

## SYLI 5: Structure - property relationships I

Time: Tuesday 11:45–12:45

Location: IFW A

SYLI 5.1 Tue 11:45 IFW A

**Hierarchically Structured Electrode Materials for Lithium-Ion Batteries** — •MICHAEL FISCHER, XIAO HUA, PRESTON SUTTON, and ULLRICH STEINER — Adolphe Merkle Institute, Chemin des Verdiers 4, 1700 Fribourg, Switzerland

We present a facile synthetic approach for the fabrication of hierarchically structured electrode materials for lithium-ion batteries. Amphiphilic block copolymers are known to form well-defined morphologies on a mesoscale. By confining sol-gel chemistry to one of the blocks of the polymer these structures can be transferred to various inorganic materials, including functional materials that find use in batteries. Blending the sol-gel precursor/block copolymer mixture with a homopolymer induces phase separation and leads to structure formation on a secondary (micrometre) scale. Following this method, we synthesized mesoporous microspheres of TiO<sub>2</sub> and Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub>/C as anode materials and hollow LiFePO<sub>4</sub>/C nanospheres as a cathode. These compounds suffer from intrinsically low ionic and electronic conductivities but are of great interest due to their inherent stability and safety. The aforementioned morphologies circumvent this drawback by providing shortened solid-state diffusion distances, facilitating ionic transport. In addition, we can tune the mesopore size by employing block copolymers of different chemical composition and molecular weight. Studying the relationship between pore size and electrochemical performance enables us to identify design principles for the optimization of electrode architectures.

SYLI 5.2 Tue 12:00 IFW A

**Comparison of different thin-film solid-state battery systems** — •SUSANN NOWAK, JULIANE MÜRTER, and GUIDO SCHMITZ — Heisenbergstr. 3, 70563 Stuttgart

Different systems of thin-film batteries will be compared in respect to their suitability of being used as a reasonable combination of materials in an all-solid-state battery. As the cathode lithium iron phosphate, lithium cobalt oxide and lithium manganese oxide are compared. The used electrolyte is LiPON and as the anodes tin, silicon and lithium are tested. All cells are prepared by ion-beam sputtering resulting in an overall thickness of one micrometer or less for the whole cell. Electrochemical and TEM investigations are used to evaluate the changes at the interfaces and in the electrodes during cycling and to show mechanisms of capacity loss in the system. The behavior of lithium iron phosphate is very different from the one of lithium cobalt oxide when covered with LiPON. The capacity of the LFP covered with LiPON is exponentially increasing with the temperature and reaching its theoretical value at around 70°C. In the case of all-solid-state batteries with other anodes than lithium, the capacity becomes dependent on the chosen balancing ratio of the cells.

SYLI 5.3 Tue 12:15 IFW A

**Ion transport and phase transformation in thin film LiFePO<sub>4</sub> intercalation electrodes** — •GUIDO SCHMITZ<sup>1</sup>, FABIAN WUNDE<sup>2</sup>, and FRANK BERKEMEIER<sup>2</sup> — <sup>1</sup>Institut für Materialwissenschaft, Univ. Stuttgart, Heisenbergstr. 3, 70569 Stuttgart — <sup>2</sup>formerly: Institut für Materialphysik, Univ. Münster

Thin film battery materials have an important potential to be applied in all-solid-state batteries. From the view point of physics, they offer a clearer model geometry than the usual particle-binder-compound electrodes. The phase diagram of LiFePO<sub>4</sub> predicts a phase separation in the Li intercalation. However, it is controversial whether phase separation might be suppressed in nanosystems due to elastic or kinetic constraints.

We study the intercalation kinetics of thin films under variation of the intercalation rate over more than 4 orders of magnitude. Also the layer thickness has been systematically varied. We show i) that the miscibility gap of the bulk system is essentially understood by elastic interaction, ii) that nanometric thin films clearly reveal phase separation and therefore undergo significant plastic relaxation, iii) that the grain boundaries represent fast transport paths with the electrochemical characteristics of an ion-conductor and iv) how the classic Randles-Sevcik analysis has to be modified in the case of a phase-separating material to derive proper diffusion coefficients.

SYLI 5.4 Tue 12:30 IFW A

**First-Principles Calculations on Structure and Reactivity of Amorphous LiPON** — •SABRINA SICOLO and KARSTEN ALBE — Institute of Material Science, Technische Universität Darmstadt, Jovanka-Bontschits-Str. 2, 64287 Darmstadt, Germany

Despite their increasing popularity as solid electrolytes, the structure/property relationship of Lithium Phosphorus Oxynitrides has not yet been clarified. Theoretical work offers an invaluable insight into the atomistic properties of solids, provided the availability of valid structural models. The simulation of glassy structures represents a main challenge from a computational point of view, and is further complicated by their non-trivial composition. In this contribution, a new approach to the ab-initio simulation of amorphous structures of virtually any desired composition is described. A realistic composition has been suggested by experiments recently conducted by academic partners. The defect thermodynamics of LiPON suggests its instability against metallic lithium. [1] The formation of a solid-electrolyte interphase (SEI) at this interface has been recently observed and quantified experimentally. Following up on this result, the interfacial structural and electronic properties have been investigated with a special focus on reactivity. This work does not only describe a novel approach to the simulation of a more realistic electrolyte, but also provides unprecedented insights, supported by experimental results, into its stability and reactivity under operational conditions.

[1] S. Sicolo, K. Albe, J. Power Sources 331, 382-390 (2016).