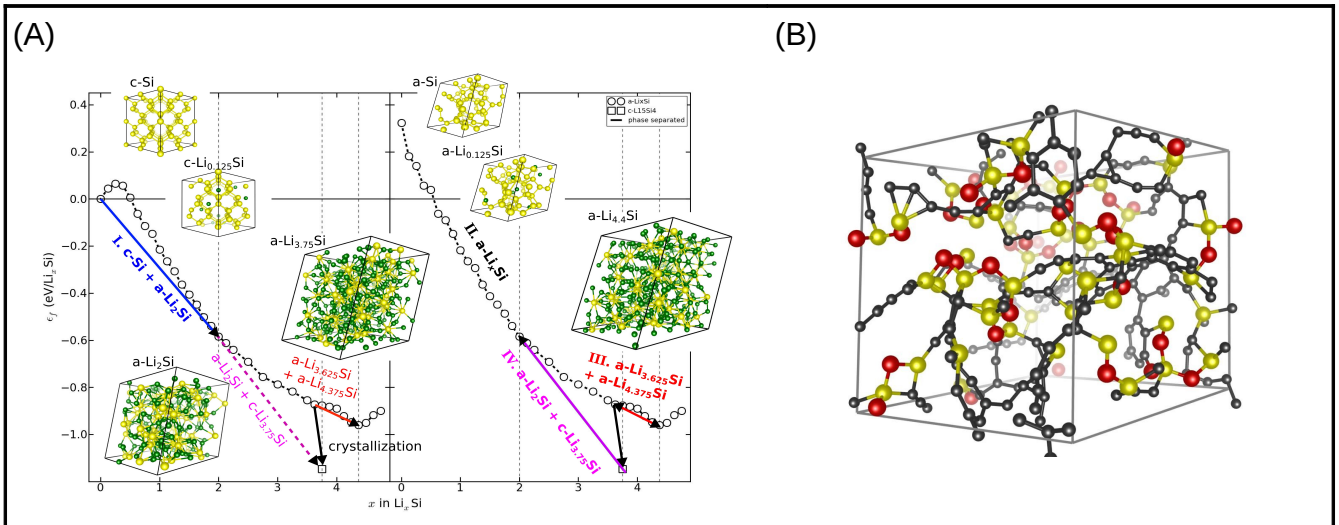

Figure 1: *(A) Model geometries, formation energies and phase evolution of Li-Si systems. Two-phase regions are identified as potential origins of internal degradation by means of cracking. (B) Structure model of a carbon-rich SiOC ceramic. Preliminary measurements suggest good energetic and structural agreement with experimental reality.*