Spatially resolved electrochemical characterization and chemical analysis of Li$_{6.4}$Al$_{0.2}$La$_3$Zr$_2$O$_{12}$ garnets

Stefanie Taibl$^{(a,*)}$, Andreas Wachter-Welzl$^{(a)}$, Reinhard Wagner$^{(b)}$, Daniel Rettenwander$^{(c)}$, Stefan Smetaczek$^{(a)}$, Andreas Limbeck$^{(a)}$, Georg Amthauer$^{(b)}$, Jürgen Fleig$^{(a)}$

$^{(a)}$Vienna University of Technology, Institute of Chemical Technologies and Analytics, 1060 Vienna, Austria

$^{(b)}$University of Salzburg, Department of Chemistry and Physics of Materials, 5020 Salzburg, Austria

$^{(c)}$Massachusetts Institute of Technology, Department of Materials Science and Engineering, 77 Massachusetts Avenue, Cambridge, MA 02139, USA

*E-mail of the Corresponding Author: stefanie.taibl@tuwien.ac.at

Rechargeable Li-ion battery (LiB) systems based on liquid organic (polymer) electrolytes became eminently important over the last few decades. Nonetheless, they exhibit distinct safety problems and suffer from capacity losses primarily related to degradation caused by the liquid electrolyte. Therefore, current research is focusing on all solid state Li-ion battery systems to overcome this problems. Among solid electrolytes, cubic Li$_7$La$_3$Zr$_2$O$_{12}$ (LLZO) is one of the most promising candidates to replace its liquid counterpart$^{[1]}$. This garnet-type oxide exhibits high ionic conductivity in combination with high chemical and electrochemical stability, as well as high stability against metallic lithium. However, the crucial parameters for a defined engineering of an optimized LLZO electrolyte are still under investigation.

In this contribution we report on spatially resolved electrical impedance measurements (EIS) performed on Li$_{6.4}$Al$_{0.2}$La$_3$Zr$_2$O$_{12}$$^{[2]}$. By applying Pt micro-electrodes on the samples it became possible to probe different areas of the material, allowing us to monitor highly resolved conductivity distributions. These local conductivities are compared to values from macroscopic measurements and revealed severe conductivity gradients. Locally, conductivities as high as $8 \cdot 10^{-4}$ S/cm were found compared to an overall conductivity of the pellet of $3.3 \cdot 10^{-4}$ S/cm. Hence, distinct variations in the elemental composition within one and the same sample can be supposed. Additional quantitative analysis by means of LA-ICP-MS and LA-ICP-OES lead to information on the exact local stoichiometry. Interpreting the analytical results in combination with the electrical impedance data, thus can help to receive LLZO electrolyte material with defined and reproducible properties.


$^{[2]}$ A. Wachter-Welzl, R. Wagner, D. Rettenwander, S. Taibl, G. Amthauer, J. Fleig; J. Electroceramics. in press