

## O 16: SYLI: Interfacial Challenges in Solid-State Li Ion Batteries - Interface-dominated behaviour

Time: Monday 15:45–16:45

Location: IFW A

O 16.1 Mon 15:45 IFW A

**Interfacial double layers in all-solid-state Li-ion batteries: theoretical insights** — ●JOHANNES VOSS<sup>1</sup>, SASKIA STEGMAIER<sup>2</sup>, KARSTEN REUTER<sup>3</sup>, and ALAN LUNTZ<sup>1,2</sup> — <sup>1</sup>SLAC National Accelerator Laboratory, Menlo Park, CA, USA — <sup>2</sup>Stanford University, Stanford, CA, USA — <sup>3</sup>Technische Universität München, Germany

Neglecting potential electrochemical and mechanical stability issues of the interface between battery electrode and solid Li-ion conducting electrolyte, we present a theoretical study of charge double layers at ideal solid-solid interfaces. Based on DFT calculations for  $\text{Li}_3\text{OCl}$ , we discuss ionic charge carrier stabilities and interactions in the bulk and at interfaces to electrodes. We furthermore employ continuum models parametrized with DFT energies to understand charging at both the anode and cathode interfaces, respectively, and discuss how spacer materials could mitigate related potential interfacial issues.

O 16.2 Mon 16:00 IFW A

**Separation of Intra Grain and Grain Boundary Processes in Solid Electrolytes by High-Resolution Impedance Spectroscopy** — ●ANDREAS MERTENS, SHICHENG YU, DENIZ GUENDUEZ, HERMANN TEMPEL, ROLAND SCHIERHOLZ, HANS KUNGL, RÜDIGER-A. EICHEL, and JOSEF GRANWEHR — Forschungszentrum Jülich, Institute of Energy and Climate Research, IEK-9, D-52425 Jülich, Germany

Solid electrolytes are intensively researched for the next generation lithium-ion batteries. Yet their ion transport mechanisms must be better understood to improve their still too low ionic conductivity. Especially distinguishing between contributions from intra grain and grain boundary processes proved to be particularly challenging. This is mainly due to similar time constants of both processes at room temperature, leading to a strong overlap of their impedance contributions.

In this work we use a distribution of relaxation times analysis (DRT) in two dimensions (2D-DRT) to increase the resolution of impedance data measured at  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  (LATP) solid electrolyte samples sintered between 950°C and 1100°C. The second dimension of the 2D-DRT is given by data measured at varying temperatures between 10°C and 50°C. With the 2D-DRT it is possible to quantify the resistances and thus the activation energies of the intra grain and the grain boundary ionic charge transport processes under realistic battery operation conditions. Moreover, the ionic conductivity within the grain and the grain boundary could be determined.

O 16.3 Mon 16:15 IFW A

**Microstructural Properties of  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  Solid-State Electrolyte** — ●DENIZ CIHAN GUNDUZ<sup>1,2</sup>, ROLAND

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Lithium-ion batteries are important in daily life. One task is to replace liquid electrolytes by solid ones and  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  (LATP) is a promising candidate exhibiting high ionic conductivities. For further development it is important to understand the influence of the microstructure on the ionic conductivity. Solid state electrolytes have been mainly investigated via means of electron microscopy (Scanning Electron Microscopy as well as Transmission Electron Microscopy). Here we combine these techniques with Laser Scanning Microscopy (LSM) to study the microstructure, such as grain size, grain boundary width, porosity and secondary phase content and distribution of LATP pellets sintered at different temperatures; and draw a relation to impedance measured on these pellets.

O 16.4 Mon 16:30 IFW A

**Development of Bulk-type All-Solid-State Lithium-ion Battery Based on Phosphate Backbone Materials** — ●SHICHENG YU, HERMANN TEMPEL, ANDREAS MERTENS, DENIZ CIHAN GUNDUZ, SVENJA BENNING, ROLAND SCHIERHOLZ, FLORIAN HAUSEN, HANS KUNGL, and RÜDIGER-A EICHEL — Forschungszentrum Jülich, Institute of Energy and Climate Research, IEK-9, D-52425 Jülich, Germany

Considerable efforts have been devoted to the development of thick-film solid-state batteries while less achievement has been published owing to the poor interfacial compatibility between electrodes and electrolytes as well as low lithium ion transfer kinetics in solid materials. Here, we demonstrate the fabrication and performance of thick-film monolithic all-phosphate ceramic solid-state Li-ion batteries with high power density and cycling stability at ambient temperature based on  $\text{LiTi}_2(\text{PO}_4)_3/\text{C}/\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3/\text{Li}_3\text{V}_2(\text{PO}_4)_3/\text{C}$ . Theoretically, due to the crystal structure matchup of  $\text{LiTi}_2(\text{PO}_4)_3$ ,  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  and  $\text{Li}_3\text{V}_2(\text{PO}_4)_3$ , the solid-solid interface between  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  electrolyte and  $\text{LiTi}_2(\text{PO}_4)_3$  anode is expected to facilitate smooth contact while partially matched on the other side for the solid-solid contact of solid electrolyte and  $\text{Li}_3\text{V}_2(\text{PO}_4)_3$  cathode. Besides, the operation voltage of the electrode pair is highly matched with the electrochemical window of solid electrolyte. Excitingly, by optimizing the morphology of electrode materials and composition of each layer, the all-solid-state lithium-ion battery, which has a controllable thickness from 0.7 up to 2 mm, showed high capacity and more than 300 stable cycles.