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Towards a better Understanding of the Conductivity of $Li_7La_3Zr_2O_{12}$: Correlating Composition Variations and local Conductivities

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Owing 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 solid-state electrolytes to be used in future Li-ion batteries [1]. Although a lot of research has been focussed on LLZO in recent years, the relationship between chemical composition and conductivity behaviour is still not completely understood. In order to further improve the material properties of LLZO garnets, information about the exact stoichiometry as well as its impact on the sample conductivity is required.

In this work, we present the analysis of various LLZO samples using several elemental analysis techniques, including laser ablation inductively coupled plasma optical emission spectroscopy (LA-ICP-OES), laser induced breakdown spectroscopy (LIBS), and secondary ion mass spectrometry (SIMS). Different spatially resolved experiments including two-dimensional imaging and depth profiling are shown, which reveal significant variations in sample stoichiometry not only in terms of metal cation composition but also with respect to oxygen and hydrogen. Further variations come into play by exposing LLZO to voltages in the range of a few volts at elevated temperatures. In order to investigate the impact of these variations on the ionic conductivity of the garnets, the chemical analysis was combined with microelectrode impedance spectroscopy (EIS) leading to spatially resolved conductivity values.

Literature:

[1] R. Murugan, V. Thangadurai, W. Weppner, Angew. Chem. Int. Ed. 2007, 46, 7778-7781.