Large-Scale Atomistic Simulations of Battery Materials and Interfaces

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We are developing software for supercomputers for atomistic electrochemical simulations under operational conditions in order to capture the essential chemistry and physics of devices such as batteries but also provide the parameters needed for bridging atomistic with higher scale continuum simulations. Our developments are within the ONETEP program [1], which is based on a linear-scaling reformulation of density functional theory (DFT) that allows simulations of orders of magnitude more atoms than conventional DFT approaches, for more realistic models. In this talk, I will outline our developments, which include solvent and electrolyte models [2], and simulations at fixed voltage with respect to a computational reference electrode [3]. Also, I will describe our ongoing uses of large-scale DFT combined with machine learning force fields to provide longer timescale simulations for processes such as the chemistry taking place during Solid-Electrolyte Interphase (SEI) formation – a very complex multi-component material crucial for battery operation and how voltage controls the deposition of SEI precursors [4]. Finally, I will summarise recent applications of these tools to the process of lithium metal deposition on anodes and its competition with Li dendrite formation [5], one of the main mechanisms of battery degradation.

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Materials Modelling of Nb based Oxides: Answering Battery Industry Relevant Research Questions

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Nb based oxides are gaining research interest for fast charging battery anode applications. They generally present in the Wadsley-Roth (W-R) crystal structures consisting of a central block, with Nb in corner sharing sites and edge sites that adjoin neighbouring blocks. In particular, the transition of Nb (3+ □ 5+) oxidation states allow for high lithium intercalation. The niobium oxides can be partnered with other transition metals (TM) such as Vanadium (V), Phosphorous (P), Tungsten (W) and Titanium (Ti) among others, to avail a plethora of materials for exploration. Ranging in the size of block (3x3 □ 3x5 and mixed block systems) and varying oxidation states which result in increasing the ion storage capacity of these materials. Which makes them very attractive for battery applications. In this talk, 3x3 WR crystal structure and Li ion intercalation will be reported using Density Functional Theory to answer industry relevant questions.

Factors Governing Thermal Conductance Behaviour of Plasticized Single-Ion Conducting Polymer Electrolytes: A Molecular Perspective

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Single-ion conducting polymer electrolytes (SICPEs) are promising candidates for next-generation batteries due to their high lithium-ion transference number, which approaches unity. While their relatively low lithium-ion conductivity can be significantly enhanced by incorporating small amounts of plasticizers, their thermal conductance – crucial for effective thermal management in batteries – has received comparatively little attention. Adequate thermal conductance is essential for rapid heat dissipation, helping to prevent heat accumulation that could potentially lead to thermal runaway or spontaneous combustion. To investigate the thermal conductance behaviour of SICPEs, we performed both equilibrium and non-equilibrium Molecular Dynamics simulations to examine lithium-ion conductivity and thermal conductivity of modified polyethylene terephthalate–based SICPE in the presence of ethylene carbonate plasticizers. The simulated lithium-ion conductivities of SICPEs at different ethylene carbonate concentrations and temperatures show good agreement with experiments, as well as the thermal conductivity of polyethylene terephthalate. Our results suggest that adding 50 wt% ethylene carbonate at 300 K yields the optimal balance, with a maximum thermal conductivity of 0.25 W/m/K and a satisfactory lithium-ion conductivity of 2.94 × 10-4 S/cm. The enhanced lithium-ion conductivity is attributed to the weakened interaction between lithium ions and both ethylene carbonate molecules and polymeric backbone. Furthermore, the predominantly thermal conductance medium shifts from polymer to ethylene carbonate upon loading ethylene carbonate. These insights advance the understanding of heat transfer behaviours in SICPEs and offer guidance for developing safer polymer electrolytes from a thermal management perspective.

A rule that's made to be broken?
Reframing the Arrhenius law and the calculation of activation energies for ion transport in solid electrolytes.

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Many different electrolytes have been proposed for use in next-generation solid state batteries, with the potential to provide greener, more efficient energy storage [1]. A key stumbling block in proposed solid electrolytes is often low conductivity. Therefore, when considering a new material, the first step is to understand ion transport.

The Arrhenius law is ubiquitous in the physical sciences. It describes how an observable property scales with temperature, T, as $\exp(\pm A/T)$ for a constant A, interpreted as an activation energy. Often transport coefficients are well-described by an Arrhenius fit. However, there are many exceptions, making it difficult to extract an activation energy from macroscopic transport properties. One reason for non-Arrhenius behaviour is correlated motion [2]. For example, the diffusion of individual ions depends upon the diffusion of neighbouring ions. Ions may also become trapped due to local structure and undergo movement but in a back-and-forth motion that does not contribute to overall transport.

