

Investigating the electrochemical stability of cubic LLZO garnets using field stress experiments

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Since performance and safety of current Li-ion batteries are limited by the instability of the utilized organic electrolytes, replacing them by solid ionic conductors is of major interest. Cubic $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) garnets are among the most promising solid electrolytes for next-generation Li-ion batteries, however, their electrochemical stability is still an unsettled matter. While wide electrochemical windows are generally reported in literature [1], theoretical studies question the stability of LLZO against high voltage cathodes [2]. To get a deeper understanding of the stability behavior of LLZO and its consequences for possible applications, further research is required.

In this work, we used field stress experiments to investigate the decomposition behavior of cubic LLZO garnets. DC voltages up to 5 V were applied on LLZO single crystals heated to elevated temperatures up to 350 °C using a combination of macro- and microscopic ionically blocking Au electrodes. Besides measuring the arising currents, we used electrochemical impedance spectroscopy (EIS) to monitor local changes in the conductivity behavior of the material. Furthermore, laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) was used to investigate stoichiometry variations induced by the electric field. Strong changes in the chemical composition close to the polarization electrodes were found and their correlation with changes of electrical properties are discussed.

[1] S. Ohta, T. Kobayashi and T. Asaoka, *J. Power Sources*, 196 (2011), 3341-3345

[2] F. Han et al., *Adv. Energy Mater.*, 6 (2016), 1501590