Book of abstracts for BATTERY 2030+ Annual Conference 2025



Invited speaker Abstracts



Poly(ionic liquids) as a versatile family of polymers for next generation alkali metal batteries

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Polymerized ionic liquids, also referred to as poly(ionic liquid)s (Poly(ILs)), represent an expanding class of ion conductors characterized by the incorporation of cations and anions coming from ionic liquids (ILs) within polymeric structures. These materials retain the unique properties of their ionic liquid counterparts, including ionic conductivity and the ability to dissolve alkali metal salts, but with the added advantage of being in their solid state with mechanical integrity and flexibility. PolyIL-in-Salt systems, where the salt is the dominant component of the polymer electrolyte, have shown unique co-ordination and transport properties with some of these materials being demonstrated in battery cells operating at elevated temperatures with high stability. PolyILs have also been used as functional binders in various cell formats. The ability to customize both the nature of the ion and the polymer backbone, provides a large degree of versatility in chemistry and function, with the ability to tailor this for specific applications.

This presentation will discuss the influence of both chemistry and composition on the ion transport and ionic structures in these novel electrolytes, including demonstrating how MD simulations might be used to understand and predict behaviour.

BASF Battery Materials: Powering Electromobility with Innovation and Commitment

The global battery market is currently experiencing a phase of slower growth and postponed investments. BASF is strategically positioned to effectively navigate the challenges in this highly competitive environment. The company's strong global footprint is complemented by an advanced CAM technology portfolio, ranging from ultra-high-Ni to Co-free and Mn-rich solutions. The materials are designed to accommodate all major cell formats and mobility segments, with the flexibility to be tailored to meet specific customer requirements. BASF's comprehensive strategy prioritizes innovation and collaboration along the entire battery value chain. This approach not only fosters world-scale CAM production capacities but also enhances the resilience of local supply chains by integrating essential components such as mining services and recycling solutions. With a long-term commitment to the battery industry, BASF is dedicated to contributing to a robust and competitive future for electromobility in Europe and beyond.

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Selected Oral Presentation Abstracts



Multi-Hydrogen Bond-Based Self-Healing Polymer Separator for Next-Generation Lithium Batteries

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Whilst not directly involved in the battery's redox reactions, the separator plays a crucial role in the functioning of lithium batteries. It must retain the electrolyte to prevent decomposition and to ensure a stable environment for electrochemical reactions. Additionally, it contributes to battery safety by preventing the diffusion of undesirable byproducts such as lithium dendrites, the presence of which could lead to compromised performance and system integrity.¹ Also, capacity fading is a drawback associated with degradation and ageing of cell components. However, these problems can be overcome by self-healing strategies.

Self-Healing smart functionalities have attracted attention for their potential use in next generation LIBs. The development of self-healing materials that can be employed in different battery compartments represents a significant advancement. This paves the way for innovative strategies that can exploit higher intrinsic durability, prolonged lifetime and safety.² This study was focused on the realization of a Self-Healing Separator (SHS) based on Ureidopyrimidinone (UPy) moieties introduced into Polyethylene-*co*-2-hydroxyethyl Methacrylate (PEHEMA) bearing hydroxyl groups by solution grafting, affording physically crosslinked PE via supramolecular interactions.³ To modulate the mechanical properties, wettability, and electrolyte diffusion of the realized self-healing polymer, it was mixed with a polyethylene oxide with a molecular weight of 300 kDa. Both stand-alone PEHEMA-UPy and PEHEMA-UPy_PEO were fully tested. The objective is to identify the separator that exhibits the optimal characteristics for implementation in a next-generation lithium battery. In half-cell tests 1M LiFSI in EC:DMC (50:50) was employed as electrolyte.

The SHS were prepared by hot-press method in a fast and easily implementable procedure. The autonomous self-healing ability was evaluated by following the repairing process of fractures caused throughout the whole film thickness that occurs in 3hours at 40°C. Moreover morphology, composition (SEM, IR), thermal, mechanical stability (TGA-DSC, DMA), and electrochemical stability window (LSV) of the SH separators were investigated. The electrochemical tests were carried out at 25°C on symmetrical Li|SHS|Li cells at a current density of 2 mA cm⁻², which showed a cycle life of over 600 hours with low overpotential and excellent cyclability. The performance of the SHS was tested through galvanostatic cycling in half-cells, with LFP as the cathode, in term of long-term stability test with a Crate of 1C (IDensity: 170 mA g⁻¹) for 1000 cycles maintaining a capacity retention of 81.5%. Accelerated rate calorimetry (ARC) analysis was finally performed to demonstrate the robustness of the realized SHS also in terms of cell thermal stability. For the sake of comparison, all the tests were also performed with a Celgard2500 benchmark as separator. A polymer separator with self-healing properties, good mechanical strength, and high potential stability was developed for use in LIBs. The autonomous self-healing ability was confirmed by tests before and after damage, which showed a good recovery both in terms of mechanical and electrochemical performances. The realized separator's structure and composition effectively suppress dendrite growth and were promising in terms of safety, stability and recovery capability over time, due to its self-healing properties.

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The role of fluorination in the development of novel salts for nextgeneration lithium-ion batteries



Figure 1: The design of novel fluorinated lithium salts for battery application

Lithium salts play a vital role in determining the performance, safety, and cost of rechargeable battery electrolytes, either as the main component or an additive. However, depending on the battery systems and operating conditions, the design requirements of electrolytes may change significantly. Modern battery electrolyte design has demonstrated that fluorination through the application of perfluorinated chemicals is key to significantly improving the thermal safety and stability of high-energy-density lithium-ion batteries.^[1] However, the perfluorinated substances (PFAS) also raise a significant concern about their impact on health and the environment. Therefore, the knowledge of the required extent of fluorination in lithium-ion batteries is critical to minimise its environmental impact without compromising the battery performance. The versatility of boron chemistry allows freedom in designing lithium salts with customisable fluorination of anion structures that provide the desirable combination of non-flammability, electrochemical stability, and interface-forming capabilities in high-voltage lithium-ion batteries. In this talk, I will discuss the design principles of these lithium fluoroborate salts as the electrolyte component for the next-generation fast charging and high-voltage Li-ion batteries (LMO, NMC and LNMO)^[2]. This talk will also provide insight into how the synthesis of novel salts allows the development of next-generation high-performance, sustainable electrolytes for lithium-ion batteries.

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Utilizing Solid-state NMR towards Understanding the Structure and Ion-Dynamics in Inorganic Solid Electrolytes

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Inorganic solid electrolytes (SEs) play a crucial role in the development of all-solid-state batteries with the promise of improved safety and energy density. Designing SEs with desired ion transport and electrochemical performance requires an in-depth understanding of the underlying structural complexity and its influence on the ion-dynamics on micro- and macroscopic scale. Solid-state NMR is a powerful technique utilized towards understanding the local structure and ion-dynamics (kHz-MHz scale) in such inorganic SEs. Here, we present some of our recent works involving structural elucidation and observing cation-dynamics in different class of solid inorganic SEs *e.g.* halides, sulfides, oxychlorides, argyrodites.

Nuclei-specific structural features are observed by performing solid-state magic-angle spinning (MAS) NMR at variable temperatures. A combination of the right nuclei and appropriate spinning frequency provides distinct structural information for Li⁺ and Na⁺ conducting SEs. For Li⁺-conductors, ⁶Li MAS NMR have differentiated between tetrahedral and octahedral Li-environment in Sn-substituted Li₃SbS₄ [1], and tracked the phase evolution in In-substituted Li₃YCl₆ as a function of In-content [2]. Similarly, ²³Na MAS NMR technique helped to distinguish between symmetric octahedral and distorted prismatic Na⁺-coordination in Na₃InCl₆ and the absence of the octahedrally coordinated Na⁺ in structurally related NaTaCl₆ [3], and to observe single (for Nb) vs. dual (for Ta) Na⁺-environment in Na*M*(=Nb,Ta)OCl₄ compounds [4].

Cation dynamics on a wide frequency scale are investigated using appropriate NMR experimental

technique and corresponding activation barriers are calculated from variable temperature (VT) measurements. Slower ion-dynamics in kHz scale are tracked with static VT ⁷Li measurements, as eminent from motional averaging based line narrowing for Li₆PS₅I. Faster Li⁺-jumps in MHz scale are accessible in spin-lattice (T₁) relaxation experiments, where the appearance of the rate curve is strongly influenced by the difference in NMR short- and long-range activation barriers [5]. Furthermore, exchange of cation between two distinct crystallographic sites are observed with two-dimensional exchange spectroscopy (EXSY), as presented for Na₃InCl₆.

Thus, the use of solid-state NMR is proven to facilitate a deeper understanding of the complex interplay between diverse structural features and the resulting ion transport in inorganic SEs, elevating the logical designing of improved SEs for future battery technologies.



Figure 1. Schematic showing applications of solid-state NMR in inorganic solid electrolyte research.

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Defect Structure Engineering for Next Generation Battery Materials

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Materials research is the key to innovation in materials advancements for next generation batteries. With increasing energy demand, new awareness of environmental benignity, and new emerging energy, transportation and information applications, the need for novel and specially engineered battery materials is growing at an accelerated pace, while the ability to quickly adapt to the needs of different applications and requirements persists. IET-1 has a long-term expertise in developing new advanced battery materials tailored to specific applications and bringing them to the cell level, while at the same time focusing on in-depth and advanced analytic techniques to understand and develop these approaches further. In this talk, we will present examples of defect engineering strategies for next generation battery materials for solid-state, Li-ion and Naion batteries. One of the basic principles for tuning the material properties is the control of phase fractions and, especially for low crystalline materials like Li₁₀GeP₂S₁₂ the corresponding defect structure i.e. amorphous/crystalline fractions. We show how the underlying mechanisms of the phase transformations are obtained by quantifying the phase fractions and amorphous fractions from X-ray data and how they correlate with electrochemical performance.^[1,2] Another popular defect engineering strategy is cationic doping. By studying the average and local structural motifs, we address the Ni-stabilizing effects in the Fe-rich high-voltage spinel lattice and gain further understanding to advance high-voltage cathodes for Li-ion batteries with more stable cycling performance.^[3,4] P2 layered oxides for Na-ion batteries are prone to undergo several phase transitions during cycling that lead to microstructural degradation of the cathode active material and the cathode composite. An anionic doping strategy is explored experimentally and by ab-initio calculations for halide doping in Fe-Mn rich P2 layered oxides where doping with F proves successful for stabilizing P2 layered oxides by stabilizing their lattice and mitigating phase transitions.^[5] These examples of advances in knowledgebased materials development will demonstrate how defect-structure engineering can help tailor and advance higher performance, more sustainable, and highly flexible materials for next-generation battery materials.

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Selected Poster Pitch Abstracts



Electrochemical and physicochemical characterization of biomassderived anodes for potassium-ion batteries

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As the global community confronts the crisis of climate change, the urgency for the transition to renewable energy sources becomes increasingly evident. A critical aspect of this transition involves coupling energy storage systems with renewable energy production. However, the reliance on lithium-ion batteries (LIBs) poses significant challenges due to concerns regarding lithium scarcity, environmental impact, and escalating raw material costs. In this context, potassium-ion batteries (KIBs) have emerged as a promising alternative for large scale applications due to the high natural abundance, cost-effectiveness, and favorable electrochemical properties of potassium [1]. Consequently, it is crucial to develop performing electrodes suitable for this application. From this perspective, hard carbon materials derived from biomass are particularly attractive as KIB anodes, offering a sustainable solution with adequate structural and electrochemical properties [2]. Our research team investigated the electrochemical performance of hard carbons obtained from lignin-rich biomass residues, chemically activated with different KOH ratios. Structural, morphological, and surface characterizations were conducted using XRD, SEM, BET, Raman spectroscopy, and XPS analysis to elucidate the influence of activation parameters on porosity, graphitization degree, and interlayer spacing. The charge/discharge tests demonstrated that the materials with a KOH-to-biomass ratio equal to 1 exhibited higher specific capacities and superior cycling stabilities, so the study shifted focus to the in-depth characterization of these materials. Various techniques such as EIS, XPS, and CV were employed to shed light on SEI chemical composition and formation mechanism, while the pseudocapacitive contribution to the charge storage has been evaluated thanks to the analysis of the VSCV with Dunn's method. As a result, we achieved cells with excellent long-cycle performance, utilizing a renewable source and aligning with circular economy principles.



Figure 1. a) Schematical representation of the KIB anode production process and b) investigation on the contributions to the charge storage.

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This research received funding from "Network 4 Energy Sustainable Transition"—NEST—Piano Nazionale di Ripresa e Resilienza (PNRR), M4C2 Spoke 6—NextGenerationEU



4D Microscale Modelling of Lithium-Ion Battery performance degradation by SEI Growth

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Lithium-ion batteries represent the state-of-the-art in portable energy storage due to their high energy and power density. However, their long-term application is hindered by ageing mechanisms such as Solid Electrolyte Interphase (SEI) growth, which leads to capacity fade and performance degradation. The SEI is a heterogeneous layer produced on the anode surface during the initial cycles of the cell through multiple simultaneous side reactions that involve electrolyte components and lithium ions. Due to its nanometric scale, sensibility to air and compositional variability, experimental SEI characterization remains challenging [1], making computational models a valuable alternative. Nevertheless, continuum models frequently rely on simplified geometries and global parameters (i.e. tortuosity, porosity), neglecting the effect of the porous

In this work, a 4D (geometry resolved and time-transient) microscale model developed in COMSOL Multiphysics is used to simulate SEI growth in a half-cell during the first charge-discharge cycles. The half-cell is composed of a lithium foil as reference electrode, graphite as active material of the working electrode, and 1 nm thin film at the graphite/electrolyte interface to represent the initial SEI layer. The film growth is driven by a parasitic reaction occurring alongside the primary lithium intercalation reaction, leading to lithium consumption and film deposition, represented exclusively by its density and molar mass without considering its detailed chemical composition. The kinetic formulation is based on the work of Pannala et al. [2], which assumes a linear concentration profile of the solvent across the SEI layer. By integrating this assumption with the Tafel equation, the model identifies two primary mechanisms governing SEI growth: the diffusion of solvent molecules through the SEI layer and the reaction rate of the solvent with electrons at the graphite surface. This approach allows the investigation of local phenomena leading to heterogeneity in electrical potential and lithium concentration, and the identification of preferential regions for SEI growth as shown in Figure 1.



Figure 1. Contour plot of local SEI thickness on the electrode active material.

ACKNOWLEDGEMENT

electrode heterogeneity on the growth process.

This work was funded by the European Commission within the Horizon Europe research and innovation programme through the GA no 101137725 (BatCat).

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Automation of High Throughput Materials Synthesis and Cell Testing for Lithium and Sodium Ion Batteries

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Next-generation battery materials remain an exciting and rapidly developing area of research, however, it is not typically possible to test a wide variety of materials and chemistries simultaneously. Materials synthesis and cell assembly are traditionally time-intensive processes and due to human error and inconsistencies during cell assembly, there is often variation between data-sets.

The DIGIBAT facility at Imperial College London provides tools to automate both material synthesis and cell assembly, removing these roadblocks. FULL-MAP aims to utilise these advantages, in conjunction with machine learning, to fast-track testing a wide range of battery chemistries and accelerate cell optimisation.

This work explores electrode materials and cell compositions for both Li-ion and Na-ion batteries, resulting in reproducible data across a range of factors. Electrode processing and methods of optimisation are explored, compared to standard systems and differences in cell assembly methodologies are discussed.



Cycle Life Enhancing Organic Compounds as Electrolyte Additives for SiO_x-Containing Lithium Ion Batteries

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Electrolyte additives play an important role in the formation of a robust solid electrolyte interphase (SEI), which is particularly crucial for dealing with the large volume changes of silicon-containing anodes in high-energydensity Li ion batteries. In recent years, molecular electrolyte additives such as dioxazolones,^[1,2] *O*-Carboxyanhydrides (OCAs),^[3] and *N*-Carboxyanhydrides (NCAs)^[4,5] have been introduced to enhance SEI robustness in LIBs. Herein, a novel and easily synthesizable compound (DOB) is presented as an electrolyte additive for NCM 523 || 10%SiO_x-graphite lithium ion cells. The effects of DOB on SEI formation, long-term cycling performance, self-discharge, gassing behavior, and interphase chemistry are investigated.



Figure 1. Illustration of electrochemical performance differences of DOB-derived and FEC-derived SEI.

Cells with DOB as an additive have longer cycle life than those with the state-of-the-art electrolyte additive, fluoroethylene carbonate (FEC), which can be attributed to the robustness of the DOB-derived SEI. Nevertheless, the instability of DOB-derived SEI at high temperatures and high states of charge results in faster self-discharge and increased gassing compared to cells with FEC. This study highlights the critical role of electrolyte additive design and the need for comprehensive additive evaluation that goes beyond improving cycle life to also address self-discharge and gas generation.

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Enhancing the Performance of Aluminium-Graphite Dual Ion Batteries: Comparison of Cathode Composition, and Electrolyte Molar Ratio, in Laboratory and Pouch Cell Formats

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Aluminium–graphite dual ion batteries (AGDIBs) are a promising solution for rechargeable energy storage, utilizing graphite as the cathode active material and aluminium foil as the anode substrate, with chloroaluminate ionic liquids like [EMIm]CI:AlCl₃ providing excellent charge-discharge performance and cyclic stability [1]. However, the scalability of AGDIBs is hindered by the corrosive nature of the electrolyte. Traditional current collectors such as Copper, stainless steel, and Nickel exhibit poor stability in these electrolytes, necessitating the use of Molybdenum (Mo).[2] Conventional binders like polyvinylidene fluoride (PVDF) also exhibit instability, leading to undesirable side reactions. These challenges have limited reports on electrode optimisation, particularly in cells relevant to application such as pouch cells.

This study aims to address this gap by investigating the performance of graphite cathodes, coated onto 25 µm Mo foil and with alginate as the binder in concentrations ranging from 1% to 4%, with a focus on their influence on cathode stability and ease of handling. The effect of the mass loading of graphite (5 to 25 mg cm⁻²) on the overall rate capability of AGDIBs showed that 10 mg cm⁻² yielded balanced performance with higher aerial capacity (0.7 mAh cm^{-2}) compared to the lower loadings (0.4 mAh cm^{-2}) and better specific capacity at 8C (40 mAh g^{-1}) compared to higher loadings (10 - 25 mAh g⁻¹). The type of natural graphite (flaky graphite and spherical graphite) was also evaluated, with spherical graphite showing a better performance with 50 mAh g⁻¹ at 10 C compared to 30 mAh g⁻¹ by flaky graphite. Alongside the role of carbon black (1% to 5%) in enhancing cathode performance in AGBs with 1:1.5 [EMIm]Cl:AlCl₃ was analysed. Furthermore, as the graphite loading increases from 5 to 25 mg cm⁻², the impact on the aluminium anode becomes more significant due to the proportional increase in the aerial current density on anode and overall cell performance was assessed in both laboratory cell EL-CELL® (ECC-Aqu, φ 18 mm electrode) and pouch cell (7.5 x 8 cm²) configurations, utilizing [EMIm]Cl:AlCl₃ as the electrolyte. Another critical limitation in AGDIB performance arises from the electrolyte itself, as the charge-discharge cycle is either terminated upon depletion of $AlCl_4^-$ ions, which halts the intercalation process in the cathode, or upon the depletion of $Al_2Cl_7^-$ ions result in the cessation of AI deposition at the anode. [3] Between 1:1.5 [EMIm]CI:AICI₃ and 1:2 [EMIm]CI:AICI₃, the concentration on Al₂Cl₇⁻ and AlCl₄⁻ differs significantly and their impact on the AGDIBs in rate capability (upto 10C) and long-term cycling at 30C are quite significant where the cell with 1:2 [EMIm]Cl:AlCl3 shows excellent performance. The role of cathode composition and the molar ratio of [EMIm]Cl:AlCl₃ on the AGDIB performance was characterized by in-depth electrochemical characterization like cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), constant current (CC) charging and discharging in both laboratory and pouch cell formats.

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Figure 1: Optimized coating for cathode sheets for AGBs

COMBINED OPERANDO AND POST-MORTEM CHARACTERISATION OF LITHIUM-ION BATTERIES

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Lithium-ion batteries (LIBs) have gained widespread popularity due to their high energy density and efficiency, which has led to their extensive use in portable electronic devices and electric vehicles. However, the phenomenon of battery degradation poses a significant challenge, resulting in several drawbacks. These include reduced energy storage capacity, diminished power performance, shortened lifespan, and increased safety risks. Consequently, it is imperative to understand the degradation mechanisms of LIBs to optimise their performance, enhance safety, extend service life, and develop more robust battery management systems.

This study presents a dual approach to investigate LIB degradation, combining real-time electrochemical techniques with post-mortem analyses. Electrochemical Impedance Spectroscopy (EIS) was implemented to monitor performance changes during cycling, while complementary post-mortem methods were used to assess morphological and compositional variations in the battery different components at different degradation stages.

A notable feature of this work is the implementation of an advanced EIS analysis technique, enabling more precise characterization of interfacial processes without invasive procedures. Through these analyses, capacitance dependence on the frequency was assessed. Patterns in the capacitance were identified and associated to the battery lost capacity, and associated with slower processes such as lithium-ion diffusion and intercalation.

Post-mortem characterisation revealed significant morphological and compositional alterations in the electrodes, particularly at the anode interface, the formation and growth of surface layers that influence ion transport and overall efficiency. Electrolyte degradation was confirmed by semi-quantitative analysis, particularly in the anode, where an increase in organic compounds was observed. These findings highlight the importance of interfacial stability and electrolyte integrity in maintaining battery performance over extended use.

