Lithium Permeation Through Thin Silicon Layers

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Lithium permeation through amorphous silicon layers with thicknesses between 2 and 100 nm and adjacent interface layers was measured by secondary ion mass spectrometry (SIMS) and *in situ* by neutron reflectometry (NR). Such experiments are interesting for research on nanostructured electrode materials in Li based batteries, Li ion selective electrodes, and sensors [1, 2]. Interface limited Li transport was recently proven to account for irreversible capacity losses [3].

Stacks with a repetition of [Si / ^{7}LiNbO$_3$ / Si / ^{6}LiNbO$_3$] multilayer units were used for analysis. Here, two isotope enriched lithium niobate layers are adjacent to a silicon layer. The lithium niobate layers serve as solid state Li reservoirs. Annealing leads to a mutual exchange of the two Li isotopes through the silicon layer and the interfaces by Li permeation. This process modifies the Li isotope fraction in the solid state Li reservoirs, which is used to derive the permeability (diffusivity × solubility).

The presence of a diffusion controlled process was proven by a significant dependence of the Li isotope exchange time on silicon layer thickness. Li permeability in silicon is approximately constant at thicknesses above 30 nm and is strongly enhanced below 30 nm. The activation enthalpy for Li permeation through silicon amounts to $\Delta H = (1 \pm 0.2)$ eV for silicon layers with a thickness below 30 nm, and to $\Delta H = (2 \pm 0.2)$ eV for larger silicon layer thicknesses.

Possible explanations for the measured higher Li permeability (diffusivity × solubility) in thinner silicon layers may be a higher Li diffusivity due to a larger concentration of fast diffusing Li species and/or a higher Li solubility due to, *e.g.*, size confinement [4].

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