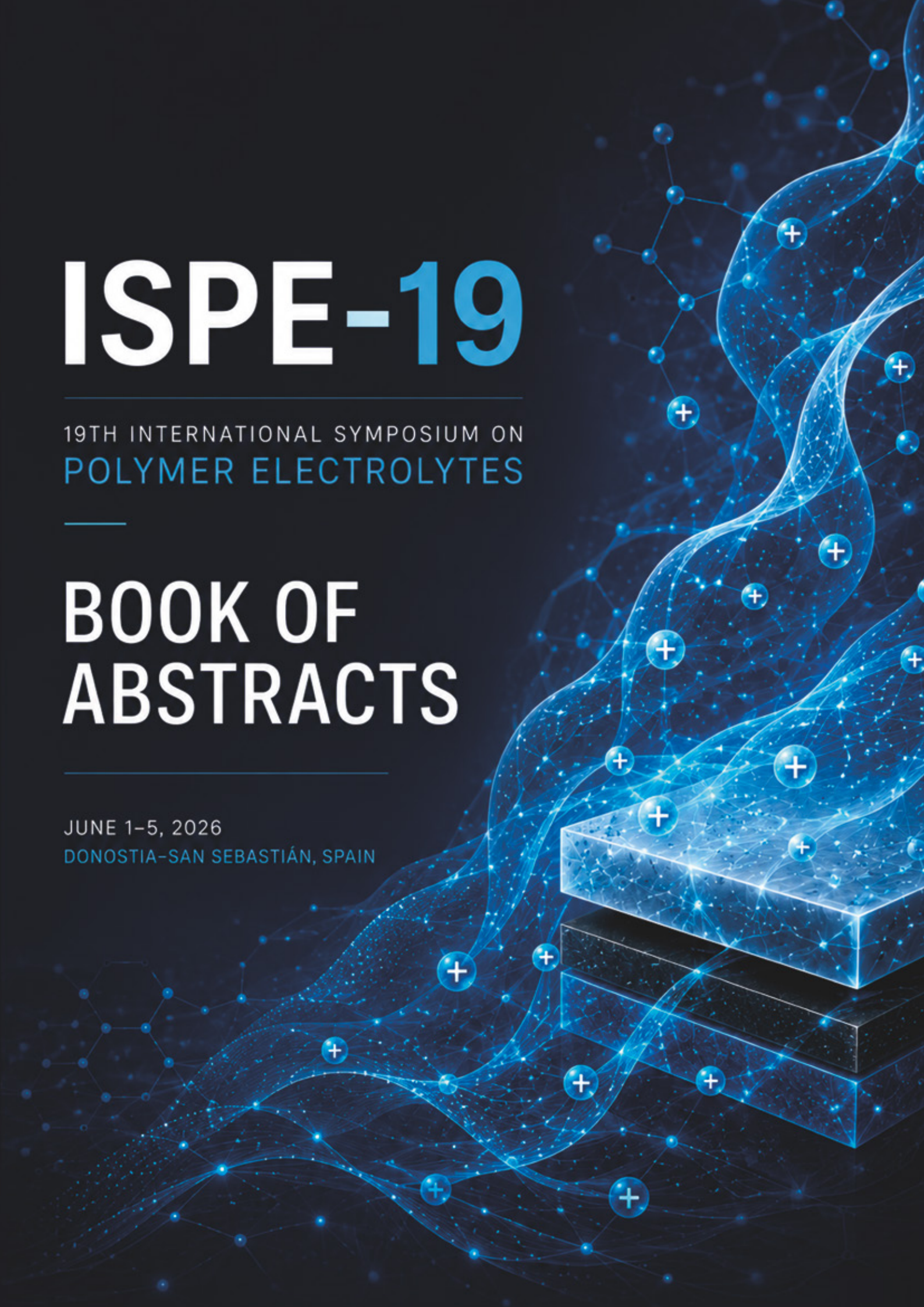
ISPE-19

19TH INTERNATIONAL SYMPOSIUM ON
POLYMER ELECTROLYTES

BOOK OF ABSTRACTS

JUNE 1–5, 2026
DONOSTIA–SAN SEBASTIÁN, SPAIN



SYMPOSIUM PROGRAM

Monday
(June 1st)

Monday, June 1st

Registration Open (8:30−)

Welcome/Opening Remarks (9:00−9:10)

Session 1, 9:00−10:30, Chairperson: Irune Villaluenga

OL 9:10-9:50 **Michel Armand, CIC Energigune, Spain**
New Solvents and Solutes for Polymer Electrolytes

PL1 9:50-10:30 **Nitash P. Balsara, UC Berkeley, USA**
Movement of Polymer Molecules under Electric Fields and Relevance to Rechargeable Batteries

Coffee Break (10:30−11:00)

Session 2, 11:00−12:55, Chairperson: Didier Devaux

K1 11:00-11:30 **Aimy Bazylak, University of Toronto, Canada**
Advanced imaging to examine the role of the membrane and ionomer in electrochemical energy devices

I1 11:30-11:50 **Kento Kimura, Tokyo University of Agriculture and Technology, Japan**
Understanding Structure-Dynamics-Functionality Relationships towards Developing Advanced Solid Polymer Electrolytes

I2 11:50-12:10 **Fabian Jeschull, Karlsruhe Institute of Technology, Germany**
Chemical and Electrochemical Degradation of a PEO- and Styrene-based Block Copolymer

O1 12:10-12:25 **Isak Bengtsson, Chalmers University of Technology, Sweden**
Modelling ionic liquid (electrolyte) conductivity using symbolic regression