Using methods developed for supercooled liquids [3], we identify ion 'jumps', particularly those that are productive for transport, and correlated motion. The method enables us to extract the true underlying activation energy from diffusion data, and recover the Arrhenius law. Here, we will demonstrate the methods for low-dimensional-networked Li-rich anti-perovskites, potential solid electrolytes [4].

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Update on the Faraday Programme and UK-US modelling discussions

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The presentation will provide an overview of the FI research programme including the new transformational challenge areas which are focussing on ultra low-cost storage and ultra high-performance energy solutions, the former for grid based long duration energy storage and the latter the aviation sector. Together with the Hartree Centre the FI jointly organised an US-UK workshop to explore the role of AI and exascale computing in the acceleration of battery modelling and manufacturing scale up. The main conclusions of the workshop will be presented including the Technical Grand Challenges identified.

Beyond Funnel-like High Throughput Screening: An Expert Guided Multi-scale Nexus for Polynary Perovskite Electrocatalyst Exploration

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Navigating the vast compositional landscape of polynary oxide electrocatalysts is historically constrained by funnel-style screening paradigms that treat high-fidelity data as binary gates rather than rich training signals. Alternatively, in this work, we introduce an expert-guided, multi-scale nexus paradigm that bypasses these limitations through recursive alignment of atomic, experimental structural, and mechanism-informed electronic features. By implementing a ‘Nearest Grid Interpolation’ strategy, computational costs for high-fidelity density functional theory (DFT) evaluations are reduced by two orders of magnitude within the search space. Through active learning loops integrated with human-in-the-loop (HITL) expertise, a localized synergistic optimum is identified at $Pr_{0.01}Sr_{0.99}Co_{0.66}Fe_{0.34}O_{3-\delta}$(PSC0.66F), achieving a state-of-the-art overpotential of 338 mV at 10 mA cm-2. Symbolic regression reveals that a complex nexus of the O 2p-band center, its bandwidth, and Fe e_g orbital filling dictates the intrinsic activity. Experimental validation confirms that this optimized electronic configuration facilitates highly efficient lattice oxygen mechanism (LOM) kinetics, supported by accelerated bulk oxygen diffusion and dynamic surface reconstruction. This multi-scale framework provides a robust template for the accelerated discovery of complex materials by transforming fragmented data tiers into a cohesive, learnable predictive landscape.

Halide-Stabilised $LiBH_4$ as a Solid Electrolyte for Li-Ion Batteries

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$LiBH_4$ is a complex hydride that has attracted significant interest in Li-ion and solid-state battery research, primarily as a solid electrolyte and, more experimentally, as a component of electrode systems [1,2]. One effective strategy to enhance its electrochemical performance is halide stabilization [3]. In this work, density functional theory (DFT) simulations are employed to systematically investigate the effects of partial anion substitution of BH_4^- with Br^- and I^- on the structural and thermodynamic stability of $LiBH_4$. Our calculations indicate that both halide substitutions preferentially stabilize the hexagonal phase over the orthorhombic phase under ambient conditions. Among the two dopants, I^- exhibits a stronger stabilization effect than Br^-, which can be attributed to its larger ionic radius and higher polarizability. The diffusion behaviour of Li^+ ions in the presence of halide substitutions is analysed and compared with that in pristine $LiBH_4$.

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Phonon driven non-equilibrium triggers for thermal runaway in battery electrodes

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Thermal management remains a critical bottleneck for next-generation battery development, as inadequate heat dissipation drives performance degradation and thermal runaway. Here, we present a full multi-scale approach to the thermal transport in battery electrodes, identifying key factors that trigger thermal runaway. We couple phonon calculations with lattice dynamics to map the intercalation-dependent thermal properties of a model electrode material, Li_xZrS_2. We show that thermal conductivity changes non-monotonically with lithium concentration, arising not from lithium vibrational modes but from charge redistribution and bond-strength modulation. Integrating these properties into multi-scale grain-resolved simulations reveals localised temperature gradients and finite-speed thermal wave interference—mechanisms that can trigger sub-grain thermal breakdown. By explicitly capturing these effects, our framework bridges atomistic physics and device-level behaviour, identifying mechanisms that result in hotspot formation and reduced thermal stability.

