KFM 12: Holistic Structural and Safety Assessment of Lithium-ion and Post-Lithium Cells and their Materials (Modelling of Battery Materials and Degradation)

The focus session is dedicated to the characterization of microstructure, electrochemical, thermal and safety properties of Lithium-ion and Post-Lithium cells and their individual active and passive materials. This is required to obtain quantitative and reliable data, which are necessary to improve the current understanding in order to design and develop better and safer materials and cells. Potential topics include, but are not limited to electrochemical characterization techniques , thermal characterization techniques, safety testing, development of safer materials and cell designs, thermodynamic modelling of materials, modelling of thermal runaway and propagation.

Chair: Carlos Ziebert (Karlsruhe Institute of Technology)

Time: Wednesday 11:00–12:45 Location: H9

Invited Talk KFM 12.1 Wed 11:00 H9

Model-assisted Insight into Degradation of Li-Ion Batteries during Thermal Abuse — •Ulrike Krewer, Leon Schmidt, and Jorge Valenzuela — Karlsruhe Institute of Technology, Institute for Applied Materials -Electrochemical Technologies, Karlsruhe, Germany

The electrolyte in Li-ion batteries is inherently thermodynamically instable; this leads to formation of the solid-electrolyte interphase and capacity loss. Exposing batteries to high temperatures above ca. 60°C accelerates interphase growth, but also leads to its dissolution and renewed formation. If the related exothermic heat is not sufficiently fast removed, this leads to self-heating and a thermal runaway of the cell. This talk uses modelling to give a deep insight into the processes and properties causing self-heating and thermal runaway of Li-ion batteries. A complex interaction of exothermic and endothermic reactions is revealed, and the effects of evaporative cooling [1], conditions during battery manufacturing and battery age. [2] Gas analysis with online electrochemical mass spectrometry aids in identifying the network and further sensitivities. [3]

[1] Baakes F. et al., J. Power Sources, 2022, 522, 230881 [2] Baakes, F. et al., Chem. Sci, 2023, 14, 13783 [3] Bläubaum, L. et al., Batter. Supercaps 2024, 7, e20230053.

KFM 12.2 Wed 11:30 H9

computational investigation of lignin based anode materials for Li- and post-Li ion batteries* — •jafar azizi[1], Holger Euchner[2], and Axel Gross[1] — [1]Institute of Theoretical Chemistry, Ulm University, 89069 Ulm, Germany — [2]2 Institute of Physical and Theoretical Chemistry, Tübingen University, 72076 Tübingen, Germany

Post-lithium ion batteries have gained a lot of attention as a promising energy storage technology for large-scale grid applications due to their high energy density and low cost. Hard Carbon is one of the most promising anode materials, but is still associated with some performance problems. It is anticipated that improved hard carbon-based cells with a higher energy density and better electrochemical performance will be highly interesting in electrochemical energy storage. Hence, in this work, we propose a novel anode material based on the structure of lignin, one of the most prevalent biomass materials. Based on ab initio molecular dynamic (AIMD) simulations and density functional theory (DFT) calculations we study the formation of lignin-based hard carbon at different temperatures which results show a promising new amorphous structure. We find a noticeable structure stability, and higher energy capacity (AMC_n, $n < 6$) compared to the usual graphite system.

KFM 12.3 Wed 11:45 H9

Stability Enhancement of Cubic $CsSnCl_3$ as Solid Electrolyte - A Computational Approach — •Johannes Döhn[1], Martin Uhrin[2], and Axel Gross[1,3] — [1]Institute of Theoretical Chemistry, Ulm University, Germany — [2]Multidisciplinary Institute in Artificial Intelligence, Université Grenoble Alpes, France — [3]Helmholtz Institute Ulm, Germany

For the transition towards renewable energy systems, efficient and reliable technologies for energy storage are needed. Batteries are one of the most widely used storage devices, but current technology based on the transfer of Li-ions faces several challenges including their dependence on critical materials with respect to both, scarcity and toxicity.