The combined operando and post-mortem characterisation approach yielded valuable insights into the degradation of LIBs, emphasising the critical role of interface growth and electrolyte decomposition, particularly under fast charge/discharge and overcharge conditions. While no major structural changes were detected in the electrodes, notable morphological and compositional alterations were observed, especially in the anode. This approach paves the way for improved battery management strategies, ultimately enhancing both performance and safety.



Powering the Future Without PFAS

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Per- and polyfluoroalkyl substances (PFAS) are a diverse class of >10 000 anthropogenic chemicals containing at least one -CF2- or -CF3 moiety (with a few exceptions).[1] Structures range from high molecular weight fluoropolymers such as polytetrafluoroethylene (PTFE) to relatively low molecular weight, water-soluble acids such as trifluoroacetic acid (TFA). Despite their diversity, all PFAS are either highly persistent or break down to persistent end products.[2] Although some have been linked to adverse health effects, hazard data are lacking for most PFAS.

Due to their unique properties and structural diversity, PFAS have found widespread use in society. One example is their uses in lithium-ion batteries (LIBs). Our recent review of PFAS uses in LIBs found that they are prevalent in electrodes and binder, electrolyte (and additives), and separator, see Figure 1.[3] In the cathode, for example, polyvinylidene fluoride (PVDF) is considered the gold standard binder today due to its excellent electrochemical stability and processing properties, among others. However, potential formation and/or emission of PFAS to the environment is of risk during LIB recycling, in addition to the risks of human exposure during LIB manufacturing. With increased activity in hydrometallurgical recycling processes because of its ability to recover Li, the use of PFAS in LIBs will lead to emissions of PFAS. Our work highlighted the need to 1) assess the emissions of PFAS from LIB recycling and 2) development of new materials based on a design-for-recycling concept.

In the present works, we assessed the currently available PFAS alternatives that could be used in LIBs.[4] We also adopt the concept of essential use when assessing the uses of PFAS in batteries, as well as apply the functional substitution framework to look beyond drop-in substitution as an alternative.[5][6] Preliminary results indicate that powering the future without PFAS can be achieved. However, the timeframe of substitution in individual components remains highly uncertain. In addition, within a wholistic sustainability assessment, it needs to be demonstrated that PFAS replacement will not lead to burden shifting. The need for innovation enabled by eminent researchers in academia and industry alike is crucial, both in providing advancements with novel materials as well as designing batteries for various uses with differing requirements.



Figure 1. Examples of per- and polyfluorinated materials in (lithium-ion) batteries (adapted from [3]). **REFERENCES**

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Estimation of battery temperature through impedance measurements and novel application of DRT- A Sensor-less approach

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Monitoring thermal state of lithium-ion battery is critical for safe operation and performance optimization. Conventional methods rely on temperature sensors (thermocouples), however equipping large number of cells with a sensor poses challenges including surface-only data, intricate wiring and added weight. In the past few years, there is a great interest shown towards temperature estimation using EIS (Electrochemical impedance spectroscopy), as it can be considered an easy and quick method without relying on complex thermal models [1], [2]. In this study, we propose a sensor-less technique utilizing impedance measurements to infer battery temperature.

To validate the methodology, high impedance/high energy (LG-M50-5Ah) and low impedance/high power (Pouch-40Ah) cells were tested over a range of different temperatures and SOCs. The different ranges of impedance among both cell types were used to assess the feasibility and limitations of the proposed approach. For the first time, Distribution of relaxation times (DRT), a model-independent method was explored to estimate battery temperature. DRT method has an advantage on conventional Nyquist plot/ ECM interpretation by separating the time constants, which could assist in extraction of temperature dependent features. The features from the EIS data were then captured using traditional linear Arrhenius relation between impedance and temperature.

We evaluated the feasibility of wide-range temperature estimation, and identified challenges in the high temperature spectrum. Hence, the study focuses on a specified range of temperatures between 25°C and 45°C. Upon validation using a single order polynomial, high energy LG cells demonstrated strong agreement with high method accuracy as compared to the high power pouch cells across all SOCs. Overall across all extracted features, temperature prediction for the LG cells lies in the range of~0.21°C to ~2.3°C, while a higher error of ~1.17°C to ~6.8°C was observed for pouch cells due to measurement accuracy limitations. The study highlights both potential and challenges when applying the proposed method on different commercial application lithium-ion cells.

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Acknowledgments:

This study is supported by the European Union's Horizon Europe project 'ENERGETIC' (Grant No 101103667).

Biodegradable-polymer-pectin based porous membrane and carbon electrodes for Na-ion hybrid capacitors and supercapacitor

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The abundance of sodium resources, low cost and sustainable options leads Na-based energy storage devices as a strong alternative to commercial Li-based devices to resolve the issues related to limited lithium reserves. Hybrid capacitors which are novel energy storage devices with the features of both batteries and supercapacitors have recently gained significant attention due to their high energy density and power density. This unique class of energy storage device combines the bridge between supercapacitors and batteries by utilizing a capacitortype cathode and battery-type anode in a single device.

In the search for long-term energy storage options, biodegradable materials for electrochemical devices have received a lot of attention in recent years. Among different types of available bio-polymers pectin can be most versatile precursor for "green" energy storage devices. In this work, pectin has been used to prepare all components for Na-ion capacitors (NICs) and supercapacitors (SCs) such as hard carbon (HC), activated carbon (AC) and porous membranes to design high performance devices.

The pectin, derived from citrus peels, has been used to prepare activated carbon as an cathode via physical and chemical activation process by utilizing KOH as activating agent. To prepare an anode for NICs, the raw pectin has been hydrothermally pretreated then thermally pyrolyzed at 1500°C. The prepared anodes and cathodes are characterized by using SEM, Raman, XRD, BET and XPS etc. to analyze their morphology and structure to confirm their suitability as carbon electrodes.

Moreover, since most of the NICs and SCs are based on organic liquid or liquid electrolytes which have drawbacks such as flammability, volatility, leakage etc. To overcome these challenges pectin based gel polymer electrolytes (GPE) have also been prepared here. To prepare the gel, first porous membrane has been prepared via non solvent evaporation method after that the vacuum dried porous membrane has been soaked in to the liquid electrolyte for 1-2 hours for complete soaking and gel formation. To increase the ionic conductivity and mechanical stability of the GPE different amounts of Polyethylene glycol (PEG) have been added. The measured conductivity of the optimized GPE has been found in the range of 3.2 - 4.5 mS cm-1 which is quite good enough to utilize it as efficient electrolyte to fabricate NICs and SCs. Further, the prepared NICs and SCs were electrochemically characterized by charge-discharge, cyclic voltammetry and impedance spectroscopy measurements.



Aqueous Processing ABC: Improving the Aqueous Processing of Ni-rich Layered Oxide Cathodes by <u>A</u>dditives, <u>B</u>inder and <u>C</u>oating.

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To ensure an extensive market penetration of electric vehicles, among others, lithium ion batteries (LIBs) need to achieve a further reduction of the environmental impact and costs of the electrode manufacturing process.^[1] Aqueous processing of LIB cathode electrodes has become a vital strategy to substitute state-of-the-art teratogenic *N*-Methyl-2-pyrrolidone (NMP) solvent and the fluorinated polymer polyvinylidene difluoride (PVDF) binder with more sustainable materials.^[2] Additionally, benefits like savings in investments and processes, *e.g., via* circumventing solvent recovery and a faster recycling can be expected.^[3]

Aqueous processing of Ni-rich layered oxide (NCM) cathodes is investigated and demonstrated^[4], though obvious issues require further investigations, *i.e.*, Li-leaching, aluminium dissolution/corrosion^[2] as well as moisture-sensitive CAM degradation.^[5] Individual and issue-targeted solutions are established in literature, while their relevance for individual processing step including meaningfulness of strategy combinations are less investigated and discussed. Therefore, the protection and stabilization of the NCM particle is considered in a bottom-up approach along the supply chain with particle modification, electrode processing as well as cell charging and discharging. In general, tailored particle coatings might not be suitable to protect the initial NCM surface while other approaches including additives and binder are reasonable to protect fresh formed surfaces during cathode processing and cycling.

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Asymmetric Benzene Sulfonamide Sodium Salt Enabling Stable Cycling in Solid-State Sodium Metal Batteries

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The growing demand for efficient and safe energy storage systems, driven by the increasing penetration of renewable energies and the expansion of electric vehicles, has accelerated the search for alternatives to conventional lithium-ion batteries. Sodium-based batteries have emerged as a promising solution due to the abundance of sodium and the potential for lower costs[1,2]. In particular, all-solid-state sodium metal batteries (ASSSMBs) have attracted considerable attention for their enhanced safety and higher energy density [3]. However, challenges such as dendrite growth, low sodium-ion transport numbers, and poor interfacial stability with sodium metal anodes (Na°) limit their practical application.

In this context, we report the synthesis and characterization of a new sodium salt, sodium (benzenesulfonyl)(trifluoromethanesulfonyl)imide (NaBTFSI), designed to address these limitations. The π - π stacking interactions introduced by the benzenesulfonyl group were expected to improve the mechanical integrity of the polymer electrolyte and to influence the sodium-ion transport properties positively.

The main objective of this work is to investigate the effect of NaBTFSI on the ionic conductivity, sodium transference number (T_{Na}), and, especially, the solid electrolyte/Na° anode interface stability. For this purpose, solid polymer electrolytes (SPEs) were prepared and, compared to the widely used NaTFSI salt. NaBTFSI-based electrolytes exhibited higher ionic conductivity and improved sodium-ion transport.

A key focus was placed on the interface behavior between the polymer electrolyte and the Na° metal anode. Through electrochemical impedance spectroscopy (EIS) and sodium plating/stripping experiments, it was demonstrated that NaBTFSI-based electrolytes significantly improve the interfacial stability. Notably, NaBTFSIbased electrolytes enabled continuous and stable cycling for more than 350 hours without short-circuit formation, whereas conventional NaTFSI-based electrolytes failed after less than 50 hours. This outstanding performance is attributed to the formation of a more robust and stable solid electrolyte interphase (SEI) promoted by NaBTFSI, which effectively suppresses dendrite growth and ensures prolonged cycling.

Overall, this work highlights the promising role of NaBTFSI as a high-performance electrolyte salt for ASSSMBs, offering improved conductivity, higher T_{Na} , and exceptional interfacial stability — essential features for the development of next-generation sodium batteries.

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Enhancing Interfacial Stability and Performance in PVDF-HFPbased Polymer Electrolytes: The Role of Crosslinking

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Solid-state secondary batteries are emerging as the next-generation electrochemical energy storage systems, offering high performance for electric vehicles and smart grid applications. To facilitate a sustainable energy transition, the development of safe and reliable solid polymer electrolytes (SPEs) is crucial. These electrolytes not only enhance energy density and safety but also enable the use of lithium metal anodes, thereby advancing more efficient and durable battery technologies. However, the practical implementation of SPEs is hindered by intrinsic limitations, such as low ionic conductivity at room temperature and inadequate mechanical strength to prevent dendrite formation [1,2].

The SOLVE European project aims to address key challenges associated with SPEs for next-generation solid-state secondary batteries. It seeks to develop mechanically robust macromolecular networks encompassing specific additives for high ionic conductivity and electrochemical stability. In particular, lithium salt and ionic liquid (IL), along with organic or inorganic crosslinkers, are incorporated into the PVDF-HFP (polyvinylidene fluoride-co-hexafluoropropylene) matrix and solid-like polymer electrolytes are obtained through a solvent-free extrusion process for component integration, followed by a fast, scalable UV-crosslinking process. This approach allows at achieving enhanced ionic conductivity (≥ 1 mS cm⁻¹), high oxidative stability (≥ 4.4 V vs. Li⁺/Li), and excellent compatibility with cathode composites. It shows great potential for enhancing interfacial stability by forming a Semi-Interpenetrating Polymeric Network (SIPN), thereby improving performance of the electrolyte, and enabling high C-rate performance and contributing to the development of safe, energy efficient and durable solid-state Li-metal battery technologies [3,4].

In summary, here we demonstrate how integrating SIPN with ionic liquids (IL) and alkali metal salts offers significant advantages, including high ionic mobility, improved cycling stability, and safety due to the inherent benefits of IL, such as low flammability, low volatility, and high chemical stability. At present, we are actively working on optimizing this system to further enhance the long-term stability and the rate performance at low temperatures of PVDF-HFP/IL electrolytes.

Acknowledgements

The SOLVE project (<u>https://www.solveproject.eu/</u>) has received funding from the European Union's Horizon 2023 Research and innovation Program under grant agreement No 101147094.

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RECOVERING VALUABLE MATERIALS FROM SPENT LITHIUM-ION BATTERIES USING DIRECT RECYCLING TECHNOLOGY

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With the widespread application of lithium-ion batteries across various markets, especially in electric vehicles, the challenge of recovering valuable materials from spent battery cells becomes increasingly critical.





AETC "mined" the cathode active material from spent lithium-ion batteries via a technology of Direct Recycling (Figure 1(a)), which includes the process of discharge, conveyorized cell disassembly, and mechanical processing to obtain the desired materials. Pilot plant setup at AETC currently allows to process several hundred cells per work shift, which generates large, representative lots of cathode active material for recycling; these went to ISPE for downstream processing to extract metals and either recycle them back into the cathode active or repurpose into other markets. The reported processing of NCA and NMC811 at ISPE made it possible to obtain engineered nickel-cobalt alloys with valuable ferromagnetic properties, pure electrolytic manganese dioxide (EMD, x-MnO₂) and lithium in the form of LiOH or Li₂CO₃ (Figure 1b), while processing at AETC generated high-purity spherical graphite, copper, aluminum, stainless steel. We established at high TRL of 6-8 prerequisites for returning these materials to the technology cycle of manufacturing of new batteries from raw materials streams generated through Direct Recycling, as well as obtaining many materials that have independent commercial value.

ISPE acknowledges support through a partnership project funded by AETC with Science and Technology Center in Ukraine, funding agreement P796, and "Horizon Europe" Program under Agreement 101137771 "STREAMS", and Agreement 101147342 "SAFELOOP".



SYNTHESIS AND INTEGRATION OF NANO-STRUCTURED SILICON INTO ANODE ACTIVE MATERIALS OF LITHIUM-ION BATTERIES

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Recent experiments conducted by researchers at CEA resulted in production in a double stage Laser pyrolysis set up [1] of short aggregate chain, 40 nm, and 18 wt.% carbon coated silicon material that was successfully co-processed by AETC into a battery-ready anode active material that displayed enhanced stability at prolonged electrochemical cycling in battery cells constructed by ISPE. Reactive synthesis at CEA focuses on optimization of chemical composition of precursor gas, variables of operation of lasers, optimization of flow rates, reactor pressure, and dilution of precursor, all of which had an effect on the resultant particle size aggregate chain length and other surface properties such as BET surface area. Work at AETC focused on using industrial scale mechanochemical [2] and continuous thermal processes to generate hybridized particles that contain natural graphite, mesophase carbon-derived synthetic graphite, recycled and healed graphite extracted from spent lithium-ion batteries, nano-structured silicon to form a composite with precise composition of individual aforementioned ingredients on a single particle level. To further stabilize anode active material from volumetric change, hybridized anode particles were surface coated with a layer of nanostructured soft carbon. Battery electrochemists at ISPE were able to successfully apply the resultant particles on to copper foil and construct half-cells of CR2032 configuration, achieving very stable cycling versus control hybridized graphite that consisted of all the same ingredients without the addition of silicon nanoparticles. The team focused on the application of 1 wt.% loading of silicon into the hybrid anode and higher loadings are being explored concurrently.





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Extending Cycle Life in Sodium-Ion Pouch Cells: Suppressing Sodium Plating with a Novel Electrolyte Additive

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The electrolyte plays a crucial role in improving key performance indicators (KPIs) such as energy efficiency, cost, safety and lifespan of a sodium-ion battery (SIB). Layered oxides represent a particularly promising cathode active material although they present certain challenges and limitations at high cut-off voltages, which are necessary to achieve a high energy throughput. Adjusting the electrolyte to mitigate active material degradation^[1], gas evolution^[2] and sodium plating on the anode^[3], key issues in SIBs for cycling at high cut-off voltages, can significantly enhance performance. Patent and literature analyses were conducted to identify relevant existing electrolyte components, providing a foundation for further electrolyte development and discovering niches for research on novel additives. Electrolyte additives are a cost-effective way to optimize the performance of a SIB cell, as they typically only make up a small amount of a few weight percent. Therefore, electrolyte additives were investigated to improve the safety and lifespan of SIBs.

The patent and literature analysis identified NaPF₆, NaFSI, and NaTFSI as the most relevant conductive salts, mainly carbonates as solvents and additives analogous to Lithium-ion technologies. The cathode active material used for the electrolyte development was the NFM334-type layered oxide, Na0,96Ca0,04[Ni0,24Fe0,30Mn0,37Zn0,09]O2, which occurs in the O3 phase. The electrolyte development, incorporating components identified through patent and literature analysis, was developed through systematic experimental salt and solvent combination approaches. The best-performing formulation was selected for the subsequent iteration, leading to 1.2 M NaFSI in a DEC:EC (30:70, by wt.) mixture as baseline formulation. During cycling (1 C = 220 mA, cut-off voltages 1.5 -4.0 V, 40 °C) with an iterative developed baseline formulation sodium plating and gas formation were observed, highlighting the need for SIB-specific electrolyte additives to mitigate these challenges. In addition, degradation of the cathode active material was detected, driven by a phase transition occurring during charge and discharge at the elevated cut-off voltage of 4.0 V. This leads to the release of transition metal ions into the electrolyte, which then migrate toward the anode, facilitating the deposition of sodium metal on the anode material and resulting in sodium plating. The transition metals and the sodium plating were detected on the anode via ICP-OES and SEM/EDX. An optimized electrolyte formulation with 1 wt.% of a novel additive led to a 30 % increase in lifespan, strongly suppressed sodium plating and reduced gas formation compared to the baseline electrolyte. Additional benchmarking in a KPI analysis against well-established electrolyte additives such as VC (vinylene carbonate), FEC (fluoroethylene carbonate) and NaDFOB (sodium difluoro oxalate borate) clearly demonstrated that the optimized formulation with the novel electrolyte additive offers enhanced overall battery performance.



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Solid-state vs. Spray-drying synthesis for Mg-doped P2-Na_{0.67}Fe_{0.5}Mn_{0.5}O₂ as cathode material for Sodium-Ion Batteries

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Among the different cathodes studied for Sodium-Ion Batteries, the P2-layered oxide structure has garnered significant attention for its electrochemical properties. To increase the electrochemical performance of the P2- $Na_{0.67}Fe_{0.5}Mn_{0.5}O_2$ oxide^[1], we performed several substitutions by introducing Mg²⁺, an electrochemically inactive cation already known in the literature for its stabilizing properties during cycling. A series of P2-Na0.67 (Mn0.5Fe0.5xMgx)O2 (x=0, 0.1, 0.2, 0.3) was synthesized by solid-state synthesis. The x=0 and 0.2 compositions were also synthesized by spray-drying synthesis followed by heat treatment to evaluate how different synthesis routes can influence the structural and electrochemical properties. The morphology and elemental composition of all materials were analyzed using SEM-EDX, ICP-AES, and XRD. Additionally, they were characterized as cathodes in coin cells between 1.5V and 4.2V. Operando XRD was also used to observe the structural evolution of the P2layered structure during cycling, revealing that Mg²⁺ inhibits P2-Zphase transition at high potential (>4V). The solid-state syntheses performed resulted in the successful synthesis of the desired P2-layered structure with a small electrochemically inactive phase impurity. Structural analysis showed that the c parameter of the elementary cell decreases as the amount of Mg increases, in contrast to what was expected. In comparison, spray-drying synthesis resulted in the formation of purer P2 oxides than those obtained by the solid-state, although with a small percentage of O3 phase. Moreover, with spray-drying synthesis, it was possible to considerably reduce the calcination temperature (800°C vs. 1000°C) and increase the sustainability of the synthesis, as the first step was conducted in aqueous solution. GITT experiments show a significant Na⁺ diffusion coefficient (D_{Na}⁺) increases as the Mg²⁺ content increases and for spray-drying materials compared to solid-state ones. The best-performing cathode was found to be Na_{0.67}Mn_{0.5}Fe_{0.3}Mg_{0.2}O₂. Compared with the x=0 sample, the specific capacities achieved are significantly higher at each of the C-rates tested (~110 vs. 70 mAh g⁻¹ @0.5C, respectively). This material also proved to be significantly better performing than x=0 oxide after many cycles, as the capacity loss observed after 100 cycles is practically halved (29% vs. 55% @1C). Comparing the registered performance for the materials obtained through the two different syntheses carried out, it was found that those synthesized by spray-drying always showed higher electrochemical performances. The best cathode was also tested in full-cell configuration with commercial Hard-Carbon (HC) as anode. In order to best optimize the cell performances, we evaluate the influence of different N/P ratios on the registered specific capacity and capacity retention after 100 cycles. We decided to simulate the effective operation of a battery by slowly charging it in CCCV (Constant Current-Constant Voltage) mode and fast discharging in CC condition.

In order to compensate for the low Na content in P2 oxides, we tried several strategies to increase the Na content and consequently limiting the irreversible loss of Na due to SEI formation during the first charge. Firstly, we electrochemically pre-sodiated the Hard Carbon by assembling half-cell vs. Na(m) at different charging states (200 and 300 mAh g-1) and then we used it in full-cell configuration. For the same purpose, we also studied the effect of Na2CO3 as sacrificial salts^[2].

