

## CPP 1: Energy Storage and Batteries I

Time: Monday 9:30–11:15

Location: H 0106

## Invited Talk

CPP 1.1 Mon 9:30 H 0106

**Customized Cathodes for All-Solid-State Battery Application** — ●TORSTEN BREZESINSKI — Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany

Quasi- and all-solid-state batteries are receiving much attention as enabling technologies for next-generation electrochemical energy storage. Similar to conventional Li-ion batteries, high-capacity, layered transition-metal oxides are being considered for use at the positive electrode in combination with superionic thiophosphate (sulfide) solid electrolytes. Yet, such composite cathodes suffer from degradation issues, such as (electro)chemical side reactions and/or particle fracture/pulverization, limiting their utilization. In this presentation, I will highlight the importance of tailoring layered Ni-rich oxides (regarding particle size, composition etc.) for increasing cycling performance of pelletized and slurry-cast electrodes. In addition, I will show data on the effect of protective nanocoatings on interfacial side reactions in solid-state battery cells using argyrodite Li<sub>6</sub>PS<sub>5</sub>Cl as ion conductor.

CPP 1.2 Mon 10:00 H 0106

**Real-time monitoring of electrochemical reactions in all-solid-state lithium batteries by simultaneous grazing-Incidence small-angle/wide-angle X-ray scattering** — ●YUXIN LIANG<sup>1</sup>, TIANLE ZHENG<sup>1</sup>, KUN SUN<sup>1</sup>, FABIAN A.C. APFELBECK<sup>1</sup>, IAN SHARP<sup>2</sup>, YAJUN CHENG<sup>3</sup>, MATTHIAS SCHWARTZKOPF<sup>4</sup>, STEPHAN V. ROTH<sup>4,5</sup>, and PETER MÜLLER-BUSCHBAUM<sup>1,6</sup> — <sup>1</sup>TUM School of Natural Sciences, Chair for Functional Materials, Garching, Germany — <sup>2</sup>WSI, TUM, Garching, Germany — <sup>3</sup>NIMTE, Chinese Academic of Sciences, Ningbo, China — <sup>4</sup>DESY, Hamburg, Germany — <sup>5</sup>KTH Royal Institute of Technology, Stockholm, Sweden — <sup>6</sup>MLZ, TUM, Garching, Germany

Polyethylene oxide (PEO)-based composite electrolytes (PCEs) are promising for lithium batteries due to its high safety. However, the material suffers from low conductivity, inhibiting its commercialization. Therefore, it is crucial to understand the electrochemical process in PCE-based batteries. Using operando grazing-incidence small-angle and wide-angle X-ray scattering, we find that the electrochemical reaction is highly correlated with the buried morphology and crystalline structure of the PCE. The PEO-Li<sup>+</sup> reduction and TFSI-decomposition cause changes in both the crystalline structure and morphology. In addition, the reversible Li diffusion process alters the inner morphology, rather than causing structure changes. This work provides a new path to monitor a working battery, thereby enabling detailed understanding of electrochemically-induced changes, which is essential for developing interface stable lithium batteries.

CPP 1.3 Mon 10:15 H 0106

**Introducing the LECA package for machine-learning guided optimization of the ionic conductivity** — ●MIRKO FISCHER<sup>1</sup>, HARRISON MARTIN<sup>1</sup>, PENG YAN<sup>2</sup>, CHRISTIAN WÖLKE<sup>2</sup>, ANAND NARAYANAN KRISHNAMOORTHY<sup>2</sup>, ISIDORA CEKIC-LASKOVIC<sup>2</sup>, DIDDO DIDDENS<sup>2</sup>, and ANDREAS HEUER<sup>1</sup> — <sup>1</sup>Institute of Physical Chemistry, University of Münster, Corrensstraße 28/30, 48149 Münster — <sup>2</sup>Helmholtz-Institute Münster (IEK-12), Forschungszentrum Jülich GmbH, Corrensstraße 46, 48149 Münster

We present the Liquid Electrolyte Composition Analysis (LECA) package as a versatile tool, which implements a simplified and semi-automatic workflow for data-driven and machine-learning guided analysis of large data sets, particularly designed for but not limited to High-Throughput-Experiments (HTE). The LECA package combines popular python-based libraries like scikit-learn, Mapie and GPyOpt to enable fast parallel training, hyperparameter-optimization, model comparison, and uncertainty calculation for various regression models. An active learning approach to reduce the amount of data needed to fit a model with high accuracy is under current development and testing.

We demonstrate the performance of the LECA package on a large HTE dataset for the ionic conductivity as an important bulk property of liquid electrolytes with over 200 individual compositions measured, including the organic electrolytes EC, EMC, and PC, and the lithium salts LiPF<sub>6</sub> and LiFSI. Furthermore, we show how the LECA package can be used to optimize the ionic conductivity and discover new compositions.