O2 12:25-12:40 **Nellie Bowen, University of Notre Dame, Notre Dame, USA**
Investigating the Mechanism of Lithium-ion Transport and Conduction in Organic Charge-transfer Composite Polymer Electrolytes

O3 12:40-12:55 **Clara Aimar, LEPMI, France**
Mechanical failure mechanisms in solid electrolytes revealed by in situ synchrotron X-ray laminography

Lunch (13:00−14:30)

Session 3, 14:30−16:05, Chairperson: Maider Zarrabeitia

K2 14:30-15:00 **Monika Schönhoff, University of Münster, Germany**
Hydrodynamics at work: How transference numbers are governed by distinct species volume fluxes

I3 15:00-15:20 **Fangfang Chen, Deakin University, Australia**
Toward Automated Molecular Simulation for Physical-Based Screening of Polymer Electrolytes

O4 15:20-15:35 **Javier Carretero González, Institute of Polymer Science and Technology-CSIC, Spain**
Ion conducting polymeric membranes for energy storage systems

O5 15:35-15:50 **Matthieu Landa, University of Grenoble Alpes, France**
Design of carbonate-based single-ion polymer electrolyte for Lithium batteries

O6 15:50-16:05 **Emmanouil Glynos, University of Crete, Greece**
Macromolecular Design of Self-Healable and Mechanical Robust Solid Polymer Electrolytes

Coffee Break (16:05−16:30)

Session 4, 16:30−17:35, Chairperson: Andriy Kvasha

I4 16:30-16:50 **Trang N.T. Phan, Aix Marseille Univ, France**
Polymer electrolytes based on poly(ethylene oxide)/poly(trimethylene carbonate derivatives) block copolymers

O7	**Nicola Boaretto, CIC Energigune, Spain**
16:50-17:05	Towards high-energy density lithium metal batteries: from hybrid solid electrolytes to in-situ cross-linked gel polymer electrolytes
O8	**Anastasia A. Teck, Uppsala University, Sweden**
17:05-17:20	Interface Stability in High-Voltage LNMO - Lithium Metal Batteries with a Polyester Solid Polymer Electrolyte
O9	**Daphné Moté, University of Savoie Mont Blanc, France**
17:20-17:35	Mechanical Analysis of the Lithium/Electrolyte Interface via 180° Peel Tests for Solid-State Batteries

Poster Session 1, 17:35−19:30 (Miramar Palace-Ground Floor)

New Solvents and Solutes for Polymer Electrolytes

Maria Martinez Ibañez[1], Lorena Garcia[1], Paul Neumann[1], Leire Meabe[1], Alexander Santiago[1], Heng Zhang[1,2], Devaraj Shanmukaraj[1] & Michel Armand[1]

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For 50 years PEO has been the workhorse of polymer electrolytes. This polyether is however limited by its mediocre anodic oxidation window (3.9 V vs. Li^+:Li°) and low transference number (~ 0.25). Poly(esters) and poly(carbonates), developed under the leadership of the group at Uppsala University, have shown decent conductivities, improved anodic stability (4.5 V vs. Li^+:Li°) and transference number (~ 0.6). These polymers however contain the polar groups in the main chain, and the inevitable reaction of the ester linkage with Li° results in loss of M_w and of mechanical properties. Besides, the dissolved salts (LiTFSI…) can catalyse depolymerisation, resulting in artificially high conductivities due to the plasticisation of the polymer by the re-formed monomer.

To solve this problem, we designed poly(esters) with the ester functionality lateral to the main chain formed of sturdy C—C linkages. The first example was high M_w poly(vinyl butyrate) prepared simply from the reaction of poly(vinyl alcohol) (PVA) on butyric anhydride in the presence of DMAP. Conductivity studies with LiTFSI as a solute show excellent values, on a par with PEO and other poly(esters) with a $T_+ \sim 0.5$. Li°||Li° cells show extended cycling, a telltale of good interfacial stability, and Li°||$LiFePO_4$ or Li°||NMC batteries behave well as expected. The poly(carbonates) made from the esterification of PVA with an alkyl chloroformate give similar conductivity values and higher transport numbers.

In the realm of $T_+ \sim 1$ electrolytes, we have shown recently that the alloys formed with PEO and a cellulose-based polysalt combined excellent electrochemical properties and outstanding film-forming ability thanks to the rigidity of the polyoside polysalt. This suggested to replace PEO by the amorphous poly(ether) made from a Jeffamine grafted onto poly(ethylene)-maleic anhydride (PEMA) that by itself lacks mechanical properties. The resulting alloys have low temperature conductivities 2 order of magnitude higher than that of the PEO equivalent and keep excellent film-forming characteristics.

In the design of new salts, we have been active with the synthesis of sulfonylimides with on one side, a lipophilic moiety, with a general formula $Li^+[R^1R^2NSO_2N^{(-)}SO_2CF_3]$ Li[(Cn,n')TFSI]. Whatever the R values, the transport number ($T_+ \sim 0.45$) is higher than in TFSI, compensating the diminished total conductivity due to a less dissociated anion. The mains studies have focussed on $R^1 = R^2 = C_6H_{13}$, which induces low surface tension in the PEO-salt complexes and superior behaviour of the Li||LFP cells upon cycling. Interestingly, the salt does not corrode the aluminium current collectors. In an attempt to get rid of the PVDF used as a reinforcement of PEO in the commercial batteries, attempts were made to co-extrude PEO-Li C6,6-TFSI with poly(ethylene), the salt acting as a compatibilizer with interesting results.