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A SUSTAINABLE DELAMINATION APPROACH FOR SIMULTANEOUS SEPARATION AND LEACHING OF CATHODES FROM END-OF-LIFE LI-ION BATTERIES

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The increasing demand for Lithium-ion batteries (LIBs) in several applications has led to a substantial rise in their production, posing risks in the supply of critical raw materials (CRM, e.g.: Li, Ni, Co). Additionally, improper disposal of end-of-life batteries can lead to environmental pollution and loss of technological value stressing the necessity for sustainable recycling. Current methods involve shredding batteries into a black mass, further processed via pyrometallurgy (energy-intensive) and/or hydrometallurgy with inorganic acids (environmentally hazardous) to recover CRMs. A more refined approach to LIBs recycling includes the dismantling and the sorting of their components, allowing for a targeted extraction.

The spent cathodes recycling process proposed involves the simultaneous delamination from the current collector and the leaching (>95%) of the cathode active material (CAM) in a citric acid solution, enabling also the recovery of Polyvinylidene fluoride (PVDF) and Carbon filler as unleached residues which are further separated by means of filtration through a silica-packed column. Transition Metals (TMs) are recovered as precursors (p-CAM) with high yields (>88%) and used to resynthesize fresh CAM, thus closing the recycling loop. To validate the proposed strategy, the recycled CAM was used in a new cathode manufacturing followed by its functional characterization in a half-cell configuration.



Figure 1. Scheme of the proposed process involving simultaneous delamination and leaching of the cathode, comprising the recovery and purification of the PVDF binder, and followed by CAM precursor recovery and CAM resynthesis

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Direct Recycling of Cathode Materials from Production Scraps

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As Lithium-ion batteries (LIBs) are increasingly used in the global economy, establishing efficient recycling processes will continue to become increasingly important.¹ With the current efforts in place to increase LIB cells production via the establishment of gigafactories, electrode production scrap is expected to become a relevant source of valuable materials. The cathode material, accounting for about 50% of the LIBs' cost, ending up in electrode scraps could be instead embedded in new LIB cells with efficient direct recycling processes.² Direct recycling of the black mass generated by production scraps has become an important element to the research within the RESPECT project, aiming to transform the recycling process for batteries by closing the loop of battery manufacturing with direct recycling with common cathode chemistries.³ Direct recycling of cathode materials is a promising alternative to classic hydrometallurgical or pyrometallurgical methods, representing a more environmentally friendly and efficient recovery method for high value products within the battery value chain.⁴

In this study, we focus on two different cathode chemistries, *i.e.* Lithium iron phosphate (LiFePO₄, or LFP) and Lithium nickel manganese cobalt oxides (NMC). We report on the study and investigation of black mass purification methods such as treatments with different solvents followed by filtration and calcination steps for an efficient removal of additional components such as Polyvinylidene Fluoride (PVDF) binder and carbon additives. Greener and sustainable solvents are investigated against N-methyl pyrrolidone (NMP). Moreover, final re-lithiation strategies are also proposed aiming at a complete recovery of the properties of the pristine cathode chemistries. Moreover, final re-lithiation strategies are also proposed aiming at a complete recovery of the properties of the pristine cathode chemistries. A comprehensive morphological, structural and physical characterization of the black masses and the treated sample has carried out to assess the efficiency of scalable regeneration procedures proposed, paving the way towards sustainable processes for direct recycling of cathode active materials. The workflow is shown in Figure 1 below.



Figure 1. Depiction of workflow processing black mass from scraps by characterizing, pre-treating (PVDF/C removal), testing and validating LFP/NMC samples.

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ORGANIC-INORGANIC HYBRID SOLID COMPOSITE ELECTROLYTES COMBINE FUNCTIONALITY, MANUFACTURABILITY, AND ELECTRODE COMPATIBILITY

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Solid or quasi-solid electrolytes can allow to maximize the active material content in lithium battery cells, enhancing energy density. They may also enable the efficient use of lithium metal while mitigating safety concerns due to the absence of flammable carbonate solvents. However, most electrolytes degrade upon contact with lithium metal or high-voltage cathodes like LiNi_{0.8}Mn_{0.1}Co_{0.1}O₂ (NMC811). Ideally, this results in passivating interfaces that prevent further breakdown while allowing lithium-ion transport, making the formation of a robust solid-electrolyte interphase (SEI) crucial for lithium metal electrodes. This can be addressed by making use of ionic liquid electrolytes (ILEs), where the decomposition of LiFSI on lithium metal leads to the formation a LiFrich SEI, improving lithium plating/stripping reversibility. In general, ILEs offer a promising alternative to conventional liquid electrolytes, with high ionic conductivity, broad electrochemical stability, thermal stability, and nonflammability. ILEs can be incorporated into a solid matrix to form solid composite electrolytes (SCEs), or ionogels, where the confined ILE acquires solid-like properties due to interactions with the host matrix, combining the advantages of both liquid and solid electrolytes. For commercial viability, SCEs must be manufacturable, which can be achieved through liquid processing. Here, a precursor solution is impregnated into a porous electrode and rapidly solidified, ensuring intimate electrode/electrolyte contact. Polymer-based SCEs offer good manufacturability but suffer from low ionic conductivity, whereas silica-based SCEs can achieve higher conductivity. Traditional non-hydrolytic silica synthesis requires acids that damage electrodes, while hydrolytic synthesis avoids acids but suffers from spontaneous and slow solidification, limiting manufacturability.

This work presents a hybrid SCE that combines functionality, electrode compatibility, and manufacturability. Stable liquid precursor solutions were developed by combining two alkoxysilanes with an ILE, water, and a solvent. One alkoxysilane features vinylidene groups that undergo rapid photo-initiated radical polymerization, forming an organic-inorganic hybrid solid matrix encapsulating the ILE. These acid-free precursor solutions were impregnated into porous electrodes and solidified instantaneously using UV irradiation, which induces molecular ordering at the ILE/silica interface, enhancing lithium-ion mobility and increasing ionic conductivity (up to 2.7 mS cm⁻¹ at room temperature, surpassing the neat ILE). Using an ILE containing bis(trifluoromethane)sulfonimide as anion, the SCE demonstrates an anodic stability >5 V vs. Li⁺/Li and compatibility with NMC811 electrodes. However, its poor stability against lithium metal limits the rate performance of NMC811/Li cells. By incorporating a mixed ILE containing two different types of anion, the SCE exhibits slightly lower anodic stability but enables the formation of robust, conductive degradation layers at the SCE/Li interface. When employing the first SCE as a catholyte and the second as an anolyte, NMC811/Li cells demonstrate high electrochemical stability, retaining 91–100% of their initial capacity after 100 cycles (0.2 C).

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INVESTIGATION AND ANALYSIS OF EUROPEAN LITHIUM-ION BATTERY MACHINERY PRODUCTIVITY

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Climate change and global warming are the two major environmental threats facing the world and the widespread use of fossil fuels as the primary energy source intensifies their impacts. In order to minimize the use of fossil fuels and eliminate the existing environmental problems caused by them, scientists and policymakers seek sustainable and renewable energy sources such as energy storage systems, solar and wind [1]. Among energy storage systems, lithium-ion batteries (LIBs) dominate the market by their outstanding advantages such as high energy density, high reliability, relatively stable charge-discharge conditions, and long cycle performance. In addition, with their superior characteristics, LIBs find widespread applications, primarily in electrical vehicles, as well as in portable electronic devices and smart power grids. On the other hand, its functionality and high-performance features not only attract increasing attention but also strengthen competition in this field. In terms of LIB production, China is the leader with 74%, followed by Europe with 8.5% and United States 11% [2]. In response, the European Union (EU) is making significant efforts to strengthen the LIB value chain in order to gain a competitive edge in this market. Although raw material supply is currently receiving the highest attention when considering the overall LIB production value chain; battery machinery is one of the most important elements influencing environmentally friendly, cost-effective and energy efficient battery production.

In this study, we analysed on the EU-wide battery machinery production. For this purpose, a map including the 174 machinery manufacturers by country have taken as reference. In the next step, publicly available product catalogues of each manufacturer were solicited by different methods such as sending e-mails, following their talks and posts on online dissemination tools and visiting their booths in the fairs. The accessed product catalogues were classified based on the LIB manufacturing steps. After categorizing the collected data, we conducted various comparisons with country-specific data across different scopes such as distribution of LIB machinery manufacturers by LIB production process stages on country basis and comparison of number of LIB machinery manufacturers and R&D expenditure relative to gross domestic product values by country. Obtained results were used to develop a critical perspective of LIB value chain of Europe from the LIB machinery point of view.



Figure 1. Distribution of lithium-ion battery machinery manufacturers across Europe

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Characterization of battery materials through high-resolution laboratory X-ray nano-CT

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X-ray nano-tomography is a powerful technique to obtain 3D structural information on a sub-micron scale. By utilizing a magnifying setup with a nano-focus X-ray tube, resolution down to 150 nm can be achieved in the laboratory without requiring optics [1]. These setups are comparatively simple and provide excellent resolution over a large energy range, enabling applications from low-density materials (e.g. tissues and plastics) [2] to denser materials (e.g. electronics and battery materials) [3]. Here we demonstrate the capabilities of laboratory nano-CT using a phone battery cathode (LiCoO2 coated on Aluminium) and anode (graphite coated on Copper).

The used system is a custom-built nano-CT, described in detail in [2], utilizing the Excillum NanoTube N3 160 kV and a DECTRIS EIGER2 1M CdTe detector. A CT scan with a voxel size of 200 nm is performed. Analysis of the grain structure is performed in Dragonfly (Comet Technologies Canada) by segmenting all grains individually enabling statistical analysis of the structure [4]. Using the reconstructed 3D volume, inspection of the material, e.g. for cracks and damaged particles, and the interface between the grain is possible. Using the segmented grains, statistical information can be extracted on their size distributions, shape, aspect ratio, etc.

Utilizing high-resolution 3D data enables the evaluation and characterization of the structure of anode and cathode materials on the sub-micron scale, facilitating a deeper understanding of the material structure. No further sample preparation is required, except for cutting out a region of interest, which is left intact for further analysis, such as electron microscopy. Additionally, high-resolution X-ray CT aids in the identification of material properties and potential issues, enhancing development and optimization of materials and components, contributing to innovations in design and performance improvements.



Figure 1. Reconstructed slices from a battery cathode and anode (left) and segmented and characterized grains by maximum diameter in cathode active material (right)

ACKNOWLEDGEMENTS

Setup development: FA4.0; Samples: Batteries Sweden (BASE); future development: FullMap

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ADVANCING STANDARDISATION OF SOLID-STATE BATTERIES' INDUSTRIALISATION FOR THE ELECTRIC VEHICLE SECTOR

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Standardisation plays a crucial role in advancing the electric vehicle (EV) market, particularly in battery development and production. It ensures safety, efficiency, and sustainability by providing consistent guidelines for manufacturing, testing, and recycling processes. Standardisation also supports the shift from mass production to customisation, aligning with evolving customer demands and competitive market dynamics.

SPINMATE^[1] project plays a pivotal role in advancing the standardisation of solid-state battery (SSB) technology, ensuring that these batteries meet the stringent requirements of the EV sector. By aligning with EU and international regulations, the project supports sustainability across the entire battery lifecycle, from eco-design and production to recycling and second-life applications. This initiative contributes to the European Green Deal's objectives for a circular economy and climate neutrality. At the core of SPINMATE is its emphasis on standardisation, which ensures that SSB cells meet the rigorous demands of the EV industry. By adopting innovative methodologies, including the use of digital twins and machine learning, the project optimises manufacturing processes, enhancing performance, safety, and sustainability. Collaboration with initiatives like SOLID4B^[2] and active participation in standardisation committees allows SPINMATE to influence the development of future standards, further connecting its contribution to the EV sector. INOVA, as the leader of standardisation activities within the project, has set clear priorities for standardisation efforts. These include monitoring existing standards, fostering collaboration with other projects, and supporting the development of new standards for advanced manufacturing systems. The project follows a structured approach to define and implement standards that align with international best practices, ensuring quality, sustainability, and competitiveness. One of SPINMATE's key achievements is the development of self-defined standards for crucial processes like cell assembly and eco-design. These standards address critical issues such as adhesive strength and environmental impact, ensuring that SPINMATE's technologies are aligned with evolving regulations. The creation and implementation of these standards help enhance the competitiveness of the EV battery industry, addressing safety concerns, reducing carbon footprints, and improving recycling efforts in line with the EU's new battery regulations. SPINMATE also integrates sustainable and digitalised production processes. By focusing on eco-design advancements that facilitate disassembly, the project promotes environmental and safety considerations throughout the lifecycle of battery packs. Key manufacturing processes, such as cathode and anode preparation, solid electrolyte membrane production, and cell assembly, are standardised to meet stringent quality and safety standards. This approach not only optimises manufacturing but also strengthens the project's positioning within the competitive EV market. Furthermore, SPINMATE prioritises standardisation in the design and end-of-life handling of battery packs. By standardising mechanical joints and data connection interfaces, the project ensures that battery packs can be easily disassembled, maximising reuse and recycling^[3], as well as enabling industry-wide testing^[4] and evaluation. This aligns with SPINMATE's sustainability goals and supports the transition to a circular economy within the EV industry. The SPINMATE project's focus on standardisation, eco-design, and sustainable production is crucial for the future of the EV battery market.

By developing self-defined standards and incorporating advanced digital and sustainable manufacturing techniques, the project strengthens its competitiveness and helps create a sustainable, regulated battery market. Through its comprehensive approach to standardisation, SPINMATE not only enhances battery performance but also sets a benchmark for the industry, supporting the EU's climate neutrality and circular economy goals.

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Bio-based electrolytes for lithium batteries: Electrochemical performance, Life Cycle Assessment, and Techno-Economic feasibility

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Bacterial cellulose (BC) is a bio-based material produced by bacteria, noted for its unique structure composed of cellulose nanofibers [1]. This study incorporated the material into the polyethylene oxide (PEO) polymer electrolyte to improve its film-forming properties in lithium-ion batteries. It also served as a separator to mitigate the risk of short circuits caused by lithium dendrites. The ionic conductivity of this polymer electrolyte, when plasticized by ionic liquid, can attain about $10^{-3}S \cdot cm^{-1}$ at 60 °C. An operando optical analysis demonstrated the potential of the BC separator for application in lithium metal batteries to mitigate dendrite formation. The potential window extended to 4.1 V relative to Li/Li^+ . A single-layer pouch cell battery was assembled utilizing the prepared PEO/BC-based polymer electrolyte, lithium iron phosphate (LFP) cathode, and lithium metal anode, followed by electrochemical performance evaluations.

To assess the broader implications of this material, a life cycle assessment (LCA) was conducted to evaluate the environmental impact of integrating BC into polymer electrolytes. Key sustainability indicators, such as global warming potential, human toxicity, land use, and abiotic resource depletion, were analyzed. A detailed LCA of the TwinVECTOR battery involved extensive data collection on both battery components and the laboratory-scale production of bacterial cellulose used in the electrolyte/separator. The results indicate that the primary environmental impact arises from energy consumption associated with equipment operation and technical building services (TBS). When TBS is excluded, the electrolyte/separator contributes the most to the overall impact, largely due to the resource-intensive nature of bacterial cellulose production, which significantly increases the environmental burden [2]. Given that all components were manufactured at a laboratory scale, the associated impacts are considerably high [3].

Additionally, techno-economic simulations were performed to explore the feasibility of scaling up BCbased polymer electrolytes for commercial applications, considering production costs and process efficiency. The assessments include key factors like materials costs, manufacturing expenses and economic feasibility. Optimizing electrolyte thickness with enhanced cell's specific capacity can potentially reduce environmental impacts. Additionally, integrating a renewable energy mix for energy consumption may further improve the overall sustainability of the battery. This work highlights the technical performance, environmental impacts and economic potential of the bio-based polymer electrolytes. The combined approach of experimental validation, environmental assessment, and economic feasibility highlights the potential of BC-enhanced polymer electrolytes as a sustainable and high-performance alternative for next-generation lithium metal batteries.

ACKNOWLEDGEMENT: This project has received funding from the European Union's Horizon Europe research and the European Research Executive Agency under Grant Agreement No. 101078935.

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Optimization of Fast-Charge Current Protocols for Electrochemical Prelithiation of Pure Si Lithium Ion Batteries

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Silicon (Si) as anode material offers high theoretical capacity (~3579 mAh/g)[1] but suffers from significant volume expansion, leading to rapid capacity fading[2]. Prelithiation is an effective strategy to enhance initial coulombic efficiency (ICE) and reduce irreversible capacity loss. This study explores a high-frequency pulsed current (1 C-2 s/1 s, where 2 s represents the charge-period and 1 s the off-period) approach for electrochemical prelithiation and compares it with conventional constant current (CC) methods[3].

Results showed that 1C-2 s/1 s and 0.1 C-CC achieved similar ICE (84.6%) and cycling stability, retaining 80% capacity after 108 and 120 cycles, respectively. However, 1 C-2 s/1 s significantly reduced prelithiation time by nearly sevenfold compared to 0.1C-CC. Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) confirmed a higher prelithiation degree under 1 C-2 s/1 s than 1C-CC. Further half-cell cycling tests revealed that the reversibility of Li introduced during 1 C-2 s/1 s prelithiation reached 72%, nearly double that of other conditions. Scanning Electron Microscopy (SEM) analysis showed that Si anodes prelithiated under 1 C-2 s/1 s and 0.1 C-CC exhibited significantly larger cracks compared to 1 C-CC, and the crack sizes under these two conditions were similar, which is related to the higher lithium content.

These findings confirm the feasibility of high-frequency pulsed current prelithiation at high C-rates. The use of pulsed current facilitates fast and efficient prelithiation, reducing time from 60 minutes (0.1 C-CC) to 9 minutes (1 C-2 s/1 s), paving the way for optimized strategies to improve Si anode performance.



Figure 1. Cycling performance of pure Si anodes under different current conditions: 0.1 C (left) and 1 C (right), with CC, 2 s/1 s, and 2 min/1 min.

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LITHIUM-ION CELLS SAFETY ASSESSMENT

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Lithium-ion batteries (LIBs) are widely employed across various applications due to their energy density and long cycle life. However, ensuring their safety is of paramount importance, given the potential risk of fires or explosions stemming from overheating, overcharging, or internal short circuits. Thermal runaway (TR) represents the most significant safety hazard in lithium-ion batteries causing battery failure. It can be triggered by mechanical, thermal, or electrical abuse conditions, each of which induces exothermic and uncontrolled chain reactions within the cell components, ultimately compromising the battery's integrity and safety [1]. This study investigates the hazards associated with thermal runaway using the standard Accelerating Rate Calorimeter (ARC), an instrument able to simulate adiabatic conditions to perform thermal abuse tests focused on assessing the thermal behaviour and critical safety limits of the tested batteries. Commercial cylindrical cells with NMC cathode were tested, starting with electrochemical characterization following a specific protocol that employed the galvanostatic intermittent titration technique (GITT) to assess the state of health (SoH) of the aged cells. This characterization focused on analysing capacity fade and the increase in internal resistance resulting from the degradation mechanism associated with aging [2]. Starting from these results, thermal abuse tests within the standard ARC were performed with the aim of identifying key thresholds for thermal instability, including the self-heating onset temperature and the amount of heat released during the process. The heat-wait-seek (HWS) operation mode was employed within the standard ARC protocol to accurately monitor the thermal behaviour of the batteries during thermal abuse testing (figure 1). This approach enabled the identification of specific critical temperatures associated with TR, related to chain reactions induced by degradation mechanisms such as the decomposition of the SEI (Solid Electrolyte Interphase) layer at the anode, which triggers the self-heating of the battery.



Figure 1. Thermal abuse tests using the standard ARC

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BIOSTORE - Biologization of Batteries and Materials

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The BIOSTORE project aims to advance battery technology by incorporating biological and bio-based materials to replace fossil-based components, thereby reducing environmental impact. Aligned with the principles of the bioeconomy and circular economy, this interdisciplinary initiative integrates biology, chemistry, battery research, political and social sciences, and economics. A key focus is the research and modification of bacterial exopolysaccharides, natural rubber, and other biopolymers and bioinspired additives. These materials are optimized through genetic and chemical modifications to enhance battery performance. Additionally, life cycle assessment will be employed to evaluate the sustainability of the materials and processes developed. To ensure broader acceptance and responsible innovation, the project also examines the political and social dimensions of new battery technologies.

Ongoing research focuses on the development of anode active materials, with particular emphasis on hard carbon, while investigations on the cathode side are directed toward binder materials and their rheological properties. To eliminate per- and polyfluoroalkyl substances (PFAS) from battery components, alternative cathode binders are being explored. Both binders and separators are derived from polysaccharides, though they exhibit distinct functionalities and material characteristics. Additionally, the project investigates the potential of biological and plant-based additives in electrolytes as a sustainable alternative to conventional components. By integrating these innovative materials and processing techniques, the project aims to advance environmentally sustainable and high-performance battery technologies.



Figure 1. A schematic overview of the scientific research projects that are currently relevant to the BIOSTORE project.


Stabilization of High-capacity Ni-rich Cathodes via Doping and Tailored CEI Chemistry within the INERRANT Project

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Lithium-ion batteries have become the primary energy storage technology driving electrification of the mobility sector. For this sector, the development is currently being driven in two different directions: design-for-performance materials which focuses primarily on high energy density and design-for-cost which makes a compromise on performance to reduce cost. The Battery 2030+ Umbrella project INERRANT will address both classes of materials through the development of Ni-rich layered oxide and LMFP cathodes. This contribution will focus primarily on the Ni-rich cathode materials and will demonstrate the holistic approach to cathode development.

