CPP 16: Energy Storage Materials and Devices II

Time: Monday 17:15–19:00 Location: ZEU/0260

Invited Talk CPP 16.1 Mon 17:15 ZEU/0260

Understanding the Passivation Properties of Solid Electrolyte Interphases (SEIs) in Batteries: Generator-Collector Experiments combined with a Transport and Reaction Model — •Bernhard Roling, Falk Krauss, Annalena Duncker, and Isabel Pantenburg — Department of Chemistry, University of Marburg, Germany

The solid electrolyte interphase (SEI) is a passivation layer in lithium-ion batteries, which has a strong impact on the battery lifetime. The SEI is formed by reductive decomposition of the electrolyte at the graphite particles of the anode during the first charging of the battery. The passivation of the SEI is not perfect, which leads to battery aging and to a reduced battery lifetime. The transport and reaction mechanism in the SEI governing the passivation properties are not well understood. Here, we elucidate transport and reaction processes during the formation of model-type SEIs by combining generator-collector experiments with a transport and reaction model. In the generator-collector experiments, we use a four-electrode-based setup to compare the electrolyte reduction current with a redox molecule reduction current at the SEI-covered electrode. We find that the current ratio depends on the SEI formation potential as well as on the formation time. The experimental results are compared to a transport and reaction model predicting four distinct transport and reaction regimes depending on the rate constant for the molecule-electron reaction. Using this combined approach, we obtain good estimates for the transport coefficients of both electrons and molecules inside the SEI.

CPP 16.2 Mon 17:45 ZEU/0260

Insight into SEI formation and dendrite growth in all-solid-state lithium metal batteries by Operando nWAXS — •Yingying Yan[1], Liangzhen Liu[2], Yuxin Liang[1], Lyuyang Cheng[1], Fabian A.C. Apfelbeck[1], Guangjiu Pan[1], Anton Davydok[3], Christina Krywka[3], Roland A. Fischer[2], and Peter Müller-Buschbaum[1] — [1]TUM School of Natural Sciences, Chair for Functional Materials, Garching, Germany — [2]TUM School of Natural Sciences, Chair of Inorganic and Metal-Organic Chemistry, Garching, Germany — [3]Helmholtz-Zentrum Hereon, Max-Planck-Str. 1, Geesthacht, Germany

Composite solid polymer electrolytes (CSPEs) have emerged as promising candidates for next-generation all-solid-state lithium batteries (ASSLBs). However, the limited ionic conductivity and poor interphase stability have seriously hindered their practical application. Herein, the anion-trapping layered double hydroxide (LDH) was introduced to enhance ionic conductivity and interfacial compatibility in poly(ethylene oxide) (PEO)-based CSPEs for stabilizing lithium metal anodes. Importantly, operando nano-focus Wide-angle X-ray scattering (nWAXS) was employed to spatially map the distribution of both the solid electrolyte interphase (SEI) layer and lithium dendrites across micron-scale lateral and vertical dimensions. Therefore, this study provides a novel strategy for designing high-performance CSPEs with stabilized interfaces and establishes an innovative approach for real-time visualization of SEI evaluation and dendrite growth in ASSLBs.

CPP 16.3 Mon 18:00 ZEU/0260

Temperature effects on the electrochemical behavior of graphite cathodes for aluminium-polymer batteries — •Mohammad Mostafizar Rahman, Mohammad Jony Rana, Shuvrodev Biswas, Amir Mohammad, Hartmut Stöcker, and Dirk C. Meyer — Institute of Experimental Physics, TU Bergakademie Freiberg, Leipziger Str. 23, Freiberg, 09599, Germany

This study assesses the performance of Aluminium-polymer batteries at variable temperatures between $-20\,^\circ$C and $80\,^\circ$C. Pouch cells were constructed utilizing a solid polymer electrolyte (SPE) coupled with two different types of graphite cathode: mesocarbon microbeads (MCMB) and spherical graphite (SpG). Cyclic voltammetry and galvanostatic cycling with potential limitation measurements were conducted at different temperatures combined with variable current densities (25...100 mA/g). The results reveal that the resistance degradation below $0\,^\circ$C is reversible, with performance being recovered when returning to positive temperatures. However, at moderately high temperatures above $60\,^\circ$C, the degradation becomes permanent. MCMB cells operate effectively between $-10\,^\circ$C and $40\,^\circ$C, while SpG cells function from $0\,^\circ$C to $60\,^\circ$C. The capacity of MCMB cells peaks at $20\,^\circ$C at all current densities and then drops significantly at $60\,^\circ$C. In contrast, SpG cells exhibit their highest capacities at $40\,^\circ$C or $60\,^\circ$C, followed by a rapid decline. Together with the good Coulombic efficiencies $> 95\,\%$ both types of cells are proven suitable for varying environmental conditions.