Lastly, the non-PFAS salt Li^+ $[FSO_2N^{(-)}SO_2CF_2H]$ (LiFDFSI) was synthesized and shows improved conductivity vs. $[CF_2HSO_2N^{(-)}SO_2CF_3]$ in all polymer solvents, and keeps the non-corrosive nature imparted by the CF_2H moieties.

Advanced imaging to examine the role of the membrane and ionomer in electrochemical energy devices

Aimy Bazylak*

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For the first time in 2024, the global average temperature over a 12-month period rose to 1.5°C higher than the pre-industrial era. This failure to achieve the Paris Agreement temperature goal has led to present day global warming crises, namely extreme weather events. In past years, the impetus for clean energy was to avoid reaching this precipice. Now, clean energy is a critical pathway for adaptation – developing energy resiliency and security. Fuel cells and electrolyzers are necessary components of an energy resilient future, particularly their use in concert with renewable energy sources; electrolyzers can be used to separate hydrogen from water or convert CO_2 into carbon neutral fuels, while fuel cells can be used to produce on-demand electricity with zero local greenhouse gas emissions when fed hydrogen. Water management in fuel cells and electrolyzers plays a strong role in determining the electrochemical performance and even lifetimes of these devices, and for the most part, this topic has been focused on the porous carbon gas diffusion layer or titanium porous transport layer. In this talk, I will discuss how our study of water management has led us to take a deeper dive into the role of the membrane and ionomer within the membrane electrode assembly of the fuel cell and electrolyzer. In particular, operando imaging has been vital for examining various mechanisms, such as membrane hydration, membrane thinning, and ionomer redistribution. Addressing these challenging mechanisms will be paramount for developing the next generation of electrochemical energy conversion for commercial adoption.

Understanding Structure-Dynamics-Functionality Relationships towards Developing Advanced Solid Polymer Electrolytes

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We have previously reported that solid polymer electrolytes (SPEs) based on specific aliphatic polycarbonates, such as poly(ethylene carbonate) (PEC) and its derivatives, exhibit unique properties, including high cationic transference numbers and an increase in ionic conductivity with increasing salt concentration [1]. These trends, which significantly differ from those of conventional polyether electrolytes, are attributed to their salt-aggregated internal structure as revealed by spectroscopic studies [1b]. Raman spectroscopy and solid-state ^{7}Li MAS NMR studies confirmed that in PEC-based electrolytes, most anions exist as weakly interacting ion pairs or aggregates even in low-concentration regions, while the materials maintain their homogeneity as films or rubbery solids.

To further elucidate and utilize the unusual behavior, it is essential to analyze the relationships between structure, dynamics, and functionality. Our dielectric spectroscopy revealed that the mobility of polymer segments in PEC/imide-type salt electrolytes is enhanced with increasing salt concentration, in contrast to polyether (P(EO/PO)) electrolytes (Fig. 1) [2]. Since the coordination environment is less constrained than in conventional electrolytes, the plasticizing effect of bulky imide-based anions becomes dominant. A comparison of various salt species, such as LiTFSI and $LiClO_4$, confirmed that the excessive interaction between Li^+ and ClO_4^- leads to the formation of more ion-insulating aggregates [2]. These findings highlight that an optimized polymer-cation-anion combination is critical for performance. Rheological analysis also serves as a powerful technique to link macroscopic mechanical properties to microscopic dynamics [3]. Preliminary results demonstrate that the effect of salt addition on the viscoelastic relaxation of polymer chains varies significantly depending on the chemical structure of the polymer matrix. Furthermore, we have demonstrated that the highly aggregated ion structures in concentrated polymer electrolytes enhance electrochemical stability and prevent current collector corrosion [4]. Based on these fundamental understandings, we have developed salt-concentrated, crosslinked poly(carbonate/ether) electrolytes that offer both high battery compatibility and adequate mechanical stability [5,6]. The knowledge of structure-dynamics-functionality relationships will serve as the foundation for the development of advanced SPEs and solid-state energy storage devices.

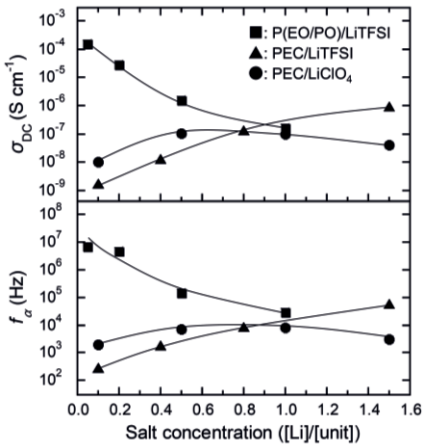

Fig.1. Salt concentration dependence of DC conductivity (σ_{DC}) and α relaxation frequency (f_α) for P(EO/PO)/LiTFSI, PEC/LiTFSI, and PEC/$LiClO_4$ at 40 °C.

References

[1] a) Y. Tominaga, *Polym. J.*, **2017**, 49, 291–299; b) K. Kimura, J. Motomatsu, Y. Tominaga, *J. Phys. Chem. C*, **2016**, 120, 12385–12391.
[2] K. Kimura, J. Motomatsu, Y. Tominaga, *J. Polym. Sci. B: Polym. Phys.*, **2016**, 54, 2442–2447.
[3] K. Kimura *et al.*, *ACS Appl. Polym. Mater.*, **2025**, 7, 4973–4981.
[4] K. Kimura, Y. Tominaga, *ChemElectroChem*, **2018**, 5, 4008–4014.
[5] N. Soontornnon, K. Kimura, Y. Tominaga, *J. Mater. Chem. A*, **2024**, 12, 20278–20287.
[6] N. Soontornnon, N. Takei, Y. Tominaga, K. Kimura, *J. Power Sources*, **2026**, 663, 238857.