CPP 1.4 Mon 10:30 H 0106

**Structural engineering of two-dimensional nanosheets for sodium-ion storage** — ●YUHUA CHEN, YULIAN DONG, HUAPING ZHAO, and YONG LEI — Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, 98693 Ilmenau, Germany

Rechargeable sodium-ion batteries (SIBs), with their low cost and abundant Na reserves, have been promising candidates for energy storage and conversion. Among all cathode materials for SIBs, two-dimensional (2D) VOPO<sub>4</sub> show high energy storage potential owing to their high working voltage, stable structure, and diverse structure. Nevertheless, the electrochemical performance of VOPO<sub>4</sub> is hindered by many factors such as poor conductivity and limited intrinsic interlayer distance. Rational design and structural engineering of 2D nanosheets by intercalation chemistry, such as cationic intercalation, conductive polymer introduction, and atomic doping, tuning their physical and chemical properties, effectively enhances the storage of sodium in thin 2D nanosheets. Here, we explore intercalation chemistry for the storage of Na<sup>+</sup> ions in layered VOPO<sub>4</sub> hydrates, delivering a high average operating voltage of approximately 3.5 V. The introduction of Fe<sup>3+</sup> ions result in a noteworthy increase in specific capacity, reaching 85 mAh g<sup>-1</sup> at 0.1 C, which exhibits a commendable capacity retention rate of 62% over 50 cycles. This investigation underscores the substantial impact of Fe<sup>3+</sup> doping on the electrochemical performance of VOPO<sub>4</sub>·2H<sub>2</sub>O nanosheets, positioning them as promising candidates for high energy density SIBs.

CPP 1.5 Mon 10:45 H 0106

**Operando neutron diffraction experiments in order to investigate the lithiation and ageing mechanism of industrial scale multi-layer 5 Ah pouch cells** — ●THIEN AN PHAM<sup>1,3</sup>, STEFAN SEIDLMEYER<sup>1</sup>, IVANA PIVARNIKOVA<sup>1,3</sup>, SVEN FRIEDRICH<sup>4</sup>, ALESSANDRO SOMMER<sup>5</sup>, CHRISTOPHE DIDIER<sup>2</sup>, ANDREAS JOSSEN<sup>4</sup>, RÜDIGER DAUB<sup>5</sup>, VANESSA PETERSON<sup>2</sup>, PETER MÜLLER-BUSCHBAUM<sup>1,3</sup>, and RALPH GILLES<sup>1</sup> — <sup>1</sup>TUM, MLZ, 85748 Garching, Germany — <sup>2</sup>ANSTO, Lucas Heights, Australia — <sup>3</sup>TUM School of Natural Sciences, Chair for Functional Materials, 85748 Garching, Germany — <sup>4</sup>TUM School of Engineering and Design, Chair of Electrical Energy Storage Technology, 80333 Munich, Germany — <sup>5</sup>TUM School of Engineering and Design, Institute for Machine Tools and Industrial Management, 85748 Garching, Germany

The demands for Li ion batteries are increasing and the improvement of battery performances remains one of the main goals of the research. Currently, switching to materials offering higher materials is one approach to achieve higher capacities. Here, the lithiation mechanism and ageing behaviour of silicon anodes and LiNi<sub>0.8</sub>Co<sub>0.15</sub>Al<sub>0.05</sub>O<sub>2</sub> (NCA) cathodes in multi-layer 5 Ah pouch cells were studied with neutron diffraction. The batteries were fabricated on a research production line in order to replicate the conditions of commercial cells as close as possible. Uncycled cells were compared to an aged cell at a state of health of 60 percent. Due to ageing, the capacity is decreased, which can be seen in the evolution of the cell parameters of NCA. During relaxation the NCA peaks are shifting indicating structure changes.

CPP 1.6 Mon 11:00 H 0106

**Ionic polymer electrolyte for aluminium batteries** — ●AMIR MOHAMMAD, THOMAS KÖHLER, SHUVRODEV BISWAS, HARTMUT STÖCKER, and DIRK C. MEYER — Institute of Experimental Physics, TU Bergakademie Freiberg, Germany

Rechargeable aluminium batteries are considered a promising alternative to lithium-based energy storage systems, but their widespread use is hindered by challenges in the electrolyte system, such as leakage and corrosion. This necessitates the use of expensive metal foils (e.g. Mo, Ta, W) as current collectors on the cathode side. However, a new ionic polymer electrolyte has been developed that offers significant improvements in performance, safety, and cost efficiency. The manufacturing process involves the complexation of polyamide-6 (PA6) with aluminium chloride (AlCl<sub>3</sub>) and the organic salt triethylamine hydrochloride (Et<sub>3</sub>NHCl), allowing for the dissolution of up to 20% PA6 in the sample.

This results in good mechanical properties and a stable electrode-electrolyte interface without compromising the cell performance. The

ionic polymer electrolyte does not require separators, has a high ionic conductivity of up to  $0.3 \text{ mScm}^{-1}$ , and can effectively dissolve and deposit aluminium electrochemically. This innovation has the potential to replace expensive metal current collectors by affordable aluminium

foils with protective coatings. The solid ionic polymer presented here is a promising and robust strategy for the development of stable, safe, flexible, and yet inexpensive aluminium batteries.