Although Ni-rich layered oxides promise high capacities and energy densities, structural instabilities lead to capacity fade during cycling. Above potentials of 4.2 V, a c-axis contraction negatively impacts the structurhas a negativecan have deleterious effects on structural

stability and subsequent volume changes can lead to intergranular cracking of polycrystalline Ni-rich secondary

particles which exacerbates with repeated cycling.21(b) Freshly exposed Ni-rich surfaces are then susceptible to

reactions with the surrounding electrolytes which further promotes degradation of the cathode material. Recent work

by UCD partner to develop operando muon spectroscopy methods has allowed to track diffusion kinetics as a

function of state of charge.22 The advent of single-crystal cathode morphologies presents an opportunity to mitigate

cracking phenomena and provide a platform for the introduction of surface coatings which could generate a more

robust CEI.23 Coating cathode materials via spray drying (a process that allows upscaling) has been demonstrated by

the Fraunhofer partner.24 Recently, the UCD partner have also demonstrated that small additions of dopants such as

W result in the formation of amorphous LixWyOz surface phases which can play a role in surface stability

Despite that different P-containing compounds such as LiPF2O2,37(a), phosphane oxide,37(b)

phosphanes,38(a) and phosphazenes,38(b) have been introduced as electrolyte additives for SEI and CEI formation, there

is strong demand for novel additives to further improve LIB performance. Novel electrolyte additives, developed in

parallel to the cathode material and coatings is essential. Some additives contain a single F atom or perfluorinated

substituents. Fluorinated groups that have been added either via the additive, e.g. LiPF2O2, the liquid component of

the electrolyte, e.g. fluoroethylene carbonate, or Li[PF6], are crucial for the CEI formation. Li[(C2F5)3PF3] (LiFAP)

has been studied as component for LIBs in the past, 39(a) while this synthesis has been established on a kilogram scale

and used in follow-up chemistry,39(b) by the JMU partner. Although a number of derivatives of the phosphorane and

the FAP anion are known, e.g. OP(C2F5)3 and [(C2F5)2PO2]-, studies in the context of LIBs are very rare



BATTERY 2030+ Annual Conference 2025

High-power recycling: upcycling to the next generation of highpower anodes for Li-ion battery applications

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The popularity of Lithium-ion batteries (LIBs) has been on the rise, especially since the global push for achieving 'net zero' emissions. High-power anodes have gained significant attention within the LIB field due to their enhanced safety and ability to rapidly charge and discharge, making them essential for high-power applications like drones, motorsports, power tools, and high-performance electric vehicles (HEVs). Lithium titanate (LTO) has been a common choice in commercial high-power batteries. However, LTO has several limitations, including a low theoretical capacity and low energy density due to its high operating voltage (1.55 V Vs Li⁺/Li) [1]. To address these shortcomings, there is growing interest in niobate anodes as the next-generation solution for high-power applications. With the growing interest in niobium-based anodes for high-power LIBs, LTO anodes will be superseded, and as such an efficient and effective method of recycling needs to be considered. These anodes feature a Wadsley-Roth structure that incorporates corner-sharing distorted octahedra (ReO₃ -like structure). These channels facilitate rapid diffusion of Li⁺ ions. Additionally, the presence of crystallographic shear planes, created by edge-sharing octahedra, accommodates the oxygen deficiency and enhances the overall stability of the niobate structure [2, 3].

As part of an investigation into LTO recycling, an approach for upcycling this current-generation anode material into the next-generation titanium-doped Wadsley–Roth anodes has been developed. The approach involves an ion exchange process, where Li⁺ cations within LTO are exchanged for protons from acidic solutions, enabling the recovery of anatase (TiO₂) for the synthesis of TiNb₂O₇ and Ti₂Nb₁₀O₂₉. Complete H⁺/Li⁺ exchange is achieved using HCl and H₂SO₄, yielding pure anatase, while Li is recovered as Li₂SO₄ and LiCl. In contrast, H₃PO₄ treatment results in anatase and LiTiOPO₄, limiting full Li extraction but allowing Li recovery as Li₃PO₄. Electrochemical testing of upcycled TiNb₂O₇ and Ti₂Nb₁₀O₂₉ in Li-half cells demonstrates high capacity and rate capability (0.1–2 A g⁻¹) comparable with the pristine materials, highlighting its potential as a high-power anode. This process offers an efficient route for LTO recycling, lithium recovery, and next-generation anode material synthesis [4].



Figure 1. (a) XRD patterns of pristine LTO (Gelon) and the obtained solids after acid exchange with 2 M: HCl, H₂SO₄, and H₃PO₄. Reference plots of LTO and TiO₂ (anatase) added. (b) XRD patterns of the crystals obtained from the LTO-acid exchanged solutions.

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BATTERY 2030+ Annual Conference 2025

Unraveling metal plating/stripping mechanism via operando EQCM-D

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The Electrochemical Quartz Crystal Microbalance with dissipation monitoring (EQCM-D) is a highly sensitive and versatile technique for investigating metal plating/stripping, providing valuable insights into mass and morphological changes on metal electrodes under real-time operating conditions.¹ In this study, the zinc plating/stripping processes were examined using operando EQCM-D and hydrodynamic spectroscopy, complemented by additional ex situ techniques.² A novel strategy was introduced to form a zincophilic interphase in situ through indium co-deposition during cycling, facilitated by the use of InCl₃ as an electrolyte additive. Our results demonstrate the multifunctional role of the indium-containing additive in influencing zinc deposition and mitigating side reactions. This study underscores the potential of metal ion co-deposition as a method for controlling zinc plating/stripping and highlights the advantages of the EQCM-D technique for studying metal plating/stripping processes.



Figure 1. Comparison of the Coulombic efficiency of Zn plating/stripping with and without 1 mM $InCl_3$ obtained from EQCM-D measurements and the mpe value of the cell with 1 mM $InCl_3$ additive during Zn plating/stripping cycles.

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https://doi.org/10.1021/acsmaterialslett.4c01561



High performance of nanostructured lead-acid batteries using rGO as additive for negative electrode

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Lead-acid batteries (LABs), while widespread, have limitations in terms of capacity, durability, and performance at high charge and discharge rates and high depths of discharge. Currently lead-acid batteries consist of a sequence of plates formed by a lead grid and a porous paste compressed on it. This configuration causes a very low utilization of the active material and promotes the hard sulphation. Almost all the problems encountered in the LABs could be solved by modifying the morphology of the electrodes and, in particular, using nanostructured materials such as proposed in our works [1, 2, 3]. As demonstrated in these works, the use of nanostructured electrodes of PbO₂ and Pb permits to obtain very good results in terms of active material utilization and duration under deep-cycling conditions. Nanostructured battery can operate at high C-rates up to 30C [4, 5] [6] for a very high number of charge/discharge cycles. In addition, these batteries have also shown good results over on a wide temperature range [7, 8] and with the use of gel electrolytes [9]. The increase on performance can be attributable to the nanostructured morphology that ensurer a considerable surface area and consequently a high number of reactive sites for redox reactions [10]. In this work we present a 12V leadacid batteries with nanostructured electrodes and using a Pb nanostructured electrode added with reduced graphene oxide (rGO) to improve their performance. This battery was cycled at high C-rate. In particular, we used a C-rate equal to 10C (6 min to complete charge) and imposing a very deep discharge. These cycling conditions are much more stressful in terms of cut-off and charge/discharge rate in comparison to the parameters usually used for commercial batteries. The rGO contributes to better electrical conduction and decreases internal resistance, for higher efficiency during charging and discharging and a decrease in sulfation. Modified batteries exhibit higher specific capacity, longer cycle life, and better reaction to high currents than classic PBAs. The results obtained indicate that the use of rGO is a viable strategy for improving the performance of lead-acid batteries, with possible applications in sectors such as industry and energy storage.

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Direct Recycling of All-Solid-State Batteries with a Halide Solid Electrolyte via Water-based Separation: Interactions of Electrode Materials in Aqueous Li₃InCl₆ Solutions

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As the demand for all-solid-state batteries (ASSBs) grows, their recyclability remains an overlooked yet essential aspect of long-term sustainability. Developing effective recycling strategies requires careful consideration of different electrode-electrolyte combinations [1]. Among solid electrolytes, halide-based Li₃InCl₆ stands out due to its remarkable structural reversibility as it can regain its initial crystal structure after dissolution in water, without significant electrochemical degradation [2]. This unique property enables a direct recycling strategy, where the electrolyte can be selectively separated from electrode materials upon dissolution in H_2O .

To evaluate its recycling potential, the dissolution behavior of Li_3InCl_6 is examined in the presence of different electrode materials ($Li_4Ti_5O_{12}$, $LiCoO_2$, $LiMn_2O_4$, carbon-coated $LiFePO_4$, $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$, and $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$). The underlying interactions, primarily governed by Lewis acid-base reactions, are analyzed using X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), and inductively coupled plasma mass spectrometry (ICP-MS). These interactions significantly impact the electrochemical properties of both the recycled Li_3InCl_6 , and the recovered electrode materials compared to their pristine counterparts.

Furthermore, a structural description of the reversible reaction mechanism of Li₃InCl₆ and its hydrated counterpart is provided, highlighting its potential for efficient electrolyte recovery in next- generation battery recycling.

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Molecular Design of Film-Forming Additives for Lithium-Ion Batteries: Impact of Molecular Substrate Parameters on Cell Performance

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The development of high-energy-density lithium-ion batteries (LIBs) relies heavily on the formation of a stable and efficient solid electrolyte interphase (SEI), which is significantly influenced by the molecular properties of organic film-forming electrolyte additives. However, many aspects of SEI formation and decomposition remain poorly understood, necessitating a thorough investigation of the underlying mechanisms.

This study explores the impact of molecular substrate parameters in *N*-carboxyanhydrides (*N*-CAs) as electrolyte additives on SEI formation and the electrochemical performance of LIBs. Earlier studies exemplified the beneficial impacts of *N*-CAs on the performance of Si-containing LIB cells when used as an electrolyte additive.^[1] A series of *N*-CA derivatives was designed and synthesized to examine the role of steric effects and electronic properties in polymerization and film formation within NCM-811||Si/graphite composite cells.

To elucidate decomposition pathways and their dependence on functional moieties, electrochemical performance analyses were conducted. The results revealed that additives with sterically demanding moieties exhibited varying degrees of performance enhancement, with steric bulk playing a crucial role in SEI flexibility and stability. Additionally, laser desorption/ionization mass spectrometry (LDI-MS) provided insights into decomposition mechanisms and polymer formation, demonstrating a direct correlation between functional group variations and SEI structure.

These findings emphasize the importance of molecular design in electrolyte additive development, particularly in fine-tuning steric and electronic effects to optimize SEI properties. This study offers valuable insights to guide future research toward a creative yet systematic and innovative approach to electrolyte additive design for high-energy-density LIB cells.



Figure 1. Molecular Design of *N*-Carboxyanhydrides (*N*-CAs) as electrolyte additives for Lithium-Ion Batteries (LIBs) and the Analysis of the formed Solid Electrolyte Interphase (SEI).

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A Sensor–Actuator Platform for In-Situ Battery Health Monitoring and Self-Healing

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Monitoring the state of health of batteries is a prerequisite for implementing triggered self-healing strategies [1]. Regardless of the cell chemistry or the specific method used to modify cell performance, achieving this relies on two pillars: digitized information about cell parameters and externally controlled healing actuation.

The HealingBat project has developed a versatile platform for in-situ sensing and actuation, based on embedding an array of planar sensor/actuator sites within the battery cell. Each site includes an interdigitated electrode area as the electronic interface, onto which functional sensor materials can be printed.

The device is fabricated using planar microsystems technology on a flexible, electrically insulating, and chemically resistant polyimide substrate. Aluminium (AI) forms the metallized layer for the electrode structures. Functional materials are deposited via colloidal inks based on tailor-made materials, enabling selective and sensitive detection of temperature, pressure, and gas species, as well as thermal actuation. A second polyimide layer insulates the sensing structures from the battery environment. The total device thickness is under 30 μ m, and the feed line width has been reduced to 900 μ m to minimize any impact on battery performance.

The first-generation device is shown in Figure 1. It has been designed for easy integration into the standard pouch cell assembly process, enabling reliable sensor deployment.



Figure 1. First generation of the HealingBat technology platform. (a) Photolithography mask layout for the Almetallization of the polyimide substrate. (b) Finished sample device and a scanning electron microscope image of a functionalized site. (c) Pouch cell during assembly with the technology platform.

Work is underway to integrate the sensors into a battery management system that will collect sensor data to monitor the battery's state of health, trigger and control self-healing actuators, and manage safety mechanisms by verifying the integrity of actuation signals.

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Capturing complexity with Synchrotron X-rays: Operando analysis of batteries from multiple perspectives

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Energy storage devices such as batteries are exquisitely complex chemical systems with multiple components interacting in ways dependent on the operating conditions. Consequently, developing tools that enable the simultaneous measurement of structural and chemical changes together with properties is critical to understanding performance. The beamline portfolio of MAX IV with X-ray energies from soft X-rays to hard X-rays provides complemental techniques to investigate batteries operando from multiple perspectives. In this poster we will demonstrate how a combination of techniques provides insight into complex structural and chemical transformations during charge and discharge of batteries from complemental perspectives. We will focus on a) soft X-ray photoemission spectroscopy and b) multimodal combinations of hard X-ray absorption spectroscopy and diffraction. The brilliance of the X-rays provided at MAX IV as well as the scanning equipment at the beamline end stations allow for high time-resolutions, and thus to follow chemical reactions like the charge and discharge of batteries in real-time.

Our recently developed tools for simultaneous multimodal combination of X-ray spectroscopy and diffraction allow for real-time *operando* measurements with sub-second time resolution. For the combination of XAS and XRD we developed arbitrary energy motion trajectories and hardware gate based data-acquisition schemes to allow for quick real-time measurements of several absorption edges combined with several X-ray diffraction exposures at various energies. Such complex measurement sequences are then executed repeatedly during operando battery cycling. This is of particular interest when investigating multinary compounds operando, e.g. Li-ion battery cathodes such as NMC or LNMO. A "simultaneous" measurement of the Ni and Mn K-edges combined with two XRD exposures can be performed repeatedly with an overall time-resolution of down to 2s, allowing for operando battery cycling experiments under high device charge/discharge currents.

Such quick multimodal measurements combined with a relatively small spot size of < 100um even allow mapping to determine spatial inhomogeneities rather than just measuring an averaged state of the device during cycling. We further developed the capability to measure up to 5 battery devices during the same time by sequentially repeating multimodal measurements on the individual devices.

Within this contribution, we will present those recent technical developments at the beamline and data acquisition system with specific scientific examples from LNMO and NMC cathodes. We will further present operando mapping results and discuss X-ray beam induced effects during investigation of electrochemical devices with highly intense hard X-rays.



DIRECT REUSE POTENTIAL AND PERFORMANCE OF RECYCLED GRAPHITE FROM LOW IMPACT AND SUSTAINABLE DIRECT RECYCLING PROCESS

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In recent decades, there has been a sharp rise in demand for Lithium-ion batteries (LIBs), accompanied by a scarcity of essential resources, resulting in a projected demand-to-supply ratio surpassing double by 2030. While increased mining and raw material extraction efforts could alleviate some supply constraints, they also pose significant challenges such as environmental pollution, safety risks, socioeconomic disparities, supply chain dependencies, and global geopolitical implications. Addressing these complex issues requires a shift towards a circular economy model in the LIBs industry to reduce reliance on extraction activities and the fragile supply chain. A pivotal strategy involves adopting highly efficient and eco-friendly processes for recycling LIBs. This approach not only creates sustainability but also unlocks innovative and profitable opportunities within the sector while promoting localized value chains. Thus, there is a pressing need for cost-effective, efficient, and high-yield recycling and reutilization strategies for graphite, highlighted by new EU regulations (Battery Regulation and Critical Raw Materials) mandating the use of recycled battery materials by 2030. The Battery Regulations currently don't require a minimum content of recycled graphite in batteries for the next 10 years, however, novel approaches that open new pathways might change this viewpoint. This study introduces a pioneering thermal recycling process designed to yield high-quality graphite from LIBs with a cost-effective approach, specifically tailored for remanufacturing in LIB applications. Detailed electrochemical performance of these materials in half-cells and recycled graphite ||NMC811 full-cells are examined, juxtaposed against commercial graphite benchmarks. The recycled graphite demonstrates notably high reversible specific capacities (e.g., ~350 mAh g-1 at C/20) with exceptional coulombic efficiencies (>99%) in half-cells. Furthermore, pouch cells utilizing recycled graphite ||NMC811 full-cells exhibit commendable cycling stability at C/3 for 1000 cycles, retaining 80% of their capacity. Importantly, comparison with pristine graphite in full-cells reveals comparable performance, emphasizing the potential of recycled and repurposed graphite as a significant step towards sustainable and reliable LIBs, aligned with the vision of the circular economy. In summary, the development of advanced recycling technologies for LIBs, such as the thermal recycling process for graphite highlighted in this study, holds promise in addressing critical resource challenges while advancing sustainability goals in the battery industry.



Figure 1. Capacity Retention versus Cycle Index for the Pouch-Cells test for Recycled Graphite. **REFERENCES**

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OPERANDO HARD X-RAY PHOTOELECTRON SPECTROSCOPY OF ANODE-FREE SOLID-STATE BATTERIES

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The safety profile of batteries is greatly improved by replacing toxic and flammable liquid organic with solid-state electrolytes. The accompanying suppression of Li dendrite formation enables the utilization of Li metal anodes (LMA). The energy density of solid-state batteries is further increased in the so-called anode-free configuration, in which the precise amount of Li needed for the battery operation is transferred from the counter electrode to the working electrode in-situ during cell charging. Unfortunately, most solid electrolytes (SEs) are not electrochemically stable at LMA operation potentials. SE reduction reactions and the formation of solid electrolyte interphases (SEI) causes lithium inventory losses and impedes the lithium transport kinetics, leading to poor battery cycle life. In anode-free batteries, the SEI is a buried interphase, making it difficult to probe by chemical-sensitive surface science methods, such as X-ray photoelectron spectroscopy. Ex situ / post-mortem studies, provide only a limited view on the underlying processes as the reactions taking place during cell operation may result in intermediates and products that are only stable under operating conditions. Here, we present a novel approach for probing buried interfaces at operating conditions by hard X-ray photoelectron spectroscopy (HAXPES), in which an ultrathin (<10 nm) uniform metal film is deposited on top of the SE as a current collector that is sufficiently transparent for photoelectrons with high kinetic energies. Three exemplary systems will be shown in this work, where the growth of the SEI will be monitored by depth-resolved in situ and operando HAXPES: (1) an inorganic SE cell based on a Li₆PS₅Cl electrolyte at room temperature, (2) a cell with a composite polymer electrolyte (polyethylene oxide with lithium bis(trifluoromethanesulfone)imide salt, PEO+LiTFSI) operating at 50°C, and (3) a cell with an organic/inorganic composite comprised of 40 wt% Li₆PS₅Cl in a PEO+LiTFSI matrix. It will be demonstrated that using HAXPES, it is possible to monitor plating/stripping cycles, revealing new insights into the underlying mechanisms of SEI growth. In particular, by combining the insights derived from examples (1) - (3), the role and interplay of organic and inorganic parts of the composite for the SEI formation will be discussed. Finally, the application limits of this novel approach will be revealed.



BATTERY 2030+ Annual Conference 2025

High-Performance Supercapacitors from Olive-Leaf-Derived Activated Carbon

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In this study, olive leaves, widely recognized as an agricultural by-product, are employed to synthesize high-performance activated carbons (ACs) for supercapacitor (SC) applications. Three chemical activators, i.e., NaHCO3, H3BO3, and KOH, are used to derive distinct porous ACs labeled OLAC-N, OLAC-H, and OLAC-K, respectively. Structural and morphological characterization through X-ray diffraction, Raman spectroscopy, scanning electron microscopy, and Brunauer-Emmett-Teller analysis indicates significant porosity and specific surface areas up to 1420 m2/g for the KOH-activated OLAC-K. Scanning electron micrographs also reveal a well-developed pore network and uniform pore-size distribution. Electrochemical evaluations, performed in both 1 M H2SO4 and 6 M KOH electrolytes using cyclic voltammetry, galvanostatic charge-discharge, and electrochemical impedance spectroscopy, consistently identify OLAC-K as the top performer. This electrode exhibits a specific capacitance of 128 F/g and demonstrates 96% capacity retention over 5000 cycles at 2 A/g in 1 M H₂SO₄, outstripping the other variants. The synergy between KOH activation and the acidic electrolyte is inferred to promote a well-developed pore network, facilitating favorable ion diffusion and charge-transfer kinetics. By harnessing agricultural waste as a sustainable carbon source, the work underlines an environmentally friendly route to high-performance energy-storage materials. The findings suggest further exploration of biomass-derived carbons to refine supercapacitor designs and bolster future clean-energy innovations.



Figure 1. Graphical Abstract.