Chemical and Electrochemical Degradation of a PEO- and Styrene-based Block Copolymer

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Block copolymer electrolytes (BPEs) based on styrene and PEO have shown improved chemical stability at the reactive electrode interphases of lithium- [1] and potassium-ion [2] batteries. In order to understand the origins of improvements, a combination of surface analytical techniques is required. However, ex-situ analysis has a tendency to alter the surface during sample preparation (e.g. when removing the electrolyte). Therefore, in-situ and operando techniques are a useful addition to acquire a more comprehensive picture of the interface chemistry [3]. Operando hard X-ray photoelectron spectroscopy (HAXPES) can be used to probe the surface layers of zero-access Ni|BPE|$K_2Fe[Fe(CN)_6]$ cells to track SEI formation at the early stages of potassium deposition. In complementary experiments chemical degradation can be induced by the deposition of metallic potassium onto a polymer electrolyte film [4].

In this presentation we will discuss the electrolyte degradation at the metallic potassium electrode and nickel current collectors of zero excess BPE-based potassium batteries. In this context focus will be placed on operando photoelectron spectroscopy studies using the extended capabilities of hard X-ray radiation at the BESSY synchrotron facility in Berlin. Because of the complexity of the spectra, chemical degradation studies were performed on the BPE electrolyte as well, which supports the analysis and interpretation of the data, but also illustrates differences in the degradation patterns between both processes (Figure 1). We will showcase how in operando techniques certain SEI components can be singled out by their charging behavior, concentration changes of mobile species can be tracked by this approach, and discuss the role of anion degradation in the early SEI formation stages.

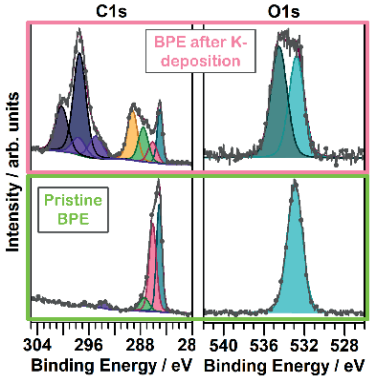

Figure 1. Changes at the BPE surface before and after potassium deposition

References

[1] A. J. Butzelaar, P. Röring, T. P. Mach, M. Hoffmann, F. Jeschull, M. Wilhelm, M. Winter, G. Brunklaus, P. Théato, *ACS Appl Mater Interfaces* **2021**, *13*, 39257–39270.
[2] A. D. Khudyshkina, A. J. Butzelaar, Y. Guo, M. Hoffmann, T. Bergfeldt, M. Schaller, S. Indris, M. Wilhelm, P. Théato, F. Jeschull, Electrochim Acta 2023, 454, 142421.
[3] F. Jeschull, *ChemElectroChem* **2024**, *11*, e202400254.
[4] E. K. W. Andersson, C. Sångeland, E. Berggren, F. O. L. Johansson, D. Kühn, A. Lindblad, J. Mindemark, M. Hahlin, J Mater Chem A Mater 2021, 9, 22462–22471.

Modelling ionic liquid (electrolyte) conductivity using symbolic regression

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The high thermal stabilities, wide electrochemical stability windows, and inherently good ionic conductivities of ionic liquids (ILs) make them promising alternatives to conventional solvents in battery electrolytes, which struggle with flammability, unwanted side reactions and even dissolution of electroactive material [1]. However, many properties of IL-based electrolytes remain poorly understood, particularly the underlying factors that govern ion transport.

As ILs constitute a vast chemical space, which could exceed 10^{18} unique compositions [2], modelling, and especially machine learning (ML) approaches, offers an efficient and scalable route to assess their properties [3]. Conventional ML methods, however, largely lack interpretability, as they map (large data) inputs to predicted properties via high-dimensional parameterized matrix product. To better understand ion transport in neat ILs, we have recently explored symbolic regression (SR), an ML method that finds analytical equations [4]. By using a general equation derived from free volume theory (FVT) as a template:

$$\sigma = \frac{f(x_1, x_2, \ldots)}{\sqrt{T}} \exp\left(-\frac{g(x_1, x_2, \ldots)}{h(T, x_1, x_2, \ldots)}\right),$$

we let SR find expressions for *f*, *g* and *h*, making the models consistent with an ion-hopping mechanism. Using molecular descriptors as inputs, we considered over 300 ILs and found models capturing ionic conductivity across wide temperature ranges.

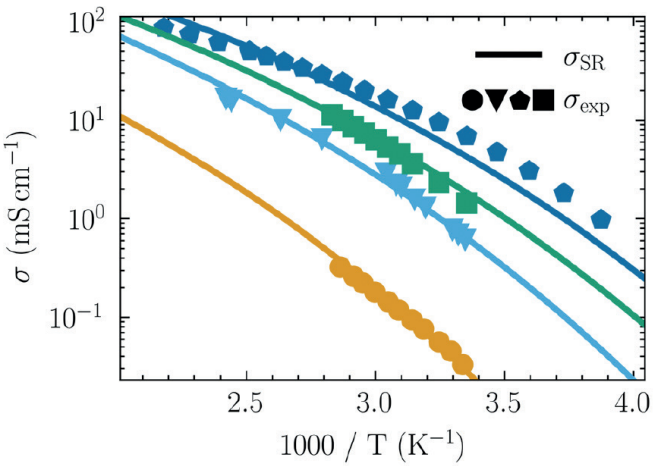

Figure 1: Measured ionic conductivities (markers) and SR model predictions (solid lines) for the ILs [P_{66614}][Hex] (orange), [N_{2228}][TFSI] (light blue), [BMIM][PF_6] (green), and [S_{221}][TFSI] (dark blue).