First-principles Modelling of Infrared and Raman Spectra

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Infrared (IR) and Raman spectroscopy are established materials-characterisation tools and are routinely employed for a wide range of energy materials, but the spectra of complex materials can be difficult to interpret. We have developed a comprehensive workflow for predicting infrared (IR) and Raman spectra using density-functional theory (DFT), which we have implemented in our open-source Phonopy-Spectroscopy code.

Phonon frequencies and eigenvectors are obtained within the harmonic approximation. IR spectra are modelled by computing the mode dipole oscillator strengths and infrared dielectric function, providing access to absorption, dielectric loss, reflectivity and transmission spectra, as well as related properties such as the static dielectric constant. Raman spectra are obtained by computing numerical polarisability derivatives from either the high-frequency dielectric constant or the energy-dependent dielectric function, the latter of which allows for partial inclusion of resonance effects. Finally, linewidths can optionally be obtained from perturbative three-phonon interactions.

The code can perform IR and Raman simulations on both single crystals and powders, accounting for the orientation of the crystal and the polarisation of the incident and detected radiation. This allows for the simulation of a wide variety of measurements, ranging from routine laboratory characterisation to the more sophisticated experiments possible with high-end or custom instrumentation. The code has also been designed in such a way as to “decouple” the preparatory DFT calculations and spectrum modelling, to better facilitate collaboration between theory and experimental groups.

We have benchmarked the code against materials for photovoltaic, thermoelectric and nuclear applications. Simulated spectra, obtained at very modest computational cost, are generally sufficient to assign the major spectral features, while careful calculations can reproduce measured spectra with near-quantitative accuracy. More recently, we have also been developing auxiliary tools for visualising and characterising vibrational modes, with a view to making complex spectra easier to interpret and e.g. identifying bands related to defects.

Autonomous Data Engineering for Chemistry using general purpose models: A Case Study on Lithium Metal Electrolytes

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Lithium metal batteries (LMB) are regarded as a promising solution to meet the market demand for energy storage systems with high specific capacity, but the implementation of lithium metal anodes is currently hindered by poor cycle life and uncontrollable side reactions between Li metal and liquid electrolytes. Liquid electrolyte engineering, which involves mixing different molecules to create electrolytes with specific properties, is ultimately the most cost-effective approach for making LMBs viable. However, there are a myriad of possible electrolyte formulations due to the large number of commercially available molecules, recently synthesized electrolyte-specific compounds, and various strategies for fine-tuning electrolyte components. Consequently, there is an increasing need for theory-guided rational design of new electrolyte formulations for LMBs, aiming to reduce research costs and avoid “trial-and-error” approaches, which is currently hindered by the fragmentary reporting of data and the lack of generalized, widely accepted performance indicators for electrolytes.

Due to recent advances in automation and computing, these challenges can be tackled with approaches such as automated retrieval of pertinent information with large language models that feed high-throughput computational workflows. These are often trained or fine-tuned for specific domains and tasks, which drastically increases the time and cost to apply them to new fields. This talk will describe the potential of using general-purpose state-of-the-art models instead, which not only lowers the cost of our approach but also lowers the barrier for entry, potentially allowing for a much faster digitalization of current domain knowledge in multiple areas of chemistry and other natural sciences. It will cover extraction performance and challenges as well as the integration with high-throughput computational workflows such as molecular dynamics and density functional theory on the use case of liquid electrolytes for lithium metal anodes.

Interface design in zero-excess Li metal batteries guided by DFT and molecular dynamics with machine-learning interatomic potentials