Development of robust hydrometallurgical process for battery recycling in REVITALISE

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Abstract:

As the need for effective battery waste management grows, it is important to recycle the batteries in order to sustain the supply of materials, mitigating the environmental impact due to end-of-life (EoL) batteries and natural resource exploitation activities¹. Traditional methods for recycling prove inadequate, lacking efficiency, and eco-friendliness. Therefore, new methodologies have been developed to counter these challenges, mainly through hydrometallurgical approach². These technologies have proven to be efficient but rapid change in battery chemistry is making recycling process difficult since, each chemistry requires specific recycling process^{3,4}. Another problem is the low recovery of Li (lithium) including its separation and purification. To address these challenges, in REVITALISE we proposed early-stage (selective) recovery of Li using mild hydrometallurgical approach and robust total acid leaching to recover cathode active materials from spent batteries. Li selective leaching by water was proposed to ease its separation and purification, which in current work involves liquid-liquid extraction, antisolvent crystallization and cooling crystallization techniques. This flow sheet was proposed to make a closed loop process through utilization of intermediate and side products including impurities back into main recycling stream. Herein, we report that Li was successfully leached, separated and purified as Li₂CO₃ from NMC black mass under optimized conditions. The obtained product has a purity of 80% and a recovery rate of 90%.

While total acid leaching was opted because it can facilitate making a robust process for different NMC and LFP battery chemistries. A design of experiments (DOE) was done to understand the interaction of lixiviants with NMC 111, NMC 622 and NMC 901, respectively. For this, inorganic acids H₂SO₄, HCl, and HNO₃ were used to leach the metals from the NMC black mass. To understand the effect of leaching, several input variables including pulp density, temperature and concentration of acid used were selected. A multilinear regression model was used to analyze the data from DOE to select the ideal lixiviant for leaching. Modelling results showed better regression coefficient for chloride leaching as compared to other lixiviants. Thus, it was observed that HCl is good to be used as it is not affected by the type of battery chemistry and no reducing agent is required for leaching.

REVITALISE is a research project that aims to develop low cost and green process recycling technology to recover of full range of battery materials funded by European Climate Infrastructure And Environment Execution Agency (CINEA) under the grant agreement number 101137585. **REFERENCES**

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BATTERY 2030+ Annual Conference 2025

RECYCLING AND REGENERATION OF CARBON-COATED LCO FROM SPENT LIBS

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Spent lithium-ion batteries (LIBs) are an excellent secondary resource of critical minerals such as lithium, cobalt, and nickel. Nevertheless, the existing recycling methods have a significant carbon footprint and are energy intensive. Because of the innate heterogeneity of battery materials based on different chemistries, the extent of recycling is largely source dependent, making environmentally benign processing of LIBs essential for the extraction and reuse of important minerals [1]. In this context, the development of the microwave-assisted hydrometallurgical recycling process employing mixed organic acids has been examined for a cobalt-rich spent LIB blackmass by varying various parameters such as acid concentration, reducing agent dosage, and microwave power. The leaching operation was optimized to achieve leaching efficiencies of Li: 96.47 %, Co: 95.10 %, Ni: 94.80 % and Mn: ~100 %. Up to 95% of metal values were coprecipitated in the form of a metal-organic coordination complex, followed by further processing to regenerate carbon-coated lithium cobalt oxide (LCO) cathode. Lastly, lithium was recovered as high-purity (~98%) lithium carbonate. The green route enables efficient recycling of spent LIBs, which paves a pathway for generating eco-friendly value-added products with high throughput processing towards a sustainable future.

Keywords: Spent LIBs; Recycling; Microwave-assisted processing; Hydrometallurgy; Organic acids.



Figure 1. Process flow diagram

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Highly Concentrated Imide-Based Aqueous Electrolytes for Supercapacitors

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Based on their high gravimetric and volumetric energy and power, long lifetime and reasonable cost, to date lithium ion batteries (LIBs) are the dominant electrochemical energy storage technology. Nevertheless, there has been an increasing emphasis on research directed towards the development of alternative battery technologies, characterized by their enhanced sustainability and potentially higher safety. Batteries utilizing highly concentrated aqueous electrolytes are promising based on their low flammability and potentially low toxicity, but suffer from a narrow electrochemical window. This, however can be expanded when utilizing high conducting salt concentrations, enabling their application in certain LIBs or supercapacitors.[1-3] Though the underlying mechanisms remains to be fully elucidated, the origin is believed to be an altered solution structure and electrolyte electrode interface and interphase.[2]

The focus of this study is on the investigation of different highly concentrated imide-based aqueous electrolytes by means of Raman spectroscopy to gain deeper insights into their ion arrangements.[3] Thereby, the impact of the salt concentration and anion size are discussed. In addition, the electrochemical properties and the performance of various highly concentrated aqueous electrolytes is demonstrated in supercapacitors and potential influences are addressed.

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Economic Viability of LFP Battery Recycling in Europe: Challenges and Optimization Strategies

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In recent years, the electric vehicle market in Europe has expanded rapidly, with lithium iron phosphate (LFP) batteries gaining significant market share due to their lower cost compared to nickel-manganese-cobalt (NMC) alternatives. However, due to the low value of their primary materials, LFP batteries are currently unprofitable to recycle [1,2]. Given their average lifespan of approximately eight years, large volumes of LFP battery waste will emerge within the next five years [3]. At the same time, EU regulations require minimum recycling efficiencies and recycled content in new batteries [1]. Beyond waste management, recycling could also reduce Europe's reliance on imported lithium.

This study assesses the economic feasibility of LFP battery recycling by modeling an industrial-scale process that integrates mechanical pre-processing with cathode material recovery. A techno-economic analysis is conducted to identify key cost drivers and evaluate potential pathways to improve profitability. Two scenarios are examined: the current state of battery recycling and an optimized process stream that could be implemented within five years.

The findings show that existing methods are unprofitable mainly due to limited product yields and the high costs required to achieve sufficient purity of the output materials. In Europe, achieving the necessary purity levels without advanced automation is costly due to high labor expenses. Thus, the optimized scenario explored in this work focuses on direct recycling combined with automation, leveraging existing and emerging technologies. In this optimized scenario, economic feasibility improves, and sensitivity analysis indicates that break-even point can be achieved under favorable lithium prices and cost reductions in labor and equipment. Therefore, a combination of these factors could make LFP recycling an economically viable strategy for battery circularity and securing critical raw materials.



Figure 1.Cost breakdown for the currently available recycling of LFP via hydrometallurgy and direct recycling, and of an optimized scenario using direct recycling.

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Unravelling Electro-Chemo-Mechanical Processes in Graphite/µ-Si Composite Electrodes for Evidence-based Design of Advanced Microstructures

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Silicon is a promising negative electrode material for high-energy automotive batteries, but its significant volume changes during cycling cause rapid degradation, limiting its loading to just 10 wt.% in commercial graphite/Si composite negative electrodes as a compromise between energy density and cycle life. Overcoming this threshold requires evidence-based design of advanced electrodes[1]. Here we combine operando optical microscopy[2], synchrotron X-ray CT 4D imaging, digital image/volume correlation and machine learningassisted image processing techniques, to elucidate the multiscale electro-chemo-mechanical processes in graphite/µ-Si composite negative electrode[3]. Presented with multimodal high-resolution videos, here we show the expansion of porous μ -Si particles strongly depend on the morphology of the intra-particle porosity. This challenges the ubiquitous assumptions about porous Si in mitigating volume effect. Moreover, charging current overload in graphite particles and abnormal lithiation of Si against thermodynamics are shown, which increase the risk of early lithium plating and capacity yet are often overlooked. Surprisingly, the electrode expansion is not necessarily governed by Si; rather, its influence only becomes pronounced at high SOCs during the first lithiation cycle but is dominated by graphite in the subsequent cycles. Severe thickness expansion (20%) and reduction in nano-pores (from 43% to 21%) are observed in the CBD, undermining accessible capacity and fast charging capability. Finally, in response to the five identified major challenges in graphite/m-Si composite electrodes, we develop a double-layered graphite/m-Si composite negative electrode, which demonstrates significantly lower polarization and mitigated capacity decay compared to its homogeneous counterparts. Overall, this study provides a comprehensive framework for advancing Si-based negative electrodes through hierarchical engineering, from particle level to the 3D architecture of the electrode.



3D Microstructural evolution vs. Performance

Operando optical microscopy

Multiscale electro-chemo-mechanical processes

Figure 1. Coupling multimodal operando microscopy to reveal the multiscale microstructure evolution and electro-chemo-mechanical processes towards design of advanced electrodes.

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Evaluation of synthesis parameters for Ti₃C₂ MXenes application in lithium-sulphur batteries

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Nowadays lithium-ion batteries (LIBs) are the most spread and reliable energy storage technology, but they also show some drawbacks that hinder the possibility to satisfy the rising demand for energy storage devices. Consequently, the study and development of alternative post-lithium-ion technologies is of crucial importance to make them commercially viable in the future. Among these systems, lithium-sulphur batteries (LiS) are one of the most interesting due to the high theoretical specific capacity of sulphur (1672 mAh g^{-1}). However, the practical application of LiS is still strongly limited by some issues. [1] The most impactful one is the so-called *shuttle effect*, caused by the diffusion of soluble lithium polysulphides formed upon cell discharge. To prevent this, different materials capable of binding these soluble species and catalysing their reduction reaction have been proposed as cathodic sulphur hosts. [2]

The present work aimed at studying a recent class of bidimensional metallic carbides and nitrides, discovered in 2011 and known as MXenes [3], and their direct and simple application in the field of lithium-sulphur batteries. Particularly, four samples of bidimensional titanium carbide, in the form of Ti_3C_2 , were prepared by varying the parameters of the classic synthesis, using a more sustainable process. The different samples of Ti_3C_2 were synthesized and characterized using physico-chemical and electrochemical techniques to evaluate their affinity to lithium polysulphides and their potential activity as catalysts for sulphur reduction reactions. Then, different cathodic formulations and the synthesized materials were studied in full cell applications.

The obtained results showed that the combination of low acid concentrations and higher reaction time allows to synthesize MXenes characterized by good morphological, physico-chemical and electrochemical properties overall. During full cell study, instead, we noticed that the employment of a thermal treatment during composite cathodic material production leads to better homogeneity and stability of the final system. In addition, using MXenes as sulphur host limits polysulphides shuttling and stabilizes cell specific capacities.

The best synthesized MXene sample was then furtherly tested up to 500 cycles and compared with standard carbon black-based sulphur cathodes. Although the obtained values are still far from the maximum theoretical specific capacity, they are promising thanks to their higher stability and capacity retention features.

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Electrochemical enhancement of Mn and Co co-doped LaNiO₃ perovskite oxides for high-performance supercapacitor applications

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Perovskite oxides are very popular for energy storage applications and show high stability characteristics in supercapacitors. Doping in perovskite oxides can further enhance their performance. Herein, the synthesis of cost-effective co-doped perovskite material for energy storage devices has been discussed. The sol-gel method is used to prepare the co-doped (LaMn_xCo_yNi_{1-(x+y)}O₃) perovskite oxide having the ratio (x = 0, y = 0), (x = 0.02, y = 0.08), and (x = 0.08, y = 0.02). Hexagonal crystal structures were confirmed by X-ray diffraction (XRD), which revealed enhancement in lattice constants as the Co concentration increased. The X-ray photoelectron spectroscopy (XPS) confirmed the presence of La³⁺, Ni³⁺, Ni²⁺, Co³⁺, Mn²⁺, and O²⁻ ions on the surface of the synthesized electrode material (LaMn_xCo_yNi_{1-(x+y)}O₃). Electrochemical performance was tested in 6M KOH aqueous solution of all materials, and it shows an enhancement in specific capacitance by increasing the concentration of Cobalt. An asymmetric supercapacitor (LaMn_{0.02}Co_{0.08}Ni_{0.9}O₃/separator/AC) was also fabricated and tested in two electrode systems.



Figure 1. About supercapacitor



Optimizing Waterborne LNMO Cathode Production: Evaluating Alternative Binders for Enhanced Electrochemical Performance and Processability

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Lithium-ion batteries (LIBs) are essential for the global transition to sustainable energy, especially in electric transportation and large-scale energy storage. As the demand for more sustainable solutions grows, optimizing cathode production processes has become crucial to improving LIB performance and scalability. "In this context, Lithium Nickel Manganese Oxide (LNMO) stands out as a promising high-voltage cathode material, offering key advantages over conventional Nickel Manganese Cobalt (NMC) due to its cobalt-free composition, which enhances sustainability and eliminates dependence on cobalt-related costs, both monetary and ethical. To further promote sustainability in battery production, it is also important to study and encourage the usage of innovative water-compatible binders for waterborne processes, aiming to improve both environmental impact and performance efficiency. This study investigates the role of alternative binders in a waterborne pathway for LNMO cathode fabrication, comparing their electrochemical performance and processability against the already commercially used Carboxymethyl Cellulose (CMC). Three alternative binders - Guar Gum (GG), Lithium Polyacrylate (Li-PAA), and Sodium Alginate (ALG) - are thus systematically evaluated. The objective is to determine their suitability as standalone binders and explore the potential benefits of blended formulations to enhance cathode performance. A Design of Experiments (DoE) approach is employed to optimize binder selection, aiming to achieve superior electrochemical stability together with high processability. Characterization techniques include galvanostatic cycling [Figure 1], Potentiostatic Electrochemical Impedance Spectroscopy (PEIS), Fourier Transform Infrared Spectroscopy (FTIR), and rheological assessments (shear rate, amplitude, and frequency sweeps). Preliminary findings indicate that the proper binder selection significantly influences electrode processing and electrochemical behavior, highlighting the potential of alternative formulations for improved performance. This study provides valuable insights into waterborne LNMO cathode production for Lithium-ion batteries, emphasizing the potential of optimized binder combinations to significantly enhance both electrochemical performance and processability, ultimately contributing to the development of more sustainable, cost-effective, and efficient LIBs.

Acknowledgements:

Funded by the European Union under grant agreement N° 101069707. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Climate, Infrastructure and Environment Executive Agency (CINEA) Neither the European Union nor CINEA can be held responsible for them.



Figure 1. Brief comparison of the rate capability and long cycling tests carried out using different binders in a LNMO cathode formulation



Analysis of the Franklinite-Gahnite (ZnFe_{2-x}Al_xO₄) Solid Solution Series: Insights into the Use of Doped Ferrites as Active Material for Zinc-ion Batteries

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The cubic spinel franklinite (zinc ferrite, $ZnFe_2O_4$) is a well-studied material with a comparatively small band gap of 1.9 eV and widely studied magnetic properties [1,2]. This makes it a compelling candidate for a number of different applications ranging from photocatalysis to data storage. Recently, there have also been attempts to use zinc ferrite as an active material at the negative electrode of lithium-ion batteries (LIBs) due to its high theoretical volumetric capacity of 1142 mA h cm⁻³, but these have failed due to the poor chemical stability of the material during repeated lithium (de)insertion [1-3]. In contrast, first attempts to use doped zinc ferrite as an active material at the positive electrode of zinc-ion batteries (ZIB) showed promising results [4]. Compared to the standard tetragonal spinel material $ZnMn_2O_4$, the cubic titanium-doped ferrite in this study showed higher potentials vs. Zn/Zn^{2+} . However, the problems of transition metal leaching in contact with aqueous electrolytes were similar to those of manganite.

In the present study on the solid solution series of franklinite-gahnite, the iron is partially or completely replaced by AI^{3+} . Since iron leaching occurs especially when Fe^{3+} in the structure is reduced to Fe^{2+} upon Zn^{2+} insertion, a sufficient amount of non-redox-active AI^{3+} could stabilize the structure in contact with aqueous electrolytes, but will strongly affect the ionic conductivity and possibly the cyclability. According to LIB studies, there might be a sweet spot in the range below x = 1.0 for $ZnFe_{2-x}AI_xO_4$ [5]. In addition, increasing the aluminum content will result in lower electron conductivity due to an increased band gap [6], which is not necessarily desirable for an active material, but can be compensated by techniques such as carbon coating. It is also expected that the magnetic and optical properties will change significantly with increasing Al content.

Homogeneous powders with the composition $ZnFe_2-xAl_xO_4$ (x = 0.0 to 2.0) were synthesized by a Pechini type synthesis method. Calcined powders were then used to produce electrode sheets for application in $ZnFe_2-xAl_xO_4$ ||Zn cells, while sintered ceramic pellets were used for materials level investigations. Using a variety of analytical techniques (XRD, impedance spectroscopy, CV, reflectance measurements, etc.) we succeeded in obtaining a holistic picture of the entire solid solution series, which will be discussed in terms of a defect model and potential applications in ZIB and beyond.

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Characterization of LFP Batteries Ageing from Automotive Applications via Single Cell Testing, Physical Modeling and Ex-Situ Analyses

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Lithium-ion batteries ageing interpretation is essential for assessing their further exploitability and to promote second-life and recycling solutions. While ex-situ analyses effectively evaluate the degradation state of a cell at material level and correlate it with capacity and power fade, they are destructive and prevent a further exploitation of the battery^[1]. Therefore developing a diagnostic methodology that provides similar information with minimal invasiveness is crucial. This study is part of AutoMAT European research project, which aims at developing aging-aware post-use strategies for lithium-ion batteries and the goal of this work is to investigate the ageing state of spent LFP lithium-ion batteries used in hybrid buses, which have faced a severe ageing in real automotive applications. This is achieved by developing a diagnostic procedure capable of determining their ageing state, distinguishing the contributions of the anode, the cathode and the electrolyte. The developed diagnostic methodology is then validated with the information gained from post-mortem analyses.

One of the goals is to establish a correlation among the field data gathered from the battery management system (BMS), and the cells degradation state. This is initially achieved through electrochemical analyses, such as quasi-OCP discharges, differential voltage analyses (DVA) and electrochemical impedance spectroscopy (EIS). The main driver of the cell degradation was the uneven operating temperature, leading to highly heterogenous aging within cell groups, with residual capacity ranging from 50% for the most aged cell to 75% for the least aged. Using the developed diagnostic method, 3 cells were selected: a pristine one and two samples at opposite aging states, which demonstrated to gather capacity retention of 78% and 48% respectively. The EIS of the three cells showed an increase in the high frequency resistance and an increase in the kinetic loop at intermediate frequencies.

A quasi-OCP model was adopted to determine the loss of lithium inventory (LLI) and the loss of active material (LAM) in the electrodes, identifying how cell degradation is mainly attributable to the reduction of cyclable lithium, followed by the LAM_n, while the influence of LAM_p results to be minimal.

To validate the results, post-mortem analyses were performed. Scanning Electron Microscopy (SEM) images allowed to determine the degradation state of the positive and the negative electrode. While the former did not display any significant change during operation, confirming a superior stability, the graphite, showed the formation of a dense superficial layer, mainly attributable to fluorinated electrolyte degradation products. This layer likely contributes to the increase in the HFR and to the loss of active material at the negative electrode, hindering lithium intercalation. Moreover this superficial layer proved to be easily removable by performing a washing with DMC, suggesting potential for performance recovery.

To quantify the loss of active material at the positive and at the negative electrode, half cells assembled with harvested materials from samples, were characterized. A good agreement was found comparing the capacity reduction showed by the half cells and the LAM estimated by the model. Eventually, to find a correlation among the loss of lithium inventory determined via physical modelling and the one that happened in the cells, X-Ray Diffraction Analyses (XRD) and Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) were performed on the cathode material its fully lithiated state, and the changes in composition among the three cells, were representative of the LLI determined via physical modelling analyses.

In conclusion the developed diagnostic procedure accurately identified the main degradation modes that occurred in the batteries during operation. The methodology proved to be effective in estimating cell ageing, which is probably linked to electrolyte degradation products that block the negative electrode active sites and consume lithium ions.

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BATTERY 2030+ Annual Conference 2025

Sulfur and sulfides as solid-state batteries electrodes

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Solid-state batteries (SSBs) hold the promise of a step improvement in safety and performance over liquid electrolyte-based batteries¹. However, they are still afflicted by significant problems, such as a narrow electrochemical stability window for most solid electrolytes, difficulty of creating an efficient ionic and electronic conductive network and significant mechanical degradation. Sulfur and sulfides (i.e. TiS₂ and CuS) are promising electrode active materials that can effectively address some of these challenges². Due to their moderate potential (compared to intercalation cathodes), these compounds should limit the electrochemical decomposition of the SE. Moreover, in the case of S, the use of an inorganic SE completely eliminates the formation of intermediate polysulfides and thus the detrimental shuttle effect, while sulfides, thanks to their limited volume fluctuation and high conductivity, greatly facilitates the formation of an efficient and resilient electronic and ionic conduction network. However, several open issues have prevented their application in commercial batteries so far.

In this work, lithium-sulfur batteries have been studied focusing on the still poorly understood interaction between sulfur and carbon. Our findings suggest that the current understanding of these systems may be incomplete and that the previously reported "sulfur spillover"³ effect may significantly influence the performance and potentially improve the battery fabrication process.

Our research on metal sulfides highlighted instead how the inclusion of a second metal center (e.g. Fe in combination with Cu, as in CuFeS₂) promotes stability by hindering the macroscopic displacement mechanism typical of these materials.

Finally, the reactions of those electrodes (e.g. $CuFeS_2$ and TiS_2) with Li has been observed for the first time in SSBs by operando techniques, revealing significant differences with the processes happening in liquid-electrolyte batteries.



Figure 1. Sulfur and sulfides as promising active materials for solid-state batteries.

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DEVELOPMENT AND INDUSTRIALIZATION OF NEXT GENERATION LITHIUM BATTERIES AT BASQUEVOLT

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BASQUEVOLT is a pioneering company in the development of next-generation solid-state batteries, with the goal of accelerating the transition toward safer, more efficient, and more sustainable electric mobility. Founded with the backing of a strong industrial and technological ecosystem, the company combines innovation, industrial capacity, and public-private collaboration to position itself as a European benchmark in energy storage.