CPP 16.4 Mon 18:15 ZEU/0260

X-Ray Absorption Near-Edge and Photoemission Spectroscopy of Deintercalation and Redox Reactions in Alluaudite-Type Na-Ion Cathodes — •Neama Imam[1], Karsten Henkel[1], Stephan Werner[2], Christoph Pratsch[2], Anna Milewska[3], Janina Molenda[3], Jan Ingo Flege[1], and Ehrenfried Zschech[1] — [1]Brandenburg University of Technology Cottbus - Senftenberg, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie, BESSY II, Germany — [3]AGH University of Science and Technology, Krakow, Poland

We have investigated alluaudite-type $\mathrm{Na}_{2.5-x}\mathrm{Fe}_{1.75}(\mathrm{SO}_4)_3$ cathodes that exhibit a three-dimensional open-channel framework with multiple Na sites. Using spatially resolved X-ray absorption near-edge spectroscopy (XANES), performed within a transmission X-ray microscope (TXM), and lab-based X-ray photoelectron spectroscopy (XPS), we have examined Na^+ deintercalation and $\mathrm{Fe}^{2+}/\mathrm{Fe}^{3+}$ charge compensation at different selected charging states. This combination of bulk- and surface-sensitive methods allows correlating Na^+ depletion with Fe^{3+} formation, potentially identifying nanoscale redox heterogeneities and tracking changes in the local Na chemical environment. Our study builds on a broader investigation [1] that includes theoretical calculations as well as structural and electrochemical analyses.

[1] J. Molenda *et al.*, *Acta Mater.* **301**, 121582 (2025).

CPP 16.5 Mon 18:30 ZEU/0260

Understanding the Impedance Spectra of Composite Materials using a 3D Electrical Network Model — •Felix Schug[1,2], Sascha Kremer[2,3], Christian Heiliger[1,2], and Janis K. Eckhardt[1,2,3] — [1]Institute for Theoretical Physics, Justus-Liebig-University Giessen, Germany — [2]Center for Materials Research (ZfM), Justus-Liebig-University Giessen, Germany — [3]Institute of Physical Chemistry, Justus-Liebig-University Giessen, Germany

All-solid-state batteries (ASSBs) are promising candidates for next-generation energy storage systems, offering enhanced energy density and safety compared to conventional lithium-ion batteries. Electrochemical Impedance Spectroscopy (EIS) is a powerful method for determining charge transport characteristics in these systems. However, the presence of multiple phases, each with distinct transport processes, as well as complex microstructures result in complex impedance spectra that are challenging to interpret.

To address this challenge, we use a microstructure-resolved 3D electrical network model to simulate charge transport on microscopic length scales. This approach enables us to disentangle the contributions of different transport processes and microstructural features to the impedance response. Our model offers insights into the local potential and current distributions influenced by material heterogeneities, directly connecting them to impedance signals. We provide insights into the impedance of composite materials, identifying the key factors that dominate the impedance signal.

CPP 16.6 Mon 18:45 ZEU/0260

Dual-Ion AlCl_4^- / AlCl_2^+ Charge Storage Mechanism in Composite Cathode for Aluminum-Polymer Batteries — •Shuvrodev Biswas, Thomas Köhler, Amir Mohammad, Hartmut Stöcker, and Dirk C. Meyer — Institute of Experimental Physics, TU Bergakademie Freiberg, Leipziger Str. 23 09599 Freiberg, Germany

Rechargeable aluminum batteries have emerged as a promising post-lithium-ion technology. Graphite, the most widely used cathode that relies on AlCl_4^- intercalation, offers limited capacity, whereas organic cathodes, such as phenanthrenequinone (PQ), coordinate with AlCl_2^+ but suffer from limited cycling stability. Here, we design a composite (SPGPQ) consisting of spherical graphite (SPG) and PQ through melt diffusion, enabling dual-ion charge storage by coupling AlCl_4^- intercalation with $AlCl_2^+$ coordination. FTIR, XPS, *in situ* XRD, and cyclic voltammetry using polyamide-6 (PA6) electrolytes (SPE) provide clear evidence of this synergistic mechanism. Whereas the PA6-SPE limits capacity due to low ionic conductivity, the higher-conductivity polyacrylonitrile (PAN)-based SPE delivers improved capacity but shows limited cycling stability. To enhance stability, we further developed an ACPQ+SPG composite through solvent-assisted impregnation using activated carbon (AC). This composite features an identical mechanism to SPGPQ, while offering significantly enhanced capacity and stable long-term cycling.