We are now extending our SR-approach to IL-based battery electrolytes, *i.e.* ILs doped with Li-, Na-, or K-salts. We are especially interested in trying to model the transport of the alkali cations, gaining molecular insights in how to design IL-based electrolytes with high cation transport numbers. One direction we are currently exploring is to combine SR with physics-informed neural networks (PINNs), which could help us extend existing theoretical frameworks with novel insights.

References

[1] M. Watanabe, et al., *Chem. Rev.* **117** (2017) 7190–7239.
[2] N.V. Plechkova, K.R. Seddon, *Chem. Soc. Rev.* **37** (2007) 123–150.
[3] I. Baskin, A. Epshtein, Y. Ein-Eli, *J. Mol. Liq.* **351** (2022) 118616.
[4] I. Bengtsson, P. Johansson, *Phys. Chem. Chem. Phys.* **28** (2026) 3841–3849.

Investigating the Mechanism of Lithium-ion Transport and Conduction in Organic Charge-transfer Composite Polymer Electrolytes

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Charge-transfer complexes (CTCs) have emerged as promising components in solid-state electrolytes and have been utilized in electrolyte materials with high room temperature ionic conductivities (2 x 10^{-3} S cm^{-1}) [1]. However, their implementation has been limited by salt dissociation via electrochemically unstable species such as water. Here, we report a charge-transfer composite polymer electrolyte (CTPE) consisting of poly(ethylene oxide) (PEO), lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), and the model CTC tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ), designed to enhance lithium-ion transport while maintaining electrochemical stability. Temperature-dependent broadband dielectric spectroscopy and electrochemical impedance spectroscopy were used to probe ion transport and relaxation dynamics. The CTPE exhibits high room-temperature ionic conductivity (order of 10^{-3} S cm^{-1}) along with signatures of mixed ionic-electronic conduction. Despite negligible changes in T_g (~-40 °C), the CTPE demonstrates significantly enhanced ionic conductivity, indicating partial decoupling of ion transport from polymer segmental dynamics. Fourier-transform infrared spectroscopy reveals increased lithium salt dissociation in the presence of TTF-TCNQ relative to neat PEO-LiTFSI, supporting a mechanism in which the charge-transfer complex alters the local coordination environment of lithium ions. These results demonstrate that CTC incorporation provides a viable strategy for tuning ion transport pathways in polymer electrolytes.

References

[1] Yang, L.; Schaefer, J. L. *ACS Energy Lett*, **8** (2023), 2426–2431.

Mechanical failure mechanisms in solid electrolytes revealed by in situ synchrotron X-ray laminography

Clara Aimar[1], Thomas Boulmier[1], Simon Bode[2], Thomas Tancogne-Dejean[3], Vincent Grolleau[3], Nicolas Lenoir[4], Alexander Rack[5], Lukas Helfen[6], Julio-Cesar Da Silva[7], Didier Devaux[1]

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Owing their excellent energy density and improved safety, lithium solid-state batteries (LiSSBs) are soon an alternative to conventional Li-ion batteries for electro-mobility. Despite their promise, the mechanical behavior of solid-state batteries remains elusive, mainly due to their complex multilayer architecture (Li metal anode, solid electrolyte (SE), composite cathode). In particular, failure mechanisms of the SE under compression are still not well defined, even though they are tightly coupled to performance degradation. Interfacial delamination directly compromises capacity [1], while electrolyte microfracturing can trigger dendrite nucleation and growth, ultimately leading to internal short circuits [2]. Furthermore, the effects of abuse loading (e.g., localized impacts) remain largely unexplored, obscuring the mechanical interplay between layers and the limits of device performance. Among relevant mechanical probes, indentation, by closely mimicking realistic loading scenarios, emerges as a particularly powerful approach to resolve the sequence of failure within the cell [2]. Accordingly, tuning the layers' properties at the cell level, under both operating and abuse conditions, is essential to investigates on the electro-mechanical response of the SE [3]. In this work, we investigate damage mechanisms in a hybrid halide SE based on Li_3InCl_6 and styrene-ethylene-butylene-styrene and assembled in various configurations of LiSSB pouch cells (e.g., Li symmetric cells vs. full LiSSBs) under mechanical indentation, using in situ 3D synchrotron imaging. Due to the large planar dimensions of pouch cells ($30 \times 30 \times 1.5$ mm³) relative to their micrometric internal architecture, achieving sufficient spatial resolution to capture microstructural evolution during loading remains highly challenging. To address this, a laminography setup was implemented at the ESRF beamline ID19, coupled with a custom-designed indentation device enabling controlled normal and abuse loading conditions, with a voxel size of 1.3 µm. During in situ experiments, mechanical (force) and electrochemical responses (open-circuit voltage and impedance) were continuously monitored. An image analysis workflow was developed to track the progressive radial and orthoradial crack propagation (see Fig. 1), and to quantify strain fields up to the onset of a short circuit. This approach provides the first quantitative insights into mechanical failure mechanisms in LiSSBs under both operating and abusive loading conditions. It further demonstrates the capability of high-resolution in situ synchrotron laminography to probe coupled structural, mechanical, and electrochemical phenomena in energy storage systems. By combining a large field of view with reduced imaging artefacts compared to conventional tomography, this method establishes a powerful framework for guiding the design of safer and more mechanically robust solid-state batteries.