Headquartered in the Basque Country, BASQUEVOLT develops technology for the world. Its business model is based on technological excellence, industrial development, and cooperation with key players in the sector.

The presentation highlights the electrolyte development towards new generation batteries going from gel type through the liquid to hybrid gel system. Scientific and engineering challenges will be discussed based on cell performance data. The differences in cell architecture significantly affects the transition from coin cell to 1Ah pouch and 20 Ah pouch cells. Adjusting electrolyte composition for proper fluidity plus cell engineering plays pivotal role in proper cell cycling. Wetting and pressure distribution become a challenge scaling up the cells. All the changes in the cell must consider safety as first keeping in ming economical aspects for future commercialization. A snapshot of cells performance progress will be given.





Development and Characterization of Anode Filaments for Fused Filament Fabrication of Sodium-ion Batteries

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Abstract

The transition to sustainable energy storage solutions has led to the exploration of sodium-ion batteries (SIBs) as an alternative to lithium-ion batteries. This study presents the development and characterization of a novel anode filament for Fused Filament Fabrication (FFF) of SIBs. The filament composition includes Polylactic Acid (PLA) as a polymer matrix, Hard Carbon as the active material, Poly(ethylene glycol) dimethyl ether (PEGDME) as a plasticizer, and Carbon Black Timcal Super-P to enhance conductivity. A mixture design approach was employed to optimize the formulation, ensuring homogeneous dispersion and improved electrochemical performance.

Rheological analysis demonstrated shear-thinning behavior advantageous for 3D printing, with comparable viscoelastic properties to conventional PLA filaments. Electrochemical characterization revealed that 3D-printed anodes exhibited a capacity of 230 mAh/g, approximately 15% lower than slurry-coated electrodes (273 mAh/g), due to reduced electrolyte infiltration and ion diffusion caused by PLA encapsulation of active particles. SEM and Raman spectroscopy confirmed uniformity in elemental distribution, while cyclic voltammetry indicated diminished electrochemical activity in 3D-printed samples. Despite these limitations, this research underscores the potential of 3D-printed anode filaments in next-generation sodium-ion batteries, offering a sustainable, scalable, and solvent-free fabrication route.

Keywords: Sodium-ion batteries, Fused Filament Fabrication, 3D printing, Hard Carbon, Electrochemical characterization.

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Figure 1. Schematic representation of filament fabrication and 3D printing process

DETERMINATION OF SOLID-PHASE DIFFUSION COEFFICIENTS AND REACTION-RATE CONSTANTS FOR LI-ION BATTERIES

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Physics-based models play an important role in simulating and optimizing the performance of Li-ion batteries. Among the physics-based models, the Doyle-Fuller-Newman (DFN) model is the most widely used [1]. However, the accuracy of this model depends heavily on its parameters. Two of the most critical parameters for the model are the solid-phase diffusion coefficient (D_s) and the kinetic reaction-rate constant (k_0) [2]. These parameters are critical because D_s controls the diffusion overpotential, while k_0 governs the charge-transfer overpotential. Together, these two overpotentials account for more than half of the total overpotential in a battery. Therefore, only precise determination of D_s and k_0 will lead to accurate battery simulations.

Since D_s and k_0 cannot be directly measured, they must be estimated, a process that can be quite complex. A literature review reveals that many studies have inaccurately determined these parameters. In this work, we have re-evaluated the estimation methods to address these inaccuracies and improve the reliability of the estimation process [3]. We use galvanostatic and potentiostatic intermittent titration techniques (GITT and PITT, respectively) to estimate D_s and k_0 using half-cells with Li(Ni_{0.4}Co_{0.6})O₂ chemistry. The two compared estimation methods are the widely used analytical approach, based on Weppner's and Huggins's work [4], and a physics-based modelling approach (DFN model), with the objective of achieving the best fit between the measured and simulated GITT or PITT pulses. The obtained D_s and k_0 parameters from both methods were implemented into the DFN model and validated using constant and dynamic current profiles. The results show that the combination of GITT measurements with the DFN model for estimating D_s and k_0 provides the highest simulation accuracy, with an average root-mean-square error (RMSE) of 12.6 mV, followed by PITT measurements combined with the DFN model, resulting in an RMSE of 15.5 mV. The determination of D_s and

 k_0 with the analytical approach demonstrates lower accuracy, particularly when combined with GITT measurements (RMSE of 53.79 mV), whereas PITT measurements show better accuracy (RMSE of 26.42 mV). Findings suggest that the commonly used analytical methods by Weppner and Huggins [4] for accurately estimating D_s and k_0 are inappropriate due to inherent limitations and assumptions. The proposed approach, combining GITT measurements with DFN model simulations, has demonstrated high accuracy and versatility in determining D_s and k_0 as a function of the lithiation degree.

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Molecular insights into an ether-functionalised ionic liquid electrolyte with hydrogen-modified anions at electrode interfaces

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The chemistry of cations and anions in an ionic liquid (IL) significantly affects its bulk phase and interface properties, requiring an in-depth understanding to advance its application in energy storage, including batteries and supercapacitors. In this work, a novel ionic liquid electrolyte featuring an ether-functionalised cation, N-methyl-N-methoxymethylpyrrolidinium [C₂₀₁mpyr], and a hydrogen-modified anion (difluoromethane-sulfonyl) (trifluoromethane sulfonyl) imide [DFTFSI] was studied computationally as a sodium battery electrolyte. The impact of the ether oxygen in the cation and hydrogen in the anion on the bulk phase structure, ion diffusion and interfacial chemistry were examined. The presence of the ether oxygen enhances ion diffusion and disrupts Na-anion aggregation through Na-O_{cation} coordination in both the bulk phase and at the electrode interface. The Na-O_{cation} coordination slightly affects the number of cations and anions in the innermost electrolyte layer but significantly influences the redox stability of the cation. The decomposition of the DFTFSI anion was explored, and two reduction routes and potential products were identified. This work provides new insights into the role of functionalised cations and anions in ionic liquid electrolytes, contributing to their development of advanced sodium batteries.



Figure 1. (a) Illustration of the IL electrolyte/electrode interface. (b) Decomposition of Na (ice blue) + DFTFSI on the Na (light green) metal surface. (c) Enhanced cation diffusion due to the interaction of ether oxygen with Na, compared to its butyl counterpart.



Unlocking the potential of a chemically modified blue phosphorene as anode materials for high-performance sodium-ion batteries

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Abstract

Blue phosphorene (Blu-Pn) has recently been identified as a new two-dimensional (2D) form of black phosphorus via the epitaxial growth method and has shown the ability to host a substantial amount of Na while serving as an anode in sodium-ion batteries (NIBs). However, its structural instability is an issue that hinders its further ex-ploitability in NIBs. In this paper, we propose to chemically modify the Blu-Pn network by substituting six phosphorus atoms with a carbon source in the form of a C6 ring, constructing a new and robust CP2 nanosheet. Based on the state-of-the-art non-local van der Waals corrected density functional theory and ab initio molecular dynamic (AIMD) simulations, we systematically explore the feasibility of the designed CP2 as a potential anode material for NIBs. Adding a C6 ring in the Blu-Pn lattice reduces its energy gap from 2 eV to 1.3 eV, which disappears after Na adsorption, indicating a conductive character. Attributed to the synergistic effect, the adsorption energy of Na on CP2 nanosheet (-2.22 eV) is greatly decreased in comparison to the pristine carbon source (e.g., graphene with -0.78 eV) and pristine Blu-Pn (-1.85 eV). The Na diffusivity on CP2 at 300 K is predicted to be $4.90 \times 10-11$ m2/s by AIMD. The specific capacity is up to 604.1 mAh/g, which exceeds that of the commercialized graphite anode (372 mA h/g). Ultimately, the designed electrode provides a small open circuit potential of 0.66 V that falls within a desirable voltage range. This work paves the way for exploring Blu- Pn 2D anode materials for NIBs technology and provides valuable insights into surface chemical modification towards the longterm stability of Blu-Pn [1] [2] [3].

Keywords: Blue phosphorene 2D material Anode, Sodium-ion batteries Density functional theory



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The Role of Electrolyte Additives in Electrochemical Performance of High-Voltage LiNi_{0.5}Mn_{1.5}O₄/Silicon-Graphite Cells

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Silicon-graphite/LiNi_{0.5}Mn_{1.5}O₄ (LNMO) is a promising cell chemistry for next-generation high-voltage lithium-ion batteries offering several advantages such as high capacity of the silicon-containing anode and a cobalt-free cathode. However, its main drawback is severe capacity fading often related to the large volume change of silicon particles in which the solid-electrolyte interphase (SEI) continuously breaks and reforms during cycling. To enable the full potential of these electrode materials, electrolyte formulations must address specific requirements such as good electrochemical stability, high conductivity, formation of stable passivation layers on both electrodes, and ability to mitigate side reactions etc.[1]

The goal of this study was to meet these requirements by introducing different electrolyte additives in a baseline carbonate electrolyte and to investigate specific roles of those additives on the cycling stability of silicon-graphite/LiNi_{0.5}Mn_{1.5}O₄ cells. Firstly, a series of cycling experiments with fresh and harvested electrodes revealed that the main degradation mechanism in these cells is lithium loss, and that the resistance increase has minimal effect on the cycling stability. Investigated additives were found to reduce lithium loss over 100 cycles and improve cycling stability. Secondly, a series of XPS studies on anode and cathode after formation and after 100 cycles showed how the formation of a stable and protective passivation layer on the cathode can help in mitigating side reactions and electrode cross-talk. Moreover, the cells with such protective layer were also shown to have a less degraded anode and hence consumed less active lithium during cycling.[2]

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Modelling Approach to Assess Electrochemical Compatibility of Electrode/Solid Polymer Electrolyte Interface in All-Solid-State Battery

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Nowadays the research for the next-generation batteries has intensified, driven by national and international policies. In this context, All-Solid-State Battery (ASSB) has gained in interest as a promising solution to make batteries safer while boosting their energy density. On top of the bulk properties of electrolyte and active materials, the stability of both cathode and anode interfaces is a key factor for long-term high performance. Electrochemical degradation of the interface can lead to the battery capacity fading and internal impedance rising as the active material is consumed and a new phase forms. Nevertheless, characterizing heterogeneous interfaces at atomistic scale presents formidable challenges, particularly through experimental methods that necessitate specialized testing facilities (in-situ and/or operando techniques). (Ab initio and reactive) Molecular Dynamics have emerged as a valuable tool to investigate buried interfaces.

Recently, Relative Bond length Change (RBC) analysis was combined with AIMD simulations to study the longterm stability and compatibility of a broad range of Solid Polymer Electrolytes (SPEs) with key negative/positive electrode materials (graphite, silicon, lithium, LFP, NMC)[1]. This approach, i.e. coupling RBC analysis and AIMD, enabled straightforward analysis of (electro)chemical degradation during dynamic calculations. The results highlighted how the functional groups interactions with electrodes affect their long-term stability and reactivity. Bond length distributions were found to respond to environmental changes and relate to the long-term reactivity. Furthermore, the balancing of the SPE polymer mobility and functional group–electrode surface attraction, respectively, kinetic and thermodynamic properties, further suggests a selective spatial orientation of functional groups when exposed to an electric field, which could have great implications for low-temperature and high-current-density environments. The obtained knowledge on how reactive key SPE polymer functional groups are and also how their reactivity changes in terms of the electric field orientation effect could provide new insights for designing new stable SPE polymers.

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Enabling Sustainable Lithium-Ion Battery Cathodes with a Water-Soluble Poly(Ionic Liquid) Binder

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Lithium-ion batteries (LIBs) face increasing concern regarding their environmental impact due to the use of toxic solvents and fluorinated polymers in their production [1]. This work addresses these challenges by exploring sustainable alternatives in both binder and electrolyte technologies. A novel approach combining aqueous processing with poly(ionic liquid) (PIL) binders and advanced electrolytes is presented to enhance battery performance while minimizing environmental footprint. Our research builds upon previous studies utilizing poly(diallyldimethylammonium) with diethyl phosphate (PDADMA-DEP) binders in NMC811 cathodes, where we demonstrated a promising alternative to PVDF/NMP systems with water-based processes, yielding capacity retention of 91% after 90 cycles [2].

Then, the application of this water-soluble PDADMA-DEP PIL was further extended to lithium manganese oxide (LMO) cathodes. This study also explores the impact of ionic liquid electrolytes, safer and non-flammable alternatives to conventional organic electrolytes, on the performance of these LMO cathodes. The combination of the PDADMA-DEP binder and ionic liquid electrolyte aims to create a more sustainable and high-performing battery system.

Electrochemical testing reveals that PDADMA-DEP binders enable high-rate capability and long-term cycling stability. As illustrated in Figure 1, LMO cathodes with PDADMA-DEP demonstrate excellent rate capability up to 10C with conventional organic electrolytes and achieve 58% capacity retention over 500 cycles at 0.5C when paired with a 50 mol% LiFSI in N-trimethyl-N-propylammonium bis(fluorosulfonyl)imide ionic liquid (IL) electrolyte. X-ray Photoelectron Spectroscopy (XPS) indicated significant interactions between the binder and LMO that enhance stability and ion conduction [3]. The development of water-soluble, PIL-based binders represents a significant advancement in sustainable battery technology, addressing environmental concerns while maintaining high electrochemical performance.



Figure 1. Electrochemical performance of LMO/lithium coin cells cycled at 25 °C using different binders (PVDF or PDADMA-DEP) and different electrolytes (LP30 or IL). (a) Rate capability at different discharge rates. (b) Long-term cycling at 0.5C.

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Harnessing AI to rapidly identify degradation mechanisms of batteries

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Diagnosing lithium-ion battery degradation is challenging due to the complex, nonlinear, and pathdependent nature of the problem. Here, we develop a generalised and rapid degradation diagnostic method with a deep learning-convolutional neural network that quantifies degradation modes of batteries aged under various conditions in 0.012 s without feature engineering. Rather than performing extensive aging experiments, synthetic aging datasets for network training are generated. This dramatically lowers training cost/time, with these datasets covering almost all the aging paths, enabling a generalised degradation diagnostic framework. We find the degradation diagnostic errors to be less than 1.22% for three leading commercial battery chemistries. Furthermore, we perform sensitivity analysis of the capacity-voltage curve under different degradation modes to provide a physically informed voltage window for diagnostics with partial charging data. By using the gradientweighted class activation mapping approach, we provide explainable insights into how these CNNs work; highlighting regions of the voltage-curve to which they are most sensitive to separate the composite electrode degradation. Quantification of degradation modes with the partial discharge/charge data using the proposed diagnostic framework validates the real-world feasibility of this approach. Robustness is validated by introducing noise to the data, with no significant negative impact on the diagnostic accuracy for noise levels below 10 mV, thus highlighting the potential for deep learning approaches in the diagnostics of lithium-ion battery performance under practical conditions. The identified mechanistic understanding of battery degradation modes will enable a unique, promising pathway to optimally extend battery lifetime while maintaining high performance.



Figure 1. Harnessing AI to rapidly quantify degradation modes and identify degradation mechanisms of batteries.

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ANALYSIS OF POLYSULFIDES IN ELECTROLYTES OF LI-S BATTERIES USING HPLC-ICP-MS METHOD

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The dissolution and diffusion of intermediate lithium polysulfide species during the operation of Li-S batteries can result in active material loss and gradual capacity decline. Characterizing these species and preventing their formation are critical for enhancing the performance of Li-S batteries. To tackle the first problem, sulfur atoms can be detected interference-free using high-resolution ICP-MS devices, such as sector-field (SF) mass spectrometers. In this study, we introduce, for the first time, a straightforward HPLC-ICP-MS method for the speciation of polysulfide anions in Li_2S_n mixtures within organic solvents [1].

The polysulfide speciation method was applied to investigate the influence of various electrolyte additives used in Li-S batteries. For instance, it was revealed that the addition of lithium 2-nitrophenolate or lithium nitrate does not affect the polysulfide distribution in the electrolyte, whereas biphenyl-4,4'-dithiol causes precipitation of (poly)sulfide species. It was shown that the main polysulfide species in fully discharged batteries is S_2^{2-} , whereas the most abundant species in fully charged batteries are S_3^{2-} and S_4^{2-} . The analysis of overall sulfur content over battery components in "anode-free" lithium-sulfide batteries, demonstrated that the electrolyte additive lithium trithiocyanurate retains the polysulfide species more effectively at the positive electrode than an interlayer of electrospun PVDF-HFP leading, nevertheless, to a faster capacity fade due to insufficient Li reactivation.



Figure 1. a) Scheme of the workflow of the analysis; b) Polysulfide distributions for different state of charge batteries with or without lithium 2-nitrophenolate additive.

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Enhancing the Electrochemical Performance of Li-ion Battery Electrodes by Atomic Layer Deposition

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Rechargeable Li-ion batteries dominate the market for high energy storage applications. Despite their widespread use, Li-ion batteries face challenges because of degradation in electrochemical performance due to uncontrolled reactions between the electrodes and the electrolyte. To address these challenges, strategies like protective surface coatings have been explored. Coatings applied via atomic layer deposition (ALD) reduce direct electrode-electrolyte interactions, suppress side reactions, prevent electrode material dissolution, stabilize structural integrity, improve Li-ion transport, and facilitate the formation of a stable Solid Electrolyte Interphase (SEI) layer.

In our study, we investigate the effects of ALD coatings on silicon/graphite-based anodes and lithium iron phosphate (LiFePO4) cathodes. The Si/Gr electrodes were prepared by tape casting a slurry containing Si/Gr powder, sulfonated-alginate binder, and carbon black as a conductive additive on copper foil. The fabricated silicon/graphite anodes were coated with ultrathin ZnO films using 5-40 ALD cycles at a deposition temperature of 100 °C. Transmission electron microscopy showed complete coverage of the silicon and graphite particles with the ZnO ultrathin layer. Electrochemical measurements revealed that the Si/Gr electrode coated with 20 ALD cycles of ZnO had the optimal ZnO thickness, exhibiting the best performance among the samples analyzed. The ZnO surface coating effectively altered the SEI layer, reducing both SEI and charge transfer resistances, which enhanced Li-ion transport. As a result, the rate capability of the ZnO-coated Si/Gr anodes improved significantly. Similarly, LiFePO₄ cathodes were modified through atomic layer deposition with 2 to 20 ALD cycles of AI_2O_3 at 100 °C. The LiFePO4 cathodes coated with 5–10 ALD cycles of Al₂O₃ showed significantly enhanced capacity at high charge/discharge rates in full-cell batteries with a graphite anode. The alumina layers improved electrochemical performance by serving as a protective barrier, reducing direct contact between the LiFePO4 surface and the electrolyte, and minimizing structural degradation in the cathode. Consequently, alumina-coated electrodes demonstrated notable improvements in rate capability, particularly at higher C-rates of 1 and 2, compared to the pristine LiFePO₄ samples.

These results emphasize the potential of ALD-based surface engineering as a promising strategy for enhancing the electrochemical performance of lithium-ion battery electrodes.

We acknowledge the support from VEGA 2/0162/22, APVV–19–0461, APVV–20–0111, EU-funded FULL-MAP, and M-ERA.NET SusHiBatt projects.



Figure 1. The electrochemical performance of Si/Gr and LiFePO₄ electrodes with atomic layer deposited ultrathin films.



Combined Capacitive and Electrochemical Charge Storage Mechanism in High Performance Graphene-Based Lithium-Ion Batteries

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Chemically produced graphene has already been successfully employed in batteries, to increase specific capacity and maximum rate of charge and discharge, and in supercapacitors, thanks to its electrical conductivity and its specific surface area. Many different synthesis routes have been proposed for graphene, each leading to different properties, useful for different applications. In this study, graphene was synthesized by thermal exfoliation of graphite oxide (TEGO), while hydrogenated TEGO (H-TEGO) was obtained by heating TEGO under hydrogen flux. Lithium half-cells were assembled either to characterize TEGO and H-TEGO or to achieve preformation of the SEI on the graphene-based electrodes for their use in full-cells.

Hydrogenated graphene boasts an impressive reversible specific capacity with fast charge/ discharge capabilities, exceeding 370 mA h g⁻¹ even at 25 C-rate. Diffusion mechanism of lithium in TEGO was characterized at different states of intercalation by means of electrochemical impedance spectroscopy. In addition, a novel combined electrostatic and electrochemical charge storage mechanism of lithium ions in graphene-based electrodes is proposed, based on three-electrode cyclic voltammetry investigation. Thanks to their dual nature, TEGO and H-TEGO can be employed both as battery and supercapacitor electrodes, making them ideal candidates in high-performance lithium-ion capacitor electrodes. Furthermore, TEGO and H-TEGO anodes were paired with commercial cathode materials, to study the feasibility of their application in full-cells. [1].



Figure 1: a) Voltage profiles for H-TEGO at different C-rates; b) proposed mechanism of Li+ diffusion in TEGO and H-TEGO.

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Graduate School Electrochemical Energy Storage

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The Graduate School Electrochemical Energy Storage (GS-EES) supports about 90 doctoral researchers at Karlsruhe Institute of Technology (KIT), Ulm University and Justus-Liebig-University Giessen and is the largest graduate school in Germany in the field of electrochemical energy storage.

The GS-EES addresses the full interdisciplinary spectrum of electrochemical energy storage and conversion, from fundamental science to processing and application. Among our topics are Lithium-ion batteries, post Lithium batteries and fuel cells.

