Synthesis and Characterization of $Li_{7-3x}Fe_xLa_3Zr_2O_{12}$ as Electrolyte in All Solid State Lithium Ion Batteries

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Conventional lithium-ion batteries consist of an electrolyte containing toxic, flammable organic liquids which lead to several safety issues such as leakage and burning of the battery. In order to overcome these safety issues solid-state electrolytes could be an alternative to obtain powerful lithium-ion batteries. Garnet type electrolytes show promising properties in terms of high lithium-ion conductivity and a good stability against lithium metal. The lithium ion conductivity of the iron substituted garnet $Li_{6.25}Fe_{0.25}La_3Zr_2O_{12}$ was found to be $1.38 \cdot 10^{-3}$ S cm^{-1}, one of the highest values for garnets so far.[1]

Objective of the work is to provide phase pure garnet type materials with cubic structure and high densities and crack free ceramics. Therefore we optimized both the synthesis of $Li_{7-x}Fe_xLa_3Zr_2O_{12}$ and the sintering based on dilatometer measurements. We focused on the density and phase purity by varying calcination and sintering time, calcination and sintering temperature and iron content. A cubic stabilization can be reached for Fe-contents with $x \geq 0.2$. Samples with lower Fe-content show partially tetragonal structures. Sintering conditions of 2 h at 1225 °C lead to stable pellets with densities up to 90 % of the single crystal density. The pellets have garnet type structure and do not show side phases indicated by X-ray diffraction. With thermal etching followed by scanning electron microscopy it is possible to make the grain structure visible. Furthermore the materials were characterized electrochemically by cycling.

References:

[1] Reinhard Wagner et al., Chem. Mater. 28 (2016) 5943-5951.