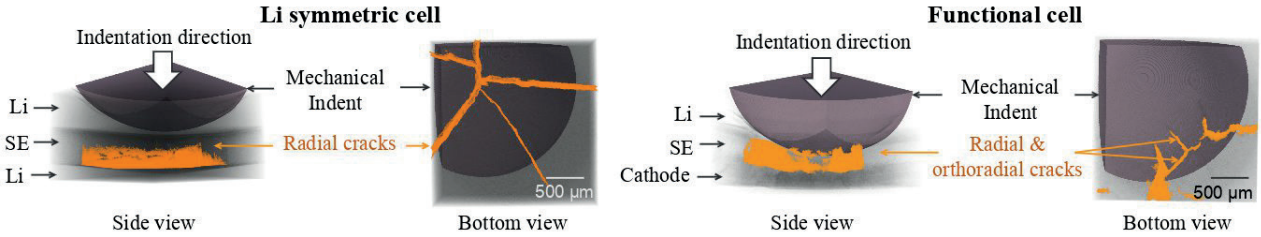

Figure 1. Image analysis of symmetric and functional cell wih a focus on crack propagation in the electrolyte.

References

[1] D. Devaux et al., *J. Electrochem. Soc.*, (2015), doi: 10.1149/2.0721507jes
[2] M. Kaliaperumal et al., *Materials*, (2021), doi: 10.3390/ma14195676
[3] L. Magnier et. al., (2021), Front. Energy Res., doi: 10.3389/fenrg.2021.657712

Hydrodynamics at work: How transference numbers are governed by distinct species volume fluxes

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The transference number T_+ of the active ion, such as Li^+, is one of the key parameters for describing the desired charge transport in an electrolyte. However, its commonly applied measurement using electrochemical methods is complicated by the challenge of controling interfacial resistances. Therefore, electrophoretic NMR (eNMR), which provides the drift velocities of the components of an electrolyte in an electric field, has emerged as an alternative that can overcome such problems. In this method, the transference number is directly calculated from experimentally determined ion mobilities. [1,2]

This lecture highlights the influence of various electrolyte components on the overall transport properties of an electrolyte. For example, in PEO-based salt-in-polymer electrolytes with different added co-solvents, drift velocities and associated volume fluxes of polymer and co-solvent are quantified. A strong influence of the volume of the co-solvent on Li ion transference numbers T_+ can be attributed to the fact that co-solvents exhibit a drift velocity in the electric field. The origin of this drift of neutral species is the need to compensate the volume flux of large anions towards the anode. This observation is in line with the relevance of the concept of volume conservation in concentrated electrolytes, as recently highlighted.[3] Furtheron, in salt-in-PEO electrolytes it is found that smaller or no co-solvents are beneficial in enhancing T_+, as in this case the chain drift velocity, and therefore the Li ion drift velocity, is enhanced, increasing T_+.[4]

Further studies dealing with a variation of the type of anion in electrolytes consisting of Li salt in PEO reveal that, due to an analogous mechanism, the volume flux of large anions can boost Li^+ transference.[5] We discuss the general predominance of anticorrelations between the electrolyte constituents, originating from volume conservation, as a governing concept which controls relative fluxes of electrolyte components. Consequently, transference numbers can be optimized by taking the species fluxes of all compoents and their interrelations into account. In conclusion, the experimental determination of species-selective flux data is generating in-depth knowledge for optimization of electrolyte compositions.

References:

[1] M. Gouverneur, J. Kopp, L. van Wüllen, M. Schönhoff, *Phys. Chem. Chem. Phys.* 2015, **17**(45), 30680-30686.
[2] M. P. Rosenwinkel, M. Schönhoff*, J. Elchem. Soc.* 2019, **166**(10), A1977-A1983.
[3] M. Lorenz, F. Kilchert, P. Nürnberg, M. Schammer, A. Latz, B. Horstmann, M. Schönhoff J. Phys. Chem. Lett. 2022 **13** (37), 8761-8767.
[4] S. Buyting, M. Schönhoff, *El.chim. Acta* 2025*,* **519***,* 145839.
[5] S. Buyting, M. Schönhoff, *ACS Appl. Polym. Mater.* 2025*, 7*(13), 8432-8444.

Toward Automated Molecular Simulation for Physical-Based Screening of Polymer Electrolytes

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Molecular simulation has become an increasingly powerful tool for understanding ion transport mechanisms and guiding the design of advanced polymer electrolytes for next-generation batteries. However, conventional molecular modelling workflows often require significant manual intervention in model construction, input preparation, and simulation setup, limiting their scalability for systematic materials exploration. The development of automated tools and modular simulation workflows is therefore essential to accelerate electrolyte discovery and enable effective physics-based computational screening.

In this work, we present our recent development toward automated molecular simulations of polymer electrolytes and their interfaces. We introduce an automated polymer generator tool, *PolyBuilder*, which constructs polymer simulation cells and generates molecular dynamics input files with minimal manual intervention, enabling ready-to-run simulations through a single command. Next, we develop modular workflows to investigate both bulk electrolyte properties and electrolyte–electrode interfacial chemistry. These workflows streamline key stages of molecular modelling, including structure preparation, system equilibration, and transport property analysis.