Our goals are :

- Educate and train the next generation of researchers and engineers in the field of electrochemical energy storage.
- **Support** them in all aspects regarding research.
- **Empower** them to foster their creativity and independence as researchers, to build scientific networks within and beyond the GS-EES, to learn develop their personal skills, to raise awareness for rights and obligations as researchers in modern society.
- **Prepare** them for successful careers in both industry or academia.



Figure 1. Main components of the GS-EES qualification program.

Highlights of the GS-EES training and support measures are:

- Three-month research stay abroad
- Broad portfolio of workshops on scientific or method-related topics and on professional skills
- Two weeks of interdisciplinary Block Course "Materials, Functioning and Technology of Batteries"
- Annual Summer School

Both the **Block Course and the Summer School are open** to open to all interested PhD or master students and post-docs, no matter at which institution they are enrolled.

The GS-EES participates in the work package "European curricula in future battery technologies" of the coordinating support action of Battery 2030+.


SOLVE – Enabling room-temperature Solid-State Batteries through European innovation and collaboration

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Solid-state batteries (SSBs) are one of the main candidates for the next generation of energy storage, offering unparalleled safety, and potential higher energy density, faster charging compared to conventional lithium-ion batteries and durability. These advantages position SSBs as critical enablers of Europe's green transition, particularly for electromobility. However, challenges such as high internal resistance, interfacial instability, scalability, and cost-effective manufacturing hinder widespread adoption.

The EU-funded SOLVE^{1,2} project unites 16 European partners to overcome these barriers and position the EU as a global SSB leader. By integrating cutting-edge innovations across materials, interfaces, and manufacturing, SOLVE targets the development of **room-temperature**, **safe-by-design 20** Ah Gen4b SSB **prototypes** and a **proof-of-concept 0.25 kWh module**, designed for industrial scalability. Key breakthroughs will include:

- **Hybrid Solid Polymer Electrolytes (HSPE):** Ultrathin (<30 μm) membranes containing ceramic fillers with high ionic conductivity (>0.5 mS/cm at 25°C) and wide electrochemical stability (>4.7 V).
- **High-Loading Cathodes:** Ni-rich NMC-based solid state electrodes (>4.0 mAh/cm², >200 mAh/g) with engineered interfaces to minimize degradation and optimized catholyte.
- Ultra-Thin Li Metal Anodes: Including lithophilic current collectors for zero Li excess SSB (<10 μm).

SOLVE integrates digital tools and models to accelerate SSB design and incorporates sustainability criteria through eco-design principles and innovative recycling processes. The project emphasizes **sustainability at every stage**, from solvent-free electrolyte production to end-of-life circularity.



Figure 1. SOLVE's LiM-SSB cell concept.

To accelerate market readiness, SOLVE fosters collaboration across academia, RTOs and industry, supported by targeted training programs and exploitation strategies. By bridging the gap between lab-scale innovation and industrial deployment, SOLVE aims to decarbonize transport, strengthen Europe's battery value chain, and deliver SSBs that outperform current technologies in energy density and safety.

Acknowledgements: the SOLVE project is funded by the European Union under Grant Agreement No. 101147094, administered by the European Climate, Infrastructure and Environment Executive Agency (CINEA).

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Understanding Battery Chemistry with Magnetic Resonance: A Path to Next-Generation Energy Storage

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Batteries play a crucial role in the energy transition. However, to develop batteries with enhanced stability, increased efficiency and higher safety understanding of material structure-property relations is essential.

Magnetic Resonance techniques, particularly Solid-State Nuclear Magnetic Resonance (NMR) and Electron Paramagnetic Resonance (EPR), provide unique insights into local structure and dynamic of energy storage materials. Even minor changes in material composition of electrolytes and/or electrodes can have remarkable impact on their local coordination environments and subsequently on their properties.

The presentation will cover applications and challenges of solid-state NMR and EPR in battery technology highlighting their potential in academic and industrial research and development. By combining operando/insitu and ex-situ Magnetic Resonance studies on battery materials we can reveal formation of solid-electrolyte interphase, monitor dendrite process, investigate ion-exchange between solid-solid interfaces and/or understand storage behavior of electrodes. Revealing these reaction pathways at atomic level contributes to the development of safer, more efficient, and longer-lasting batteries.



Stable Electrochemical Performance of Recovered Graphite for Reuse in Lithium-Ion Batteries

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Lithium-ion batteries (LIBs) were first introduced to the market by Sony Corporation in 1991 and have since become essential in various applications, including consumer electronics, electric vehicles, military technology, and medical devices. Their popularity is attributed to their high energy density, long cycle life, and low self-discharge rate [1,2]. As the demand for LIBs continues to rise, the need for efficient recycling strategies has gained significant attention. While extensive research has focused on recovering transition metals from spent cathodes, the recycling of graphite anodes has been largely overlooked [3]. However, cost-effective regeneration of graphite is crucial for reducing environmental impact and ensuring sustainable resource utilization. In the context of the RENOVATE project, our approach involves froth flotation, followed by an attrition stage, to improve graphite recovery with a 90% recovery rate. A subsequent post-leaching treatment then further boosts purity to 99.9%. This approach improves both the purity and structural integrity of recovered graphite, enhancing thus its electrochemical performance. The spent graphite initially delivers a specific capacity of 301.1 mAh/g in the first cycle at 0.5 C, which decrease to 79.5 mAh/g after 200 cycles. In contrast, the regenerated graphite demonstrates a higher initial capacity of 279 mAh/g and retains stable at the same value after 200 cycles at 0.5 C, highlighting its improved cycling stability.



Fig. 1. (A,B) Charge–discharge curves and coulombic efficiency (CE) of FAAM and Indian graphite at 0.5 C. **(C,D)** Cyclic voltammetry (CV) curves of Li⁺/Li at 0.1 mV s⁻¹ from 1 to 3 mV of FAAM and Indian graphite.

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OStructural and Electrochemical Insights into Novel Doped Niobates

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Niobium oxide anodes are emerging as a strong alternative for high-power lithium-ion battery (LIB) applications, addressing the limitations of conventional anode materials. While graphite anodes, with a theoretical capacity of 372 mA h g⁻¹, are widely used, their performance is hindered at high charge rates due to lithium plating and dendrite formation, raising safety concerns. ¹ Lithium titanate (LTO) offers improved safety with a working potential of 1.5 V vs Li/Li⁺ but is limited by its lower capacity of 175 mA h g⁻¹. ² Developing new anode materials combining high energy density with enhanced safety at higher rates could significantly improve on LTOs' and graphite's limitations. Such properties would benefit mining, maritime, aviation and rail transport applications, industries, which require decarbonisation strategies to combat the risks posed by rising CO₂ emissions. In this respect, niobium oxides provide a compelling alternative, delivering both high capacity and rapid lithium-ion diffusion. Their Wadsley-Roth structures are particularly promising, featuring oxygen-deficient ReO₃-like frameworks with no A-site cation occupancy. These structures consist of distorted MO₆ octahedral blocks connected by shear planes that compensate for oxygen deficiencies, enhancing structural rigidity and limiting volumetric expansion. ^{3,4} The presence of these shear planes also facilitates fast lithium-ion diffusion, making niobium oxides highly suitable for high-rate battery applications. ⁵

This study has investigated modifications of a new 4 x 4 block Wadsley-Roth phase, Nb₉Ti_{1.5}W_{1.5}O₃₀ which has potential for high-power battery applications. ^{6,7} We illustrate the electrochemical performance of this phase, showing excellent high-rate capability, and report a study investigating the solid-solution range for this system. These findings highlight the potential of new niobate Wadsley-Roth phases as promising high-power anode materials, offering a balance of stability, capacity, and fast lithium-ion diffusion for next-generation lithium-ion batteries.



The crystal structure of Nb₂O₅ polymorph, N-Nb₂O₅ (4×4 block)⁵

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Development of an automated cell disassembly concept

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The increasing demand for sustainable battery recycling solutions necessitates efficient disassembly techniques for lithium iron phosphate (LFP) batteries. This work focuses on the development of an automated cell disassembly concept to optimize material recovery from LFP cells. The project commences with manual deconstruction to identify essential process steps and precision robotic tools for effective separation. Key technologies include advanced robotic arms with cutting grippers and pick-and-place mechanisms, which are crucial for handling delicate components in confined environments.

The automated process is further enhanced by the integration of computer vision technology, enabling real-time monitoring and analysis. This ensures accurate alignment and positioning of robotic tools while enhancing component identification and overall separation efficiency. The synergy between robotics and computer vision is vital in the limited space of a glovebox, where variability in geometries of end-of-life battery cells can result from manufacturing differences and aging. The robotic system must adapt to these geometric variations in real time, ensuring reliable disassembly. Initial phases will involve the atmospheric disassembly of dry reject cells using a lab-scale, semi-automatic line for effective separation of anode and cathode foils. The confined glovebox requires careful handling to prevent damage and ensure safety, and robotic systems will be programmed to perform precise manoeuvres, minimizing contamination risk and optimizing workspace utilization. By the conclusion of the construction phase, the disassembly line is expected to achieve semi-automation for wet reject cells, reliant on manual loading. The anticipated outcome is the production of separate, dry, and salt-free anodes and cathodes, safely released for further processing. Additionally, fractions such as casings, busbars, internal fixtures, and separator foils will be effectively managed in subsequent tasks.



Tailored electrode architectures for enhanced specific energy and cycling stability in high areal capacity sodium-zinc chloride batteries

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Commercial sodium-nickel chloride (Na-NiCl₂) batteries provide reliable energy storage for industrial applications. Replacing nickel, an increasingly critical raw material, by cheap and abundant zinc, may provide an economically viable and environmentally sustainable sodium-zinc chloride (Na-ZnCl₂) battery.¹ However, the lower melting point of metallic zinc and of zinc chlorides, together with a lower cell voltage, adversely impacts cycle life and specific energy of Na-ZnCl₂ cells.²

In this study,³ we discuss how the positive electrode architecture influences the cycling performance and ageing mechanisms in Na-ZnCl₂ cells operated at 300 °C. We present cycling results for two different architectures, namely particle-based Zn/NaCl granules and foam-based Zn/NaCl electrodes.

Particle-based electrodes, cycled in tubular cells with 150 mAh cm⁻² areal capacity, achieve a specific energy of up to 231 Wh kg⁻¹ at 15 mA cm⁻² but suffer from degradation in voltage efficiency, due to zinc agglomeration. To address this challenge, foam-based Zn/NaCl electrodes with over 200 mAh cm⁻² areal capacity were developed. Cycled in planar Na-ZnCl₂ cells, these electrodes achieve a specific energy of 336 Wh kg⁻¹ at 15 mA cm⁻², with stable voltage profiles.



Figure 1. Schematic of **a**) particle-based Zn/NaCl granules and **b**) foam-based Zn/NaCl electrodes applied at the positive electrode in Na-ZnCl₂ cells.

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Towards an autonomous robotic battery materials research platform powered by automated workflow and ontologized FAIR data management

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The discovery of novel battery materials has been accelerated by machine learning and advanced modeling. However, their integration into battery cells remains constrained by the necessity for experimental validation. Here, we present the status of development and validation of our automated robotic battery materials research platform Aurora enabling rapid testing of scientific hypotheses and validation of physical models. Aurora integrates electrolyte formulation, battery cell assembly, and battery cell cycling into an automated application-relevant closed-loop workflow. We illustrate how the different features of the Aurora platform can be leveraged to design experiments elucidating the impact of cycling parameters, electrode composition and balancing, and electrolyte formulation on battery performance and long-term cycling stability at the example of NMC||graphite and LFP||graphite cells with carbonate-based electrolytes, which serve as a benchmark battery cell chemistries. We conclude with an outlook on how we plan to integrate Aurora into autonomous data-driven workflows. Our study establishes Aurora as a powerful research platform for accelerating battery materials research.



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Architectured Synthesis of a NMC core/shell cathode: Tailoring microstructure with tungsten oxide barrier for improved stability of Lithium- ion batteries

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Lithium-ion batteries play a crucial role in modern energy storage applications, driven by the demand for high-performance and widely applicable energy storage solutions. High Ni-content layered oxide cathodes, such as LiNixMnyCozO2 (NMC) applied in Lithium-ion batteries (LIBs), are at the forefront of next-generation cathode materials due to their high energy density and cost-effectiveness. However, their commercialization is hindered by several challenges, including irreversible capacity loss during the initial cycle, voltage hysteresis, poor rate capability, and gradual voltage decay. These issues primarily stem from the high Ni-content, which promotes Ni²⁺ migration into vacant lithium sites via tetrahedral interstices after Li⁺ extraction. This migration is accompanied by oxygen lattice loss, leading to structural instability and performance degradation [1]. To address these challenges, we propose a core/shell NMC architecture, where a high-Ni NMC90 core ensures high capacity, while a Mn-rich NMC622 shell enhances structural and thermal stability [2]. Furthermore, introduction of a tungsten oxide (WO₃) interlayer between the core and shell may offer additional benefits to mitigate interfacial degradation and improve long-term cycling stability. This WO₃ layer acting as a diffusion barrier may prevent undesirable elemental migration and preserve the structural and electrochemical integrity of the cathode material. The synthesis of this multi-layered core/shell NMC particles has been performed using a coprecipitation method, employing oxalic acid as a cost-effective and environmentally friendly precipitant. Additionally, lithium incorporation has been carried out directly during the co-precipitation step to ensure a homogeneous precursor composition. Structural and electrochemical characterizations have been conducted using SEM, EDX, XRD and GCD analysis. The XRD results in Figure 1(a) confirm that both NMC90 and NMC622 achieve the desired $R\overline{3}m$ crystal structure on calcination at 850°C. Furthermore, GCD measurements, shown in Figure 1(b), reveal that the WO₃-modified core/shell cathode exhibits a degradation rate that is reduced by a factor of over two compared to the uncoated counterpart during cycling. This combined strategy of integrating a core/shell architecture with a WO₃ diffusion barrier, demonstrates a promising approach for enhancing the performance, stability, and lifespan of high-Ni NMC cathodes in next-generation LIBs.



Figure 1: XRD of NMC90 (core), NMC622 (shell) and core/shell structured NMC cathode material and (b) cycling performance of core/shell structured NMC with and without WO₃ Interlayer

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Accelerating battery innovation through operando gas analysis

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The comprehensive understanding of interface formation and evolution remains one of the most important challenges for rapid battery innovation, as these interfaces play a pivotal role on both performance and safety[1]. The gas evolution signature of interfacial processes can be highly informative, and operando gas analysis techniques have become an essential tool for accelerating battery innovation[2-6]. Electrochemical mass spectrometry (EMS) can provide quantitative information of individual processes occurring in the cell, as well as their interconnectivity (e.g., cathode-anode crosstalk and anode slippage) with high sensitivity and resolution, as it can be seen in the figure[1].

Here we present online electrochemical mass spectrometry (OEMS) studies of systems with both highnickel cathode materials and high-voltage anode materials that show the nature, onset, extent and interconnectivity of parasitic reactions leading to cell aging using very short test protocols. Our results indicate the need for better understanding these processes in order to accurately target solutions such as electrolyte formulations, coatings, etc., to accelerate materials innovation in batteries.



Figure 1. Operando gas analysis data (OEMS) showing voltage profile (top) alongside typical gas evolution profiles (bottom; m/z = 2 for H_2 ; m/z = 27 for C_2H_4 ; m/z = 28 for CO; m/z = 44 for CO₂) of electrochemical processes contributing to the formation/evolution of interfaces in lithium-ion batteries.

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The EU Battery's Regulation: Reshaping Business Models, Overcoming Barriers and Bridging the Gap in the transition to a Circular Electric Vehicle Battery Value Chain?

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The EU Battery Regulation that came into force in July 2023 represents a pivotal legislative framework designed to accelerate the transition towards a circular electric vehicle battery (EVB) value chain.¹ This article critically examines the Regulation's capacity to reconcile ambitious sustainability mandates with pragmatic business realities, identifying systemic barriers, such as fragmented supply chains, technological limitations, and economic and financial inertia that hinder the achievement of circularity.

This article will examine the barriers that the stakeholders should overcome and focuses on the key actors within the electric vehicle (EV) battery value chain, with particular emphasis on the role of policymakers and regulatory frameworks. Policymakers and the policies they enact serve as a critical regulatory bridge, facilitating the overcoming of systemic barriers within the EV battery ecosystem. By establishing stringent thresholds and standards, policymakers can compel various stakeholders ranging from raw material miners and battery producers to electric vehicle manufacturers, recyclers, and other participants in the value chain to adopt practices that ensure compliance and handling with end-of-life (EoL) for EV batteries. The Regulation introduces several transformative provisions, which in a way compel businesses to rethink, reshape and transform their models and integrate sustainability into their core strategies. These include recycled content requirements, battery passport, due diligence requirement and end of life management. These provisions can be seen as codifying core circular economy principles into law, transforming abstract sustainability goals into actionable obligations. By legally mandating circular practices, the Regulation disrupts traditional take-make-waste models and sets the stage for a systemic shift toward sustainability.

While circular business models are essential, they operate within the constraints of market incentives, technological feasibility, and economic structures. This article argues that without a robust regulatory framework, business-led initiatives remain fragmented, voluntary, and vulnerable to market uncertainties. Hence, this article further argues that the Battery Regulation is the true enabler of systemic change.

Policymakers, through legislation and governance, serve as the architects of circularity, providing the legal certainty and economic incentives necessary for businesses to invest in sustainable practices. By mandating extended producer responsibility (EPR), enforcing material recovery targets, and establishing closed-loop supply chain requirements, the EU Battery Regulation creates the structural foundation for a truly circular EV battery value chain. While the Regulation establishes rigorous obligations that unlock opportunities for innovation in recycling, remanufacturing, and resource efficiency, it also reveals tensions between its aspirational objectives and operational hurdles, which may undermine its transformative potential. The Regulation provides the necessary framework, but its transformative potential can only be realized through collaboration, innovation of various stakeholders in the EV battery value chain, and a shared commitment to sustainability.

¹ • REGULATION (EU) 2023/1542 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 12 July 2023 concerning batteries and waste batteries, amending Directive 2008/98/EC and Regulation (EU) 2019/1020 and repealing Directive 2006/66/EC L 191/1(Text with EEA relevance) ('Battery Regulation').



Smart sensors and self-healing functionalities embedded for battery longevity with manufacturability and economic recyclability (SALAMANDER)

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The European Commission has a goal of transitioning towards a carbon neutral society and reducing greenhouse gas emissions by at least 55% by 2030. It is therefore crucial to develop batteries with ultra-high performance beyond current capabilities to fulfill these goals and keep up with the increasing demand for batteries. [1] The main objective of the SALAMANDER project is to demonstrate a longer lasting battery cell by integrating self-healing functionalities embedded directly into the electrode which is triggered by external stimuli on internal sensors measuring temperature, electrode electrical conductivity and Mn-ion dissolution which communicates with a battery management system (BMS) without detrimentally affecting manufacturability and recycling, as shown in Figure 1. To achieve this, the project aims to demonstrate a successful embedment of sensors and self-healing functionalities into single battery cell, with sensors being capable to detect defective operation and trigger self-repairing functionalities via BMS. In the SALAMANDER project, we developed such a solution for Sibased and other high-capacity anodes materials, integrating computationally and experimentally optimized self-healing polymers to counteract the mechanical fracturing due to repeated volume change, and for NMC-based cathodes vulnerable to transition metal dissolution which results in solid-electrolyte interphase (SEI) degradation and loss of lithium inventory. The targeted self-healing functionalities aims to enhance the quality, reliability and lifetime of Li-ion batteries. [2]



Figure 1. The core concept of the SALAMANDER battery in which the BMS receives input from multiple embedded sensors and triggers external heating stimulus to induce self-healing functionalities.

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CL-SP-ICP-OES Analysis of Cathode Active Materials

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In recent years layered lithium transition metal oxides as cathode active material (CAM), especially $LiNi_xMn_yCo_zO_2$ ($N_xM_yC_z$; x + y + z = 1), excel due to their high theoretical specific capacity (~275 mAh g⁻¹) and further drift towards Ni-rich compositions, *e.g.* NMC811. The elevated Ni content increases the capacity and therefore the energy density of the cell at the cost of shorter cycle life. [1]

Furthermore, the influence of the particle morphology on the cycling performance, as well as the degree of inhomogeneity in the active lithium content in the respective CAM after long-term cycling were investigated in this work. As particles are separated from their electronic surroundings, the active lithium content within these structures can be isolated for further electrochemical processes, resulting in individual particles, which contain a diverging degree of lithiation (DOL) compared to those in the main electronic cathode network. [2] Thereby, the formation of an inhomogeneity in the State-of-Charge (SOC) distribution within the electrode possibly takes place.

A Classification-Single Particle-ICP-OES (CL-SP-ICP-OES) method was utilized after long-term cycling to get an insight into the degree of SOC heterogeneity of the respective CAM. In addition Scanning Electron Microscopy (SEM) was used for visualization of particle degradation (Particle cracking, surface degradation, etc.). Alternatively, the method is shown to be applicable to investigate a mixture of different NMC compositions in an artificial black mass matrix, utilizing the different transition metal ratios in the respective materials.



Figure 1. Histograms of the Mn to Ni signal intensity ratios of the four different NMC compositions. A Gaussian fit is implemented to obtain the mean of the respective signal distribution.

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Pioneering Lithium-Sulfur Battery Innovation: Theion GmbH's Breakthroughs in High-Density Energy Storage

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Theon GmbH is at the forefront of transforming energy storage through its cutting-edge lithium-sulfur (Li-S) battery technology. This presentation delves into the scientific and engineering innovations driving Theion's sulfur crystal battery, which leverages direct crystal implantation to target energy densities of up to 1,000 Wh/kg—far surpassing conventional lithium-ion systems. We will explore how Theion harnesses sulfur, an abundant and eco-friendly element, integrating it into battery cells with advanced coatings to overcome persistent challenges such as the polysulfide shuttle effect and volumetric expansion during cycling.

