CPP 16: Energy Storage and Batteries I

Time: Tuesday 9:30–11:15 Location: H34

Invited Talk CPP 16.1 Tue 9:30 H34

Multifunctional structural batteries — •Göran Lindbergh — KTH Royal Institute of Technology, Stockholm, Sweden

A multifunctional lightweight material combines several functions in a single material entity able to simultaneously bear mechanical loads by acting as a carbon fibre composite component while providing one or more other functionalities at the same time, e.g. storing energy. In this work, we will show functionalities coming from alkali ion insertion in carbon fibres that goes beyond the battery function, and how these functions can be combined in a single device.

The starting point for these concepts is so called structural batteries, which are multifunctional composite materials able to carry load while storing electrical energy like lithium-ion batteries. In such a battery, carbon fibres are used as the load carrying material thanks to their excellent strength and stiffness properties, but also as the active negative electrode. They also include a multifunctional matrix system, a structural battery electrolyte (SBE), the material holding the fibres together to make a solid-state load carrying material, but which at the same time is ionically conductive and chemically stable. It includes a multifunctional positive electrode, a composite electrode based on conventional ion inserting materials like lithium iron phosphate (LFP) supported by electrochemically active carbon fibres for load carrying and electrical conduction. This device can then also be used for inherent strain sensing in the material and can also be made to shape-morph with electrical control at low voltages and for harvesting energy by converting mechanical strain to electrical power.

CPP 16.2 Tue 10:00 H34

Structural response of advanced silicon-containing graphite anodes in commercial Li-ion batteries — •Tobias Hölderle[1,2], Dominik Petz[1,2], Vladislav Kochetov[2], Peter Müller-Buschbaum[1], and Anatoliy Senyshyn[2] — [1]TUM School of Natural Sciences, Chair for Functional Materials, 85748 Garching, Germany — [2]MLZ, TUM, 85748 Garching, Germany

This study explores the structural behavior of graphite anodes containing silicon additives in commercial lithium-ion batteries (LIBs) through detailed X-ray and neutron powder diffraction analyses. Silicon is increasingly utilized in advanced LIB anodes due to its potential to significantly increase energy capacity. However, its substantial volume expansion and amorphization during lithiation, complicate direct observation of lithium redistribution within the anode via diffraction techniques. Nonetheless, an indirect structural response linked to the lithiation of silicon was identified, indicated by a delayed lithium intercalation into the graphite structure. Furthermore, aging effects on high-silicon-content anodes were explored, showing the role of silicon in accelerating electrode degradation over time. These findings enhance our understanding of the complex relationship between the silicon content in the anode, structural stability, and lithium transport in advanced LIBs.

CPP 16.3 Tue 10:15 H34

XPS study of redox mechanism in Na2.5-xFe1.75(SO4)3 cathode material for high-voltage sodium-ion batteries — Neama Imam[1], •Karsten Henkel[1], Anna Milewska[2], Janina Molenda[2], and Jan Ingo Flege[1] — [1]Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus-Senftenberg, Germany — [2]AGH University of Krakow, Faculty of Energy and Fuels, Krakow, Poland

A multiplet splitting model based on the original work by Gupta and Sen has been used to track the redox mechanism and electronic structure of Na2.5-xFe1.75(SO4)3, a high-performance cathode material for sodium-ion batteries (SIBs). This high-purity, off-stoichiometric open-channel cathode material with a tailored sodium-ion distribution, synthesized using an optimized solid-state route, demonstrates a high operating voltage of ~3.8 V, surpassing the values reported for other cathode materials in the literature. X-ray photoelectron spectroscopy (XPS) was employed to analyze the evolution of the material's electronic structure at various charging potentials. Fe2p3/2 spectra decomposition using the multiplet splitting model revealed the gradual oxidation of Fe2+ to Fe3+ during sodium de-intercalation while transitioning from its pristine state with the presence of Fe2+ at the cathode surface only to the fully charged state (Na0.89Fe1.75(SO4)3 at 4.5 V). This result is consistent with the electrochemical analysis.

