Lithium Transport through Ultrathin Silicon Layers

E. Hüger,[1] L. Dörrer,[1] T. Geue,[2] J. Stahn,[2] H. Schmidt[1]

[1] Technische Universität Clausthal, Institut für Metallurgie, AG Mikrokinetik, Germany
[2] Laboratory for Neutron Scattering, Paul Scherrer Institute, Villigen PSI, Switzerland

Corresponding author: Erwin Hüger, E-Mail: erwin.hueger@tu-clausthal.de

The investigation of lithium transport through ultrathin layers is an important task in lithium-ion battery research. Such layers are present in batteries at the solid electrode/electrolyte interface (SEI layer) [1], as ion selective membranes [2] or in form of thin film electrodes [3]. The knowledge of diffusivities and permeabilities in these layers is essential for charging and discharging times and consequently for the power density of the battery. A tailored modification of the SEI layer by the insertion of ultrathin solid state layers like lithium niobate or silicon may improve the performance of a battery by a drastic reduction of the interface resistance for Li transport at the electrode/electrolyte interface and is nowadays in the focus of academic and industrial research [1,2]. Further, thin layers made e. g. of silicon or other materials are also developed as high capacity electrode materials [3]. Hence, it is of high interest to develop techniques that directly allow to measure Li transport from a lithium reservoir (like it is present in an electrolyte) through the electrode/electrolyte interface and within the electrode on the nanometer scale.
This contribution presents non-destructive measurements of lithium transport parameters (diffusivity, permeability) in nanometer sized silicon layers done by a novel neutron reflectometry (NR) based approach [4]. The basic structure used for the investigations is a triple layer of the form $^{7}LiNbO_3/Si/^{6}LiNbO_3$ (deposited on an appropriate substrate by ion-beam sputtering) with single layer thicknesses in the nanometer range (5-50 nm). Here, two isotope enriched lithium niobate layers (as Li reservoirs) are adjacent to a silicon layer. Annealing at elevated temperatures leads to a mutual exchange of the two Li isotopes through the silicon layer and the interfaces. For analysis by NR a multilayer arrangement of the given structural unit is used. Two types of Bragg peaks were detected in the NR pattern. One originates from $LiNbO_3/Si$ chemical contrast, the other one from $^{6}Li/^{7}Li$ isotope contrast. Annealing reduces the intensity of second type of Bragg peak only, demonstrating that this decrease is a measure of the ^{6}Li and ^{7}Li isotope exchange through the Si layer [4]. From the decrease of the Bragg peak, the transport parameters can be calculated and information on the rate determining step of the transport process can be derived. Recent results from the experiments done between 180° and 360°C will be presented and discussed.

References

[1] C. Yada, C. Brasse, Springer Automotive Media, ATZelektronik **9** (2014) 20; T. Ohtomo et al., J. Power Sources **233** (2013) 231; N. Kamaya et al., *Nature materials* **10** (2011) 682; M. Ogawa et al., *J. Power Sources* **205** (2012) 487; N. Ohta et al., *Electrochem. Commun*. **9** (2007) 1486.
[2] T.T. Truong, et al., *Adv. Mater.* **23** (2011) 4947.
[3] M. T. McDowell et al., *Adv. Mater.* **25** (2013) 4966.
[4] E. Hüger, L. Dörrer, J. Rahn, T. Panzner, J. Stahn, G. Lilienkamp, H. Schmidt, *Nano Lett.* **13** (2013) 1237.

This work is licensed under a Creative Commons Attribution 4.0 International License.
To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/