In our contribution, we will present atomic-scale investigations of potential future battery materials carried out using a combination of density functional theory (DFT) and machine learning interatomic potential (MLIP) calculations. We employed a high-throughput approach in order to evaluate potential dopants for the well-known Cl-ion conductor $CsSnCl_3$; a solid electrolyte material for chloride ion batteries (CIBs) which is ascribed the capability to fully exploit the potential of this alternative battery type. The investigated dopants where chosen based on a dual doping strategy: Cation doping aims at enhancing the stability of the material while the introduction of mobile species, i.e., Cl vacancies/interstitials, balances the formal charge of the system and aims at improving the Cl-ion conductivity.

KFM 12.4 Wed 12:00 H9

Stability of $MgSc_2Se_4$ Surfaces — •Sebastian Utz[1] and Axel Gross[1,2] — [1]Ulm University, Ulm, Germany — [2]Helmholtz Institute Ulm, Ulm, Germany

Magnesium metal anodes are a promising material for post-lithium battery systems because of their high theoretical gravimetric energy density. One issue that hinders their application is the low ionic conductivity of many magnesium electrolytes. One of the few electrolytes that show good magnesium ion conductivity is the solid electrolyte $MgSc_2Se_4$ with a spinel structure. While the structural and diffusion properties of the bulk material are already well understood, its surfaces and interfaces are hardly explored. To shed some light on the surface properties of this solid-state electrolyte, first principles calculations within the framework of periodic density functional theory were conducted. The stability of low index {100} and high index {222} surfaces are being compared. Additionally, common structural features of the different surfaces that may lead to a stabilisation or destabilisation will be explored.

KFM 12.5 Wed 12:15 H9

Hybrid Interfaces in Focus – Decoding the Berlinite Surface with a Synergetic NMR-DFT Approach — •Javier Valenzuela Reina[1], Vera Barysch[2], Simone Köcher[2,1], and Christoph Scheurer[1,2] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]Institute of Energy Technologies (IET-1), Forschungszentrum Jülich GmbH

One of the milestones in the development of the next generation of high-performance lithium batteries is the understanding and improvement of hybrid electrolytes and their interfaces. Nuclear magnetic resonance (NMR) spectroscopy is a non-destructive, powerful technique for unraveling the intricate interface structure and ion dynamics in these materials.

We exploit the synergies between NMR experiments and density-functional theory (DFT) simulations for investigating Berlinite ($AlPO_4$) as a model for the surface of the well-known solid ion conductor $Li_{1+x}Al_xTi_{2-x}P_3O_{12}$ with $0 \leq x \leq 1$ (LATP), a promising candidate for a hybrid electrolyte. By supporting surface-selective NMR techniques such as cross-polarization (CP) and transfer of populations in double resonance (TRAPDOR) on $AlPO_4$ powder with DFT calculations of NMR observables, we study multiple surface models, infere structural characteristics of the sample, and study its interactions with water as well as organic molecules. We demonstrate that the joint experimental-theoretical approach holds future potential for understanding and improving materials whose performance relies on the properties and behavior of complex organic/ceramic interfaces.

KFM 12.6 Wed 12:30 H9

Pits and Traps in the Impedance Analysis of Ionic Conductors — •Janis K. Eckhardt — Center for Materials Research (ZfM), Justus Liebig University, Giessen D-35392, Germany

The development of innovative electrochemical storage systems, such as solid-state batteries, is critical for achieving climate neutrality and sustainability goals. Several hurdles must be overcome before such technologies are ready for the market. Impedance spectroscopy is a powerful method for characterizing the electrical transport properties of new materials and for monitoring systems in operation. Although it is an older measurement technique, the interpretation of data for inhomogeneous solid-state systems lacks established concepts for reliable results. Therefore, we use 3D electrical network models for spatially-resolved transport simulations and systematic investigation of the influence of sample microstructure and solid-solid interface morphology. The material-specific transport quantities derived from 1D models (e.g., brick layer model) sometimes exhibit inaccuracies of several orders of magnitude. In addition, the impedance response of the system exhibits geometric signatures that cannot be adequately represented in physically motivated circuit models, e.g., current constriction phenomena.