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The concept of anode-free or zero-excess Li metal batteries (ZELMBs), has attracted a lot of interest as it simplifies the battery manufacturing process, improves safety, and could bring significant cost reduction. However, the Li stripping/plating occurring on the surface of the commonly adopted Cu current collector, are one of the main disadvantages of ZELMBs.[1] In this work, we present an efficient computational workflow to screen the performance of interlayer metals at the anode side, combining DFT, AIMD and MD with fine-tuned machine-learning potentials, aiming to obtain stable deposition/stripping process of Li during operation. Initially, we performed density functional theory (DFT) calculations to investigate Li deposition on 19 interlayer metals, including those forming solid solutions with Li (Ca, Mg, Zn, Al, Ag, Au, Pt, Pb, Tl, In, Y, Tm, Sb, Pd), those forming intermetallic alloys with Li (Sn, Si, Ge), and those non-alloying with Li (Cu, Ni). We derived a relationship between the Li deposition overpotential and diffusion barriers revealing that Mg-Li alloys with low Li content (<11.25 at. % Li), as well as Li-In, Li-Tl, Li-Pb, Li-Tm, and Li-Y alloys with high Li atomic concentration (>98 at. % Li), offer the best performance for efficient Li deposition. We conducted large-scale molecular dynamics simulations using machine learning interatomic potentials (MLIPs), fine-tuned with AIMD and DFT data, for the Li-Cu, Li-Zn, Li-Mg, and Li-Bi systems, considering varying amounts of Li metal, and we studied the dynamics of the crystallization and alloying of Li on those different interfaces. For the Li-Cu interface, we observed that with low amounts of Li metal, Li aggregates are formed, leaving empty spaces on the Cu surface. Solid solution formation was observed at the Li-Zn and Li-Mg interfaces, with a larger presence *hcp* lattice of Li in the former and *bcc* lattice of Li in the latter. Finally, we predicted that Li preferentially forms Li_3Bi intermetallic alloys at the Li-Bi interface, based on the morphology and coordination analysis. Experimental validation through SEM, XRD, AFM, and ToF-ERDA corroborates the predicted alloying and phase evolution. These insights provide a general atomistic framework for rationally designing metal interfaces for ZELMBs.

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Understanding Interfacial Reactions by Combining Operando Soft X-ray Spectroscopies with Spectral Simulations

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The performance of Li-ion batteries depends critically on reactions taking place at the interfaces between their solid electrodes and the liquid or solid electrolyte. Obtaining chemical information from these interfaces under realistic conditions is critical to the selection and design of improved materials. Core loss spectroscopies can provide powerful element-specific insights, but this requires methods to extract information with interface-sensitivity, and approaches to model the spectral features observed.

In this direction, I will summarise our results combining a suite of complementary core-loss spectroscopies to obtain a depth-resolved (10 nm to >10 μm) account of the redox processes in Ni-rich cathode materials used in Li-ion batteries.[1] Soft X-ray absorption (sXAS), X-ray Raman, and Electron Energy Loss Spectroscopies distinguish reversible bulk redox processes from near-surface degradation. Comparison with density-functional theory (DFT) and charge transfer multiplet (CTM) calculations,[2] provides new insight into the speciation of the Ni environments, showing that bulk redox occurs through Ni–O rehybridization and importantly does not involve molecular O_2.

I will further discuss our work with enclosed electrochemical cells sealed with thin (<100 nm) silicon nitride membranes, that allow sXAS of solid-liquid interfaces under electrochemical control.[3] We have thereby observed solid-electrolyte interphase (SEI) formation on silicon anodes in Li-ion batteries, and determined how the presence of the widely-used additive fluoroethylene carbonate proves beneficial in this process. A full understanding again relies on DFT simulations to identify the origins on key spectral features and thereby assign the chemical species present.

These examples demonstrate the importance of combining powerful interface-sensitive core loss spectroscopies with advanced spectral simulation tools to obtain a detailed understanding of interfacial electrochemical reactions.

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Atomistic Understanding of Interfaces in Next-Generation Batteries

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The interfaces between and within materials are where energy technologies, particularly batteries, succeed or fail. The performance of commercial lithium-ion batteries is governed by the chemistry of solid- and cathode-electrolyte interphases, while next-generation solid-state batteries experience failure from lithium dendrites and anode-free batteries suffer from inhomogeneous lithium plating and stripping. In this presentation, I will demonstrate how atomistic simulations can help us in the understanding, design and improvement of next-generation batteries. In particular, I will present our recent research as part of the Faraday Institution SOLBAT and Degradation projects. In the first part, I will discuss our findings on the fundamentally different mechanisms of dendrite initiation at the grain boundaries of garnet Li7La3Zr2O12 and argyrodite Li6PS5Cl solid electrolytes. In the second part, I will focus on the optimisation of formation cycling based on electrolyte solvation structure engineering in anode-free batteries.

Amorphous-like thermal conductivity and high thermoelectric figure of merit in “*π*” SnS and SnSe

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The tin chalcogenides SnS and SnSe are an important family of optoelectronic materials with applications to clean-energy technologies including photovoltaics (PV) and thermoelectric (TE) generators. In particular, SnS has been widely studied as a potential PV material, and orthorhombic *Pnma* SnSe is one of the current flagship high-performance TEs. The recently-discovered *π*-cubic phases of SnS and SnSe have similar local structures to the *Pnma* phases, have been shown to be both dynamically stable and close to the convex hull, and have been identified as potential high-performance TEs. However, the physical properties required to determine the TE figure of merit, *zT*, have yet to be measured.