We demonstrate the capability of these tools through rapid prediction of ion transport behaviour and interfacial properties in polymer electrolyte systems. The integration of automated and standardized simulation workflows establishes a foundation for large-scale, physics-informed computational screening and the future development of AI-driven materials discovery frameworks.

Ion conducting polymeric membranes for energy storage systems

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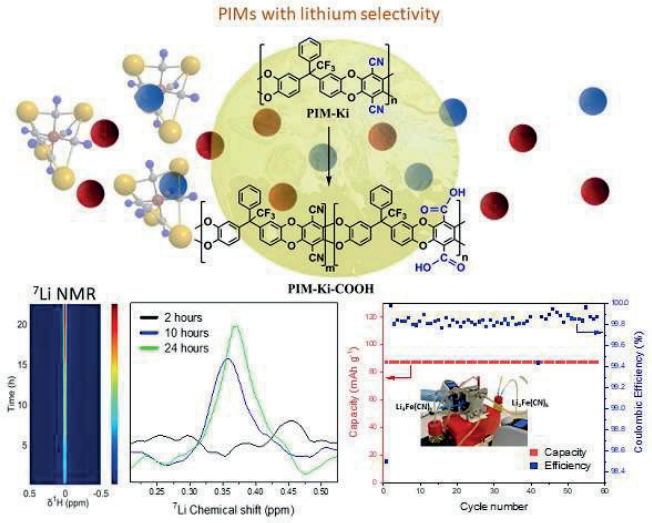
Ion exchange membranes (IEM) are key components of redox flow batteries (RFB). These systems that act as separators between electrolyte compartments need to ensure high-ion conductivity and selectivity while effectively minimizing crossover of the redox species. Because commercial perfluorinated membranes, such as Nafion®, face low selectivity and high costs, compromising both performance and economic feasibility would be needed. To overcome these limitations, current research is directed towards the development of materials with low or zero fluorine content that provide excellent balance and high-capacity retention rates in the battery [1]. In this communication we are going to show a new family of Polymers of intrinsic microporosity (PIMs) that we have synthesized and characterized [2]. Besides, they have been modified to adapt its structure to be used as selective membranes in redox flow batteries.



Different reaction conditions have been used to study how the insertion of COOH groups into the polymer affects the ion transport in aqueous media. FTIR, ^{1}H/^{13}C-NMR, N_2/CO_2 adsorption and ion permeation have been done. Moreover, in-line crossover and ^{7}Li/^{35}Cl diffusion NMR experiments have been done in those membranes exhibiting optimum properties, followed by flow half-cell studies to know the cyclability and capacity reached using $Li_3/_4Fe(CN)_6$ as redox species and the PIM membrane in aqueous media. A 65% modification of CN into COOH groups has been found to be the one with enhanced ion permeability and preference for Li ion. Moreover, the electrochemical tests showed low polarization, high-rate capability and capacity retention values of 99 % when cycled at 10 mA cm^{-2} for more than 50 cycles in a symmetric redox flow-cell. These materials can be potentially used as highly selective and conducting membranes in redox flow batteries but also in separation processes and as a protective interlayer in metal-ion batteries.

This research is supported by the Project TED2021-130372B-C43 funded by MCIN/AEI/10.13039/501100011033 and the European Union through the "NextGenerationEU"/PRTR action.

References

[1] McKeown, N. B. Science China Chemistry. Science in China Press August 1, 2017, pp. 1023–1032
[2] J. C. Martínez-López et al., "Dibenzodioxin-Based Polymers of Intrinsic Microporosity with Enhanced Transport Properties for Lithium Ions in Aqueous Media," Macromolecules, vol. 57, no. 19, pp. 9442–9456, Oct. 2024, doi:10.1021/acs.macromol.4c01243.

Design of carbonate-based single-ion polymer electrolyte for Lithium batteries

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Solid Polymer Electrolyte (SPE), composed of a polymer matrix in which a Li salt is dissolved, are promising candidates for safer lithium batteries due to their low flammability, good thermal stability and absence of leakage compared to conventional liquid electrolytes used in the Li-ion technology. [1] Additionally, their compatibility with lithium metal anodes makes them attractive for next generation batteries with increased energy density. [2]

Conventional SPEs based on poly(ethylene oxide) (PEO) suffer from limited lithium-ion mobility due to strong coordination of Li^+ within the polymer repeating units, as well as insufficient electrochemical stability at high voltage. Typically, PEO-based electrolytes exhibit a lithium transference number of around 0.2 and an electrochemical stability window below 4V, which restricts their use to low-voltage cathode materials such as LFP in full-cell configurations. [3]

Single-ion polymer electrolytes (SIPEs) have emerged as an effective strategy to enhance the lithium transference number by covalently grafting the anionic species onto the polymer backbone. This approach suppresses anion mobility, thereby reducing cell polarization and limiting lithium dendrite growth. [4] In parallel, amorphous polycarbonate matrices, especially poly(trimethylene carbonate), have been explored as alternatives to PEO, offering improved oxidative stability (up to 5V vs Li^+/Li) and favorable lithium transport properties ($T_{Li+} \approx 0.8$).[5]

In this presentation, we report the synthesis of a novel carbonate-based SIPE obtained via a post-polymerization functionalization strategy. In our approach, a functionalized carbonate copolymer is first synthesized. The pendant functional groups introduced along the polymer backbone are then reacted with a pre-synthesized functionalized lithium salt to form the SIPE. This strategy enables the covalent grafting of anionic moieties in close proximity to the carbonate groups responsible for lithium-ion transport. Such a molecular design is expected to promote efficient Li^+ conduction while avoiding phase segregation and diffusion limitations.

