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Lithium Permeation through Ultrathin Silicon Layers measured by Neutron Reflectometry

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Neutron reflectometry (NR) was applied to measure non-destructively and in-situ Li permeation through thin silicon layers [1,2]. Such experiments are interesting for research on nanostructured electrode materials in lithium based batteries, Li ion selective electrodes, and sensors [1,2]. Understanding Li transport through interface layers in rechargeable batteries by in situ methods is a hot topic of current academic and industrial research [2]. Interface limited Li transport was recently proven to account for irreversible capacity losses [3].

Using NR we identified the control mechanism (diffusion or interface controlled) of Li permeation through thin Si layers embedded in oxide based solid state Li reservoirs. Stacks with a repetition of five [Si / 7LiNbO_3 / Si / 6LiNbO_3] units were used for analysis. Si layer thicknesses between 2 and 22 nm were studied. Two types of Bragg peaks were detected in the NR pattern. One originates from LiNbO_3/Si chemical contrast, the other from $^6\text{Li}/^7\text{Li}$ isotope contrast. Diffusion annealing at 240°C reduced only the intensity of second type of Bragg peak, demonstrating that the decrease is a measure of the ^6Li and ^7Li isotope exchange through the Si and interface layers. The presence of a diffusion controlled process was proven by a significant dependence of the Li isotope exchange time on Si layer thickness. The experiments showed that the lithium silicate interface layer, which appears by the proximity of silicon to oxide based solid state electrolytes or cathodes, does not limit Li permeation.

The temperature dependence of Li transport through thin silicon layers was also measured by NR between 85°C and 420°C . The obtained activation enthalpy of the Li permeability (diffusivity X solubility) of 1 eV is nearly identical for all Si layer thicknesses. Hence, the intrinsic process of Li transport is independent of the Si layer thickness. A strong increase of the pre-exponential factor (entropy factor) with decreasing Si layer thickness is also observed. This enhances the Li permeability by three orders of magnitude when the Si layer thickness is decreased from 22 to 2 nm at each temperature studied.

References:

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