We have applied a fully *ab initio* modelling approach to determine the TE properties and *zT* of *π*-cubic SnS and SnSe as a function of temperature and carrier concentration with both p- and n-type doping. The complex structures push the “particle-like” contribution to the lattice thermal conductivity, κ_{latt}, below the amorphous limit, resulting in an ultra-low κ_{latt} from room temperature upwards. The *π*-cubic symmetry supports larger Seebeck coefficients than the *Pnma* phases, but higher carrier effective masses and stronger electron scattering require high doping levels to optimise the conductivity and power factors. For *π* SnSe, we predict a low-temperature n-type *zT* comparable to Bi_2Te_3, and a high-temperature *zT* competitive with the flagship *Pnma* SnSe. These results demonstrate the exceptional promise of these systems as high-performance thermoelectrics, and highlight structural complexity as a route to optimising low-temperature *zT* by minimising the κ_{latt}.

From Constant-Potential Modelling to Sulfur-Engineered Hard Carbons: Ab-Initio Simulation of Electrochemical Interfaces

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Electrochemical systems operate under applied bias, yet most first-principles simulations are performed at fixed charge, creating a boundary-condition mismatch that obscures voltage-dependent structure and kinetics. In the first part of this talk, I present computational methodologies that enable constant-potential modelling within density-functional theory, from ion-imbalance approaches to open-boundary electronic formalisms. These methods provide thermodynamically consistent descriptions of electrified interfaces, capturing interfacial polarization and charge transfer under controlled bias.

In the second part, I apply these ideas to sodium-ion batteries, focusing on sulfur-doped hard carbon anodes. Combining experiment with atomistic modelling, we show that sulfur chemisorption weakens sodium binding at defect-rich carbon sites, lowering the energetic penalty for desodiation and improving reversibility. First-principles models incorporating curvature and defects reveal how tuning adsorption energetics enhances initial Coulombic efficiency and cycling stability.

Modeling Redox Potentials and Electrolyte-Organic Conjugated Polymer Interactions for Electrochemical Energy Storage Electrode Applications

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Organic conjugated polymer electrodes are emerging as promising alternatives to inorganic materials for rechargeable batteries due to their low weight, mechanical flexibility, tunable structure, and sustainable processability. Their intrinsic redox activity enables reversible electrochemical reactions, while their modular architecture comprising a π-conjugated backbone and functional side chains offers multiple molecular design parameters to optimize performance. The conjugated backbone governs redox potential and electronic charge transport, whereas side-chain engineering modulates ionic conductivity, electrolyte compatibility, and cycling stability. Efficient polaron formation, stabilization and optimal redox potentials critically depend on a balanced interplay of backbone planarity, sidechain functionality, and microstructural order. Despite recent reports showing that polymer electrodes can approach the energy densities of inorganic electrodes, the molecular mechanisms governing polaron stabilization, ion coordination, and electrolyte-polymer interactions remain incompletely understood. In this work, we employ molecular dynamics simulations to investigate polaron formation and redox behaviour in diketopyrrolopyrrole-thiophene-based conjugated polymers. By systematically varying backbone and side-chain chemistry, as well as solvent and electrolyte environments, we establish structure-property relationships linking molecular design to redox potential, ion coordination, and polaron stabilization. These insights provide a mechanistic framework for rationally designing high-performance, durable, and sustainable conjugated polymer electrodes for next-generation energy storage systems.

Directly following Potassium Intercalation in Prussian Blue Cathodes via Potassium K-edge X-ray Absorption Spectroscopy

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Prussian blue (PB) frameworks offer promise as low-cost cathodes for Na^+, K^+ and Mg^{2+} aqueous and non-aqueous batteries due to their open, ion-exchangeable structure.

However, significant structural complexity—especially $[Fe(CN)_6]^{4-}$ vacancies, $(H_2O)_6$ clusters, and occluded water—complicates understanding of intercalation mechanisms. Here we demonstrate that operando K-edge potassium XANES, supported by theoretical modelling, can directly probe the local coordination of K^+ during cycling. This approach enables real-time correlation of electrochemical behaviour with ion-site evolution in PB and PBA samples of differing crystallinity.

Differentiated K^+ species were clearly observed by XANES, associated with the fully intercalated Prussian white structure, partial intercalation within Prussian blue, and solvated K^+ in the electrolyte. Theoretical simulations of Prussian blue, and associated XANES simulations, show that K^+ resides within the cavities of the structure, but located away from the centre of the primitive cubic cavities (8c site) and towards the corners of the cavities (32f′ position), due to structural defects and occluded water.

