Material development for solid state electrolytes for advanced Li-ion cells

Author 1: Dr. Magnus Rohde, Karlsruhe Institute of Technology (KIT), IAM-AWP

Author 2: Dr. Carlos Ziebert, Karlsruhe Institute of Technology (KIT), IAM-AWP

Author 3: Prof. Dr. Hans Jürgen Seifert, Karlsruhe Institute of Technology (KIT), IAM-AWP

Keywords: Solid state electrolyte; Lithium ion battery; glass-ceramic; thin films; ionic conductivity

ABSTRACT

Within the current development state Li-ion batteries for energy storage with high capacity and power density liquid electrolytes are applied. These organic liquids are not thermally stable and tend to degrade at higher temperatures. Most of these liquids are flammable which can be an important security issue in high power battery systems. In contrast to the organic liquids solid electrolytes made of a glass-ceramic do not have these disadvantages. Even at higher temperatures they are very stable and do not degrade and therefore reduce the efforts of a sophisticated thermal management. However, one of the main obstacles for the application of solid electrolytes in Li-ion cells is the relatively low ionic conductivity which can be about one order of magnitude lower than in liquid electrolytes at room temperature.

Within this work three Li_2O-based systems which are candidate materials for solid state electrolytes were studied. LAGP (Li_{1+x} $Al_xGe_{2-x}(PO_4)_3$, x≈0.5) and LATP (Li_{1+x} $Al_xTi_{2-x}(PO_4)_3$, x≈0.5) glass-ceramic substrates were prepared using a melt quenching route and by applying different compaction methods, whereas thin films of the LVSO (Li_2O – V_2O_5 – SiO_2) system were deposited by non-reactive magnetron sputtering using a combinatorial materials science approach based on segmented targets. In each sputter experiment, thin films of different composition and/or microstructure were simultaneously deposited by placing the Si substrates in five individual positions relative to the segmented target consisting of two half parts of circular Li_4SiO_4 and V_2O_5 ceramics and the sputter pressure was varied between (0.5 –25 Pa).

In order to develop a better understanding of the relationship between the specific microstructure and the ionic conductivity the samples were characterized by scanning electron microscopy, x-ray diffraction, Raman spectroscopy and impedance spectroscopy.

The measured values of the ionic conductivities of the two glass ceramics were in the range of 10^{-4} to 10^{-3} S/cm at room temperature, but exhibited an increasing behaviour as a function of temperature reaching a level of the order 10^{-2} S/cm above 200 °C. Additionally, thermal analysis and measurements of the thermal transport parameters were performed in order to evaluate the thermal stability a higher temperatures and also to identify the optimum temperature range of the thermal post-processing.

Amorphous $Li_{1.2}V_{1.3}Si_{0.7}O_4$ thin films could be deposited at a pressure of 0.5 Pa that showed an ionic conductivity at room temperature of up to 6.5×10^{-5} S/cm, which is significantly higher than all values for the LVSO system or the standard Lipon (lithium phosphorous oxynitride) thin film electrolytes reported in literature up to now.

These results confirm the potential of both approaches for the development of improved solid state electrolytes for advanced Li ion cells.