Investigating stoichiometry and conductivity variations in LLZO caused by preparation or field stress

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Since currently used Li-ion batteries suffer from problems caused by the instability of the utilized organic electrolytes, it is of major interest to replace these liquid electrolytes by more stable inorganic solid ion conductors. Due to their high Li-ion conductivity as well as chemical and electrochemical stability, cubic $Li_7La_3Zr_2O_{12}$ (LLZO) garnets are among the most promising candidates for future all solid state Li-ion batteries. Although a lot of research is focussed on LLZO in recent years, the reproducible synthesis of highly conductive garnets remains challenging. In order to further improve the material properties of LLZO garnets, a better understanding of the causes of these conductivity variations is required.

In this work, we show how local conductivity variations of LLZO samples correlate with local stoichiometry changes. Polycrystalline Al-stabilized LLZO pellets were investigated by employing spatially resolved electrochemical impedance spectroscopy (EIS) in combination with laser ablation inductively coupled plasma optical emission spectroscopy (LA-ICP-OES). Furthermore, field-stress experiments were conducted on single crystals of Ga-doped LLZO and Ta-doped LLZO. In order to create stoichiometry gradients in a controllable way, DC fields were applied to the samples at elevated temperatures. The thereby generated polarisation phenomena were investigated by microelectrode measurements.