CPP 16.4 Tue 10:30 H34

Multipoint Anionic Bridge: Asymmetric Solvation Structure Improves the Stability of Lithium-Ion Batteries — •Tianle Zheng[1], Ya-jun Cheng[2], and Peter Müller-Buschbaum[1] — [1]TUM School of Natural Sciences, Chair for Functional Materials, Garching, Germany — [2]Ningbo Institute of Materials Technology & Engineering, Chinese Academy of Sciences, Ningbo, 315201, Zhejiang Province, P. R. China

In this study, a novel concept of multipoint anionic bridge (MAB) is proposed and proved, which utilizes anions with different sites to connect with the asymmetric solvation structure (ASS). Compared to usual solvation structures, this study uses the multifunctional groups of difluoro(oxalate)borate anion, which can connect with Li ions. By tailoring the concentration, the anion serves as a bridge between different solvated structures. In-situ techniques and simulations investigate electrolytes to draw correlations between solvation structures and reaction pathways. The proposed design demonstrates remarkable high-temperature performance on both the anode and cathode sides, enabling stable cycling of lithium cobalt oxide (LCO)||graphite (0.5 Ah, 1.0 C) pouch cell for over 200 cycles at 80 °C, and facilitating Li||MCMB and Li||lithium iron phosphate (LFP) cells to deliver stable performance for 200 cycles at 100 °C. This work paves a way to develop high-performance electrolyte systems by designing and using new multipoint anions to construct ASSs.

CPP 16.5 Tue 10:45 H34

In situ Mechanochemical Synthesis of Organic Anodes in Electrode Processing for Ultra-long Sodium Storage — •Ying Qi, Huaping Zhao, and Yong Lei — TU Ilmenau

Currently, hard carbon is commonly used as an anode material in sodium-ion batteries (SIBs). However, hard carbon anodes face several challenges, including time-consuming and energy-intensive synthesis processes, low-voltage operating platforms, and capacity storage with a steep slope. These issues contribute to severe dendrite growth and reduced full-cell capacity, negatively affecting electrochemical performance. In contrast, organic anodes are becoming a focal point in SIBs research due to their abundant availability, low cost, simple synthesis, and stable potential plateaus. In this study, we directly prepared a series of acid anhydrides including perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA), 1,4,5,8-naphthalenetetracarboxylic dianhydride (NTCDA), and pyromellitic dianhydride (PMDA) into electrodes using a one-step milling process during slurry preparation. The prepared Na4PTC electrodes show low potential plateaus and ultra-stable cycling performance in SIBs. It exhibited exceptional performance, demonstrating ultra-long cycling stability lasting nearly one year. Additionally, the assembled full cell delivered a high energy density of ~200 Wh kg-1.

CPP 16.6 Tue 11:00 H34

Localized highly concentrated electrolytes with perfluorocarbonate diluents enable high-energy-density lithium/sodium metal batteries — •Zhuijun Xu[1], Yinghui Li[2], Yajun Cheng[2], Yonggao Xia[2], and Peter Müller-Buschbaum[1] — [1]TUM School of Natural Sciences, Chair for Functional Materials, 85748 Garching, Germany — [2]Ningbo Institute of Materials Technology & Engineering, Chinese Academy of Sciences, Ningbo, Zhejiang, 315201, P. R. China

Localized high-concentration electrolytes (LHCEs) have been widely recognized for their critical role in managing the reversible plating/stripping of alkali metals. multi-functional diluents with more stable physical and chemical properties are crucial for enhancing battery performance and safety. In this study, a variety of solvents, including 4-fluoro-1,3-dioxolan-2-one, 4,4-difluoro-1,3-dioxolan-2-one, cis-4,5-difluoro-1,3-dioxolan-2-one, trans-4,5-difluoro-1,3-dioxolan-2-one, 4,4,5-trifluoro-1,3-dioxolan-2-one, and 4,4,5,5-tetrafluoro-1,3-dioxolan-2-one, were analyzed using Density Functional Theory (DFT) calculations. Further, molecular dynamics (MD) simulations were conducted on the aforementioned solvents and their corresponding LHCEs for lithium/sodium metal batteries (LMBs/ SMBs). A comprehensive set of physical and chemical properties was examined, including the density of the solvent molecules, their highest occupied molecular orbital

(HOMO) and lowest unoccupied molecular orbital (LUMO) energies, and their binding energies with lithium or sodium ions, distinct solvation structures within various LHCEs.