A key focus will be Theion's development of a lithium metal host anode, engineered to suppress dendrite growth even at high charging rates, enhancing both safety and cycle life. These advancements position Theion's Li-S batteries as a game-changing solution for demanding applications, including electric vehicles, aerospace, and grid-scale renewable energy storage.

Beyond performance, Theon's technology prioritizes sustainability by eliminating toxic and scarce materials like cobalt and nickel, while ensuring full recyclability of the battery system. This presentation will offer a comprehensive analysis of the materials science, electrochemical principles, and scalable design strategies behind Theon's Li-S batteries, underscoring their transformative potential for the future of energy storage.



Tailor-made functional material system for in-situ gas sensing in Li-S batteries

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Lithium-sulfur (Li-S) chemistries are among the most promising candidates for next generation electrochemical energy storage owing to their potentially high energy density, low raw material cost, and high capacity [1]. However, numerous scientific challenges currently hinder the realization of this potential, including poor cycling stability and electrolyte consumption. Several mechanisms can lead to the failure of Li-S batteries, such as overcharging. This leads to high temperatures and localized short circuits, which, in turn, can trigger the decomposition of the electrolyte. Ultimately, these processes result in a decrease in the pH of the electrolyte system, leading to reactions with the polysulfides or sulfides in the cathode, resulting in the generation of hydrogen sulfide (H_2S).

To detect this failure mechanism and subsequently initiate self-healing processes, the specific detection of H_2S at room temperature is required. At the same time, the gas sensing technology has to be compatible with the HealingBat technology platform, which relies on planar microsystems technology. Therefore, a functional layer based on a copper(I)oxide (Cu₂O) functional ink, synthesized via a precipitation reaction [2], has been developed. This approach allows for the production of spherical Cu₂O nanoparticles with a narrow size distribution, which are dispersed in a water-based solution serving as ink. Upon thermal treatment, the solvent completely evaporates and Cu₂O is converted to CuO while maintaining the particle morphology [3]. Figure 1 shows the functional layer deposited onto an interdigitated electrode structure to enable the determination of the layer's resistivity. Using this material system, the detection of H₂S developing inside the battery is based on a H₂S-specific percolation phase transition [4], which leads to a change in resistivity by at least one order of magnitude.



Figure 1. Scanning electron microscopy image of a Cu_2O layer deposited onto interdigitated electrode structures to read-out the layer's resistivity. The use of inkjet printing allows for adjusting the layer morphology and thickness.

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ENHANCING LITHIUM-ION-BATTERY PERFORMANCE: METAL-ORGANIC-FRAMEWORKS INTEGRATED SEPARATORS TO IMPROVE THE CYCLE LIFE

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Lithium-Ion Batteries (LIBs) are the most widely used energy storage systems in modern technology, from portable electronics to electric vehicles [1]. Despite their advantages, LIBs face challenges such as capacity loss, safety concerns, and gas evolution during operation, which can degrade battery performance and longevity [2]. The growing demand for efficient and sustainable batteries has led to extensive research to improve battery components, especially separators and electrode materials. One major issue in LIBs is the formation of gases like CO₂, H₂, and O₂ due to electrolyte decomposition and instability of Ni-rich cathodes at high states of charge [3]. These gases can lead to volume expansion, electrode degradation, and safety risks. Metal-organic frameworks (MOFs) have emerged as promising materials to mitigate these challenges due to their highly porous structure and excellent gas adsorption capabilities [4]. By incorporating MOFs into battery separators, the produced gases can be captured, thereby improving cycle life and stability. This work focuses on the development of several MOF-integrated separators to enhance LIB performance. MOFs, particularly zirconium-based MOFs (Zr-MOF-808) and nickel-based MOFs (Ni-MOF-74), have been synthesized using a scalable solvothermal method and characterized using XRD, SEM, and EDX techniques. Afterwards, these MOFs have been incorporated into polyvinylidene fluoride-co-hexafluoropropylene (PVDF-HFP) thin film solid polymer separators (Figure 1a), glass fiber and Celgard 3501 separators to enhance electrolyte uptake, ion transport, and enable gas absorption. To investigate the electrochemical performance, half-cell Li-ion batteries have been fabricated and tested using commercial Lithium Nickel Manganese Cobalt Oxide (LiNi0.6Mn0.2C00.2O2, NMC622) as cathode, Li-anode, 1 M LiPF6 in EC/DMC (50/50) (v/v) electrolyte and fabricated MOF-integrated separators. The integration of MOFs into separators provides improved electrochemical stability (Figure 1b) by capturing gases generated during operation and thereby increasing the cycle life and efficiency of LIBs. This research, supported by the EU-funded PHOENIX project (grant agreement No. 101103702), represents an important step towards more sustainable and safer lithium-ion batteries for future applications.



Figure 1 (a) Ni-MOF-74 integrated PVDF-HFP solid polymer separator and (b) long cycle testing of LIBs with and without MOF-integrated PVDF-HFP solid polymer-separators.

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Visualizing the Dynamic Wetting and Redistribution of Electrolyte in Lean-Electrolyte Li-S Pouch Cells via Operando Neutron Imaging

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The lean electrolyte in lithium-sulfur (Li-S) batteries commonly presents inhomogeneous distribution and inadequate electrolyte wetting, resulting in uneven electrochemical reaction interface, suboptimal performance, and cell failure or accelerated degradation.[1] Non-destructive operando/in-situ methods that can visualize electrolyte wetting and dynamics during cell operation in liquid metal-sulfur batteries have unfortunately not been described. [2] In this study, operando neutron tomography technique is employed to nondestructively visualize and analyze the electrolyte distribution in practical lean-electrolyte double-layer Li-S pouch cells. Through real-time electrolyte observation across different pouch-cell layers, we unambiguously reveal conglomerations and diminishment of unwetted areas during cell rest, lead to localized electrolyte redistribution. Remarkably, discharge/charge processes can enhance electrolyte homogeneity remarkably, boosting the electrochemical activation of sulfur. Unique periodic "breath-in" and "breath-out" behaviors of electrolyte wetting were observed in the dynamic evolution with the status of discharge and charge, which is correlated to the dissolution and precipitation of sulfur species. These results provide solid evidence of an inhomogeneous distribution of lean electrolyte in practical Li-S pouch cells and offer insights into the correlation between electrolyte redistribution and lithium-sulfur (electro-)chemistry. This work develops valuable guidelines for optimizing testing protocols and strategies for electrolyte wetting in Li-S pouch cells and other metal-sulfur batteries.[3]



Figure 1. Illustration of the neutron imaging of the electrolyte and its dynamic wetting and redistribution

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ENHANCING LITHIUM ANODE STABILITY IN LI-S BATTERIES VIA SELF-HEALING CHEMISTRY

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Lithium-sulfur batteries (LSBs) have emerged as a highly promising next-generation energy storage technology, offering superior theoretical capacity, cost-effectiveness, and environmental benefits compared to conventional lithium-ion batteries (LIBs). However, the uncontrolled growth of lithium dendrites significantly hinders their practical application by leading to internal short circuits and severe safety hazards during repeated charge-discharge cycles. In this work, inspired by nature, we introduce a novel self-repairing strategy for lithium metal anodes to enhance the long-term stability of LSBs. By incorporating a self-healing additive into the electrolyte, we demonstrate that the cycling lifespan of lithium anodes can be significantly extended. This additive actively interacts with the predominant component of the corrosive surface layer on lithium metal anodes, thereby greatly improving lithium utilization. Our approach offers a simple, cost-effective, and highly efficient solution to the persistent issue of lithium metal corrosion in Li-S batteries. Moreover, this strategy can be readily adapted to other metal-based battery systems, including sodium-oxygen/sulfur (Na-O₂/S) and lithium-oxygen (Li-O₂) batteries, paving the way for broader advancements in next-generation energy storage technologies.

ACKNOWLEDGMENTS

We acknowledge financial support from the European Horizon project (Grant agreement No. 101104006 – HEALING BAT – HORIZON-CL5-2022-D2-01).

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RECYCLING OF LITHIUM-ION BATTERIES WITH THE FOCUS ON EXTRACTION OF GRAPHITE CONCENTRATE AND ITS FURTHER PROCESSING FOR THE SECOND LIFE USE AS BATTERY ANODES

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This TRL6 to TRL7 work is devoted to the extraction of valuable materials (cobalt, nickel, manganese, lithium, graphite, copper, aluminum), from spent lithium-ion batteries, recovered from consumer electronics, BESS working in green utilities (solar and wind) as well as from electric and hybrid vehicles. UNE reports on the engineering data package which it has developed to support a fully functional production line which was installed and rendered operational at pilot scale at AETC. This line focuses on extraction and "healing" of graphite anodes, which pay for the recycling process irrespective of whether the cathode has NMC, LNMO or an LFP chemistry.

The processing line includes: (1) Battery Direct Recycling line, including cell discharge, cryogenic deactivation and precise mechanical disassembly using simple semi-continuous and continuous machine shop equipment which allows to separate streams of anodes, cathodes and separators. The anode foils are cleaned from graphite, the cathode powders are separated from the aluminum current collectors; spent separators are upcycled by heat treatment to obtain graphite. Water after vacuum filtration of the anode and cathode stations is reused in the next cycle. The final products of the line: powdered graphite, clean and dry metal foils and cathodes; (2) Dry graphite is then repaired by heat treatment at 2200-2300°C crucibles of an induction furnace, for which large diameter graphite crucibles are machined on a precision CNC milling mill. The high-temperature induction furnace operates in a protective atmosphere (N₂) for removing metal oxides from powdered graphite. (3) Dry cathode active material is supplied to ISPE which uses plating baths to generate controlled composition metal alloy on cathodes and MnO2 on anodes. Metal alloy is then converted into new NMC by salt precipitation. A high-temperature roller hearth furnace of UNE design is then employed for lithiation of recycled cathode.

Work by UNE and ISPE was funded by the Horizon Europe Programme under Agreement No. 101137771, Action entitled "Sustainable Technologies for Reducing Europe's battery raw MaterialS dependence", or "STREAMS".



Figure 1. Schema of a fully functional pilot line designed with the assistance from UNE and installed and started up at AETC for extraction of graphite anode concentrate from spent lithium-ion batteries.

1 - Table for raw material; 2 - Discharge Station; 3 - Freezer Tenney/TJR; 4 - Lathe Hardinge / DV-59; 5 - Surface Grinder Boyar-Schultz / 6 x 12; 6 - Device used to cut open electrode; 7 - Winding Machine Winding Machine; 8 - Vacuum Pump Agilent / DS 202; 9 - Calciner; 10 -Ultrasonic Cleaner VEVOR / E-100; 11 – Conveyor; 12 - Furnace Thermcraft / XSB-12-12-1V-F01-P; 13 – Furnace Binder / ED 115-UL



SOLVOMETALLURGICAL APPROACH FOR THE RECOVERY OF VALUABLE METALS FROM EOL BATTERIES

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On the website of the European Commission is reported: "Raw materials are crucial to Europe's economy. They form a strong industrial base, producing a broad range of goods and applications used in everyday life and modern technologies. Reliable and unhindered access to certain raw materials is a growing concern within the EU and across the globe. To address this challenge, the European Commission has created a list of critical raw materials (CRMs) for the EU, which is subject to a regular review and update. CRMs combine raw materials of high importance to the EU economy and of high risk associated with their supply [1]." In the list of the critical raw materials are included Cobalt, Nickel and Lithium. These metals are essential components of the Lithium-ion batteries (LIBs). This family of batteries are spread world wide, for this reason in the last few years the attention of scientific community is focused on the recycling of the spent LIBs in order to extract the CRMs and use them for the development of second life batteries. The methods used until now for the extraction of metals from the spent batteries involve hydrometallurgy and pyrometallurgy which are high energy-consuming approaches[2]. In the last few years the approach to the extraction of these metals is shifting towards more sustainable methods. A good option that is gaining ground is to recover metals from cathodes via Deep Eutectic Solvents (DES). The use of DES in metals recovery processes has several advantages, for example the solvent can be formed by green, non-toxic and very cheap components, the operating conditions are milder than those used in pyrometallurgy and hydrometallurgy and in some cases the same deep eutectic solvent might be used for several subsequent extractions[3][4]. As a task of the RENOVATE WP4 "CAM (Cathodic Active Materials) recovery: solvometallurgy and direct recycling" in this study we present a novel three-component deep eutectic solvent (DES) composed by Choline chloride : Lactic acid : Urea for the recovery of Li, Ni and Co from End of Life (EOL) batteries via Microwave Assisted Extractions (MAE). The ternal DES composition and the parameters of the microwave extractions procedure are optimized through the Design of Experiments (DoE) performing a mixture-process design. The metals extracted by the DES are precipitated, characterized and used to assemble second life CAM which preliminary electrochemical performances are compared to those of the commercial cathodic active materials.



Figure 1. Recycling process of LNCO cathodes using DES ChCl:LA:Ur via microwave assisted extraction.

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ANODE EMBEDDED INTERNAL HEATER TECHNOLOGY FOR LOW TEMPERATURE OPERATION LITHIUM-ION BATTERIES

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The performance of a lithium-ion battery is dictated by the kinetic mechanisms, in terms of charge transport, of each of the constituent materials. Such mechanisms are the electronic conductivity and lithium diffusion inside the anodes and cathodes, the ionic conductivity of the electrolyte as well as the ionic conductivity and charge transfer of the formed interphases and interfaces respectively. On the cell level, lithium-ion technology aims to optimize these kinetic mechanisms based on the engineering of materials and geometries.

All the kinetic mechanisms are affected by temperature [1]. As the temperature decreases, charge transport is slowed down and the battery cannot reach its rated current capability and capacity, significantly impacting specific energy and power. Apart from the kinetic limitation, at temperatures well below 0 °C, liquid electrolytes can freeze making the battery inoperable.

To overcome this problem, different heating methods are proposed and classified in external and internal, with respect to their placement either outside the battery module or acting inside the cell respectively [2]. External heating introduces significant increase in the bulk and weight of the system and complexity in the installation, while it requires an additional power source. Most significant, however, is the non-uniform distribution of temperature inside the module [3]. On the other hand, Internal heating methods aim towards exploiting the energy of the battery itself, operating in a more efficient way but modifications on a cell level are required.

We propose an efficient way to heat a cell by intervening directly to the active material. Using microwires, a heating element can be embedded in the anode. Based on this concept, insulated nichrome 80 microwires with a diameter of 40 μ m were placed firmly on top of a copper current collector sheet, which was then coated with the anode slurry. The modified anode and an LFP cathode were combined in a pouch cell with four terminals, two for the cell operation and two for the heating element, as seen in figure 1 (a). The internal heating was applied in three different controlled environment temperatures: 0 °C, -18 °C and -37 °C. The cell temperature was measured as a function of time as seen in Figure 1 (b) for two different heating rates, by applying different currents through the heating element. Based on the dynamics of Li ion transport and electrolyte conductivity, it was possible to determine the exact temperature inside the cell.

The proposed method provides heating locally, at the point where the limiting charge transport processes take place. That is, the intercalation of lithium ions in the graphite and the charge transfer between the lithium ions and the graphite. The cells demonstrated stability and high values in their capacity with less than 0.5 % average capacity fade ratio. They showed high coulombic efficiency, > 99.5 %, during C/2 Charge – C/3 Discharge at an ambient temperature of 0 °C, while retaining 99.5 % of the initial RT capacity after 30 cycles in low temperature.



Figure 1. (a) Schematic of the embedded heater element. (b) Cell temperature vs time in a controlled temperature of 0 °C, -20 °C and -40 °C.

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EFFICIENT DELAMINATION AND RECOVERY OF LFP CATHODE MATERIALS: A SUSTAINABLE APPROACH FOR LITHIUM-ION BATTERY RECYCLING

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By 2030, the global volume of end-of-life (EoL) lithium ion batteries is estimated to exceed 2 million tons per year, while production scrap could account for between 5% and 7% of the manufactured material,1 equating to hundreds of thousands of additional tons annually. In this context, the efficient recovery of active materials from these waste streams is not only a key challenge for the development of sustainable recycling processes but also a crucial strategy to enhance economic feasibility and reduce dependence on critical raw materials.



Figure 1: a) Time-dependent pH curves of the oxidative medium during the delamination process at different oxidant concentrations. b) SEM image of an LFP particle, recovered at a concentration of 0.4% by w/w of oxidant.

This study presents a delamination method for separating the black mass from the current collectors of commercial lithium iron phosphate (LFP) battery cathodes, aiming to optimize the recovery of active material for reuse in new batteries. The developed protocol consists of two consecutive stages: a chemical treatment followed by a mechanical process. First, cathode scraps were treated in an oxidative aqueous medium. Subsequently, low-energy ball milling and sieving were employed to optimize the recovery of the active material. During the aqueous treatment, key experimental variables such as oxidant concentration and temperature were evaluated, along with the analysis of metal concentrations in solution. Additionally, chemical composition analysis of the black mass was conducted, and the structural quality of the recovered active material was assessed.

The results demonstrated an effective separation of the current collector, with a black mass recovery yield exceeding 98 wt.%. No significant structural changes were observed in the recovered LFP active material compared to the original sample, while the concentration of contaminants was significantly reduced.

The developed delamination protocol emerges as a promising complementary process to traditional hydro- and pyrometallurgical methods. Its robustness and scalability make it suitable for both direct recycling of cathodic materials and as a pretreatment for waste before active material leaching, contributing to more efficient and sustainable lithium ion battery recycling strategies.

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SUPERCRITICAL CO₂ EXTRACTION FOR LITHIUM RECOVERY AND ORGANIC CONTAMINANT REMOVAL IN BATTERY RECYCLING PROCESSES

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Over the past decade, the consumption of lithium batteries has been driven primarily by the increasing demand for electromobility and stationary energy storage applications. Projections indicate that, in the coming decades, large volumes of end-of-life batteries will be discarded. Consequently, a proportional amount of waste containing highly toxic substances and critical materials—important not only for their economic value but also for their limited geographical availability—will be generated.^[1] Scalable protocols that complement traditional pyro/hydrometallurgical processes are required to enable the treatment of effluents for both environmental remediation and the recovery of critical materials.

Supercritical solvents are substances at temperatures and pressures above their critical point, exhibiting densities similar to liquids and diffusivities comparable to gases. This unique state offers a wide range of possibilities for extracting organic components from solid and liquid matrices bv adjusting the process pressure and temperature.^[2] In this study, the use of supercritical CO2 was evaluated for the decontamination and lithium recovery from process water generated in lithium battery recycling. The protocols involved a combination of static and dynamic extractions, as well as the use of organic chelating agents for lithium recovery.



Figure 1: Lithium recovery yields (%) reached by supercritical CO_2 extraction as function of different experimental parameters.

In the first stage, the extracted organic compounds were purified and quantified using a novel solidphase extraction (SPE) method combined with gas chromatography (GC-FID).^[3] This approach enabled the identification and recovery of concentrated solutions containing organic carbonates, ageing by-products, and organic solvents commonly used in battery manufacturing. The correlation between extraction yields and process conditions allowed for the determination of optimal parameters for the selective recovery of each component.

For lithium recovery, the effluent was alkalinized to facilitate chelation-using HTTA (a diketone) in combination with TOPO (a phosphine). A series of different experimental conditions were evaluated, achieving lithium recovery yields exceeding 70% in a single static-dynamic cycle. However, the addition of co-solvents with different polarities (Methanol and Ethyl Acetate), as well as an increase in pressure (from 160 to 190 bar) and CO_2 flow rate, negatively affected the recovery efficiency. Nevertheless, using lower-polarity solvents in the supercritical phase enhanced the selectivity of the chelating system for lithium.

Supercritical CO₂ extraction protocols proved to be effective for both the removal and concentration of organic compounds and for the chelation-mediated recovery of critical metals.

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Pushing the Limit of Sub-Millimeter Batteries by On-Chip Self-Assembly

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The development of tiny energy storage devices with high energy density is critical to realizing autonomous intelligent microsystems and small-scale robotic applications.¹ In this presentation, I will explore the potential of zinc battery chemistries for sub-millimeter-scale energy storage, highlighting their advantages over conventional lithium-based systems. Zn batteries, with their superior air stability and seamless compatibility with microfabrication processes, offer significant promise in overcoming the challenges of miniaturization to sub-millimeter footprints. I will discuss recent innovations in using micro-origami technology to develop tiny Zn batteries, which have crushed the footprint limit of 1 mm², achieving a footprint capacity of more than 1 mAh/cm² and extending the boundaries to deep-submillimeter scale (< 0.1 mm^2 , Figure 1).^{2,3}

Additionally, this talk will emphasize the material innovations for microfabrication techniques that have enabled Zn batteries to take center stage in the quest for reliable, efficient energy storage at sub-millimeter scales. I will highlight the development of different photolithographable polymer electrolytes to improve the cycling stability of Zn and therefore the lifetime of a full cell.^{4,5} Moreover, emerging battery chemistries such as cathode-free configuration and decoupled electrolyte design will be briefly discussed to show the potential of Zn batteries in advancing the performance of sub-millimeter batteries.⁶

Finally, I will demonstrate the proof-of-concept integration of sub-millimeter zinc batteries with microactuators, highlighting the significance of modulating zinc ion behavior in an aqueous solution to simultaneously realize efficient actuation and energy storage.



Figure 1. Deep-submillimeter microbatteries

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