The grafting rate can be tuned through the molar ratio of functionalized comonomer, although it is also limited by the reactivity between the pendant functional groups and the lithium salt. Particular attention is given to the influence of the grafting rate on the physicochemical and electrochemical properties of the electrolyte. The performance of these materials is compared to that of conventional polycarbonate-based electrolytes to assess the benefits of this strategy. By tuning the density of grafted ionic groups, we investigate the trade-off between ionic density and segmental mobility, and its impact on ionic conductivity and transport properties.

References

[1] Y. An, X. Han, Y. Liu, A. Azhar, J. Na, A. K. Nanjundan, S. Wang, J. Yu, Y. Yamauchi, Small, 18 (2022) 2103617
[2] Y.-K. Sun, ACS Energy Lett. 5 (2020) 3221−3223.
[3] J. Mindemark, M. J. Lacey, T. Bowden, D. Brandell, Prog. Polym. Sci. 81(2018), 114–143.
[4] K.M. Diederichsen, E. J. McShane, B.D. McCloskey, ACS Energy Lett. 2 (2017) 2563–257
[5] B. Sun, J. Mindemark, E. V. Morozov, L. T. Costa, M. Bergman, P. Johansson, Y. Fang, I. Furó, D. Brandell, Phys. Chem. Chem. Phys. 18 (2016) 9504–9513

Macromolecular Design of Self-Healable and Mechanical Robust Solid Polymer Electrolytes

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The development of solid polymer electrolytes with simultaneously high ionic conductivity, mechanical robustness, and damage tolerance remains a major challenge for next-generation electrochemical energy storage systems. In this work, a new materials design strategy based on architecturally controlled mikto-arm star polymers is introduced to create self-healing polymer electrolytes capable of autonomous recovery of mechanical properties while maintaining efficient ion transport.

Mikto-arm star polymers composed of rigid poly(methyl methacrylate) (PMMA) and flexible poly(lauryl methacrylate) (PLMA) arms were synthesized and shown to form densely packed, nanoparticle-like assemblies exhibiting solid-like behavior and spontaneous recovery after yielding, without relying on dynamic covalent bonds or supramolecular interactions. These self-healable nanoparticles were subsequently used as multifunctional additives in fast-conducting low molecular weight PEO/LiTFSI electrolytes in order to create mechanically reinforced yet conductive nanocomposite electrolytes.

Systematic variation of nanoparticle loading and arm composition revealed that the balance between hard–soft architectural contrast and miscibility with the PEO phase governs the resulting morphology, segmental dynamics, ionic conductivity, and recovery efficiency. SAXS/WAXS and DSC measurements indicate the formation of semicontinuous nanostructures in which the PEO-rich phase provides ion-transport pathways, while the mikto-arm nanoparticles form a percolated, reversible load-bearing network. Remarkably, complete autonomous recovery of mechanical properties is achieved at high nanoparticle concentrations, demonstrating that self-healing in these systems arises from reversible particle–particle interactions rather than polymer chain diffusion.

These results establish mikto-arm star nanoparticles as a versatile platform for the design of self-healing solid polymer electrolytes and highlight molecular architecture as a key parameter for future energy-storage materials.

Polymer electrolytes based on poly(ethylene oxide)/poly(trimethylene carbonate derivatives) block copolymers

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Solid-state batteries (SSBs) using lithium metal anodes represent a transformative advancement in energy storage technology. They overcome the limitation of liquid electrolytes by offering higher energy density, enhanced safety, longer cycle life and a wide operational temperature range. Having all of these advantages, the quest for superior solid-state electrolytes (SSEs) has been ongoing for decades. Among SSEs, solid polymer electrolytes (SPEs) have gained popularity due to their excellent processability, low mass density and thickness required. However, designing a SPE that combines high ionic conductivity, a wide electrochemical stability window (ESW), a high transference number, good interface compatibility, and a high dielectric constant along with superior mechanical and thermal properties remains a significant challenge.

The classic poly(ethylene oxide) (PEO)-based Li electrolytes exhibit low transition temperature (T_g) and good ionic conductivities above the PEO melting point. However, their ESW is below 4.0 V *vs*. Li^+/Li, making them unsuitable for the high-voltage cathode materials in the next generation high energy batteries. In contrast, aliphatic polycarbonates have emerged as promising ion-conductive matrices, potentially achieving a high Li^+ transference number (≈ 0.6 – 0.8) and an ESW of up to 5V. Poly(ethylene carbonate) electrolytes with short ethylene oxide side chains have demonstrated that longer side chains improve ionic conductivity [1]. Additionally, copolymer electrolytes based on random copolymers of ethylene carbonate (EC) and ethylene oxide (EO) with varying EC/EO ratios exhibited higher conductivity and lower T_g compared to the EC homopolymer system [2]. Incorporating EO units into the main chains of polycarbonates also enhanced ionic conductivity, making them suitable for ambient temperature batteries [3]. Interpenetrating blends of amorphous PEO and poly(trimethylene carbonate) (PTMC) matrices doped with LiTFSI have shown significant improvement in ionic conductivity [4].

In this study, block copolymers composed of PEO and poly(trimethylene carbonate derivatives) with varying EO/carbonate ratios were synthesized via ring-opening polymerization (Figure 1). The influence of the EO/carbonate ratios and the nature of the TMC derivatives on the physicochemical and electrochemical properties of the resulting electrolytes were systematically investigated.