Operando potassium XANES during electrochemical redox reactions of Prussian blue and Mn analogues definitively showed that K^+ was the charge balancing species with the structural. Clear evolution between the 3 observed potassium states is seen, but with little evidence of solid solutions (i.e. a loss of K signal without observed phase change). In large >100 nm domain sized Prussian blue samples, the intercalation process was reproducible over repeated cycles, although full deintercalation of K^+ was kinetically hindered. Significant degradation in electrochemical performance of a Prussian blue sample with smaller domains (<14 nm), was identified as being caused by K^+ becoming trapped within the partially intercalated structure.

The findings show the potential of XANES to monitor the speciation of alkaline charge carriers, in a host of different battery and other electrochemical processes, has exciting future potential in characterization and development of new materials.

JPhys Energy at IOP Publishing

Aswathy Girija[*]

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Journal of Physics: Energy is an interdisciplinary, fully open-access journal from the Institute of Physics (IOP) Publishing. Since its launch in 2018, the journal has brought together physicists, chemists, materials scientists, and engineers to share advances across the energy landscape. Building on the 50-plus-year legacy of the *Journal of Physics* series, *JPhys Energy* combines rigorous peer review with a strong commitment to open-science principles, ensuring that high-quality research reaches the widest possible community.

In this talk, I will introduce the journal's vision and scope and outline how we support researchers through trusted peer review, diverse article formats including Tutorials, Technical Notes and Quick Start Guides, and our flagship Roadmap programme. I will spotlight a selection of articles that showcase the breadth and ambition of the field, and highlight a recent event organised for early career researchers in energy. Above all, this session aims to welcome the community and demonstrate how *JPhys Energy* can serve as a vibrant home for cutting-edge computational and experimental research in materials for energy.

Performance and Circularity in Sodium-Ion Batteries

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Delivering high energy density storage for global decarbonisation demands innovation beyond incremental improvements to established lithium-ion systems. While lithium-ion technology continues to dominate due to its energy density and manufacturing maturity, sodium-ion batteries are emerging as viable complementary technologies. However, when considered within a circular economy framework, the lower intrinsic material value of sodium-ion systems challenges conventional recycling economics. Alternative drivers, such as supply chain resilience, geopolitical criticality, and resource security, may therefore provide stronger justification for recovery and reuse strategies.

This talk integrates advances in materials chemistry with circular design principles. The landscape of sodium-ion systems is outlined, highlighting how material selection influences performance, durability, and end-of-life considerations. A layered oxide–hard carbon configuration is used as a case study. Morphological optimisation of an O3-type layered sodium cathode via boron oxide modification is presented, where boron functions both as a crystal growth flux and a lattice dopant. This dual role directs dominant (003) facet exposure, reinforces the metal–oxygen framework, enlarges Na-layer spacing, and mitigates the durability–power trade-off by enabling fast in-plane Na^+ diffusion alongside enhanced structural stability. Circularity is further addressed through a low-temperature, direct recycling route for hard carbon anodes. Mechanical “ice-stripping” for electrode separation, combined with binder negation at 300 °C under nitrogen, preserves graphene-layer integrity and maintains near-pristine electrochemical performance. Morphology control again proves critical, as higher temperatures induce irreversible structural damage.

Together, these advances demonstrate that morphology engineering and end-of-life strategy must be co-designed to enable technically robust and resource-secure sodium-ion battery systems within a circular economy framework.

Investigating the asymmetric electrochemical, structural and electronic properties of Mn-rich Li(Mn,Fe)PO4 electrode materials

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Olivine-type phosphates $LiMn_xFe_{1-x}PO_4$ are attracting increasing interest as positive electrode materials for lithium-ion batteries due to their low cost and good electrochemical performance. However, the effects of the mixed Mn/Fe composition on lithium intercalation and ion transport are not fully characterised, especially in Mn-rich compositions. In this study, the electrochemical, structural and ion transport properties of Mn-rich $LiMn_xFe_{1-x}PO_4$ ($x = 0.6, 0.7, 0.8$) (LMFP) are investigated using a combination of experimental and materials modelling techniques. Considerable asymmetry in charge/discharge profiles is found, which highlights the complexity of the mixed-metal system. An intricate lithium intercalation mechanism is observed, including both solid solution and two-phase regions. While the Fe/Mn cation disorder causes the oxidation to proceed mostly via a solid solution mechanism, the Mn plateau remains associated with a two-phase process. Ab initio simulations indicate that Li+ diffusion occurs along one-dimensional channels parallel to the crystallographic b-axis following a curved trajectory, and find favourable Li/Fe and Li/Mn anti-site defect formation. Analysing the band gaps of the lithiated and delithiated phases revealed that Mn substitution of Fe can improve the electronic conductivity, suggesting asymmetric electronic behaviour.

