Li Diffusion in TaS_2 Single Crystals – Effects of Temperature, Pressure and Crystallographic Orientation

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A basic process in rechargeable lithium batteries is the reversible intercalation/deintercalation of Li ions into/from host electrode materials. Kinetics of this process is not well understood yet. Tantalum disulfide as a layered transition metal dichalcogenide attracts interest as a model system due to its specific electrical properties.

TaS_2 single crystals were grown via the chemical vapor transport (CVT) method with iodine as a transport agent. X-ray diffraction powder pattern ascertained that this synthesis yielded a single phase 1T-TaS_2 crystal. To establish one-dimensional diffusion conditions parallel to *ab*-plane, crystals with well developed surfaces with minimum size of 2×2 mm were encapsulated in epoxy and then one side was cut open by laser. Subsequently samples were intercalation with 1.6 $\text{mol} \cdot \text{L}^{-1}$ *n*-butyl lithium solutions in *n*-hexane (BuLi). The lithium intercalation duration was varied from 20 h to 136 h, while the reaction temperature ranged from 280 to 329 K. After intercalation experiments, profiles of Li, Ta and S were measured parallel to the *ab*-plane of the crystal by LA-ICP-MS (LASER Ablation - Inductively Coupled Plasma - Mass Spectroscopy). In order to study the lithium diffusion coefficients perpendicular to *ab*-plane and compare them to diffusion parallel to *ab*-plane, embedded crystals in epoxy with one open surface were lithiated by the same intercalation method and subsequently measured using SIMS (Secondary Ion Mass Spectrometry). The results indicate that lithium diffusion parallel to *ab*-plane is at least three orders of magnitude faster than diffusion perpendicular to *ab*-plane.

Additionally, preliminary results of the diffusion coefficients derived from the experiments under uniaxial pressure between 225–590 MPa are presented and compared to diffusion coefficients obtained at ambient pressure. The developed cell enabled us to conduct experiments without having BuLi solution being under pressure and with less degradation in comparison to hydrostatic pressure experiments.

All the lithium diffusion profiles measured perpendicular and parallel to *ab*-plane by using SIMS and LA-ICP-MS were fitted to specific solution of Fick's second law considering the given boundary conditions and from the fits lithium diffusivities (D_{Li}) and lithium contents at the crystal edges ($x_{\text{Li,S}}$) were calculated.