AI-Accelerated Screening of Electrolyte Solvents for High-Safety Batteries

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Flammability and volatility of conventional carbonate-based electrolytes remain central obstacles to improving battery safety, particularly for high-energy applications. We present a data-science-driven, high-throughput workflow to identify safer nonaqueous electrolyte solvents and functional additives by directly screening molecular structure for key safety and transport-relevant properties. Graph neural networks trained on the DIPPR 801 database are used to predict flash point, melting point, dielectric constant, and liquid viscosity from SMILES-derived molecular graphs and 3D conformers. We combine these predictors with chemically motivated substructure filters, a synthetic accessibility criterion, and a fast dynamic-stability screen using machine-learned vibrational analysis. Applied to two complementary molecule datasets—a large catalog of purchasable molecules (Mcule) and a chemically enumerated set of small molecules (GDB 13)—the approach evaluates over one billion candidates and yields thousands of promising molecules, including both readily available compounds and de novo structures. We highlight representative candidates, including a known battery additive rediscovered by the pipeline and multiple repurposable molecules used in other industries, and discuss how this workflow can guide experimental down-selection and next-generation electrolyte design.

Modelling and Design of 3D Electrode Microstructures for Energy Storage Batteries

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Electrode microstructure plays an important role in the performance of energy storage batteries [1]. Building a clear understanding of how battery performance is affected by the electrode microstructure is necessary to design the optimal electrode microstructure for high performance.

This talk will introduce a 3D pore-scale lattice Boltzmann (LB) model developed at Surrey to simulate multi-physics transport phenomenon and gas-liquid two phase flow, coupled with electrochemical reactions, for linking the complex 3D porous electrode microstructures with electrochemical performance. The LB model is developed to simulate 3D porous electrodes in redox flow batteries (RFBs). Three electrode structures (SGL paper, Freudenberg paper, Carbon Cloth) are reconstructed from X-ray computed tomography (CT) and input to the model for performance simulations. The simulated pressure drop and electrochemical performance are compared with the experimental measurement in an organic RFB. Excellent agreement is achieved between the simulated and experimentally measured electrochemical performance, indicating the validity of our model [2]. The optimal electrode structures are also revealed.

Recently we have applied the model to simulate the composite cathodes of all solid-state Li batteries (ASSLBs). First, we simulate the heat conduction in computer-generated 3D composite cathodes with different particle size, porosity and tortuosity. Tortuosity of different phases emerges as a nonnegligible factor influencing the effective thermal conductivity, and a polynomial regression method is applied to relate the effective thermal conductivity with the tortuosity values of all the three phases in the composite cathodes [3]. The performance of ASSLBs is critically governed by the effective ionic conductivity (EIC) and effective electronic conductivity (EEC) within their composite cathodes. The LB model is applied to an X-ray tomography obtained sulfide-NMC composite cathode and showed excellent agreement in predicted EIC and EEC values with experimental measurements. EIC and EEC of various composite cathodes with different particle size, materials ratio and porosity are investigated. We proposed the overall effective conductivity concept by balancing the EIC and EEC contributions, to identify optimal ratios of cathode active materials to solid electrolyte [4]. These findings offer critical insights into the design of high-performance composite cathodes and underscore the importance of integrated microstructural optimization.

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Modelling Structural Disorder and Ionic Transport in Sodium Oxychlorides

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Sodium-based solid-state batteries are attracting growing interest as a scalable and sustainable alternative to lithium-ion batteries, motivated by sodium's abundance and wide geographical distribution. Beyond economic advantages, solid-state sodium batteries offer further benefits including improved safety, higher energy density and longer lifetimes enabled by solid electrolytes. Among candidate solid electrolytes, halide-based materials have shown favourable electrochemical stability; however, their application is often limited by poor ionic transport arising from highly ordered crystal structures.

Recent studies have identified $NaTaCl_6$ as a promising endmember in halide-based sodium solid electrolytes, presenting good electrochemical stability relative to other candidate solid electrolytes. However, in its ordered form sodium mobility is intrinsically low [1]. Advancements in ion transport properties have been made by introducing structural disorder, particularly via mixed-anion chemistries. One such example $NaTaOCl_4$ has exhibited significantly enhanced sodium mobility and promising catholyte behaviour [2, 3].

The mixed-anion series $NaTaO_xCl_{6-2x}$ ($x = 0.5,1$) provides a platform to assess the impact of oxygen incorporation on the structure and sodium ion transport in this material. To explore this, we employ a computational workflow combining density functional theory with ab initio random structure searching and machine-learned interatomic potentials to map the energetic landscape, identify low-energy configurations and dominant local motifs. Calculations show that oxygen incorporation disrupts long range ordering through linking previously isolated $TaCl_6$ octahedra via corner sharing oxygen. The coherence length of these units increases as oxygen content increases to $x = 1$, ultimately forming trans-linked $TaCl_4O_2$ chains. Molecular dynamics simulations are used to directly link these emerging motifs to enhanced sodium-ion transport, providing an atomistic explanation for the experimentally observed conductivity improvements in compositions such as $NaTaO_{0.5}Cl_5$.

Close collaboration with experimental partners allows comparison of computational predictions with diffraction, spectroscopic, and electrochemical measurements providing validation of the proposed structure–property relationships. Together, these results highlight the critical role of mixed-anion disorder in enabling fast-ion conduction in amorphous sodium oxyhalide electrolytes.

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Modelling Fast Lithium-Ion transport in the Halide Argyrodites

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Halide argyrodite systems, Li_6PS_5X (X = Cl, Br, I), are promising solid-state battery electrolytes, with structural disorder and aliovalent doping enabling a vast compositional design space and strongly influencing ionic conductivity. Recent computational studies have primarily relied on *ab initio* molecular dynamics and static calculations, limiting simulations to short timescales.

Here, machine-learned interatomic potentials are employed to extend molecular dynamics into the nanosecond regime across representative compositions of Li_6PS_5X and Zn-doped $Li_{6-2x}Zn_xPS_5X$. Access to these longer timescales enables analysis of correlated ion motion and collective transport processes beyond the reach of conventional ab initio simulations. The results across both undoped and Zn-substituted systems provide a mechanistic picture of ion transport and demonstrate how atomistic simulations can deconvolute the coupled effects of structural disorder, aliovalent substitution, and vacancy formation on ionic conductivity.

Predicting energy density retention in battery electrode materials using first-principles based modelling

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Lithium-rich manganese-based layered oxides are high energy density positive electrode (cathode) materials for next-generation lithium-ion batteries. Their high energy density is due to very high capacity from a combination of transition metal ion redox and oxygen redox. However, this high energy density is only partially reversible upon cycling, and they suffer a loss of energy density over time, predominantly through a loss of voltage at each cycle, termed 'voltage fade' [1,2,3,4]. To improve the performance of these cathode materials, the mechanisms behind the voltage fade must be prevented. Voltage fade has been linked to the observation of nanoscale voids appearing and growing in the material over time [1], but the mechanisms by which the voids form and grow are not well understood. The conventional atomistic modelling tool for understanding battery cathode materials, density functional theory (DFT), cannot be used to directly investigate nanoscale void formation and growth, because it requires system sizes that are too large to be computed with DFT.

In this work, we develop, a strategy, using DFT calculations, a cluster expansion, and Monte Carlo simulations, to simulate the void formation and growth over extended cycling. We apply this approach to a series of Li-rich Mn-based cathodes in the Li_2MnO_3–$LiMnO_2$ compositional space. We show that nanoscale voids in the bulk of the material arise from the formation of O_2 molecules within the bulk, and extensive transition metal migration to form TM-deficient regions, in a phase segregation mechanism. Over extended cycling, the voids coalesce, which is driven by minimisation of their surface energy, in a process akin to Oswald ripening. We show that the process of void coalescence depends on the initial structure of Li sites in the Mn-rich layer of the cathode, revealing that different initial structures can limit structural changes over long-term cycling. We show that voltage loss arises directly from structural changes as the voids grow, suggesting that by preventing void coalescence, voltage loss can be inhibited. Finally, by systematically mapping the voltage fade arising from different starting structures in the Li_2MnO_3–$LiMnO_2$ compositional space, we identify materials with the optimum structures and composition to minimise voltage fade and retain energy density over long-term cycling. Our study provides clear structural and compositional design rules for Li-rich cathode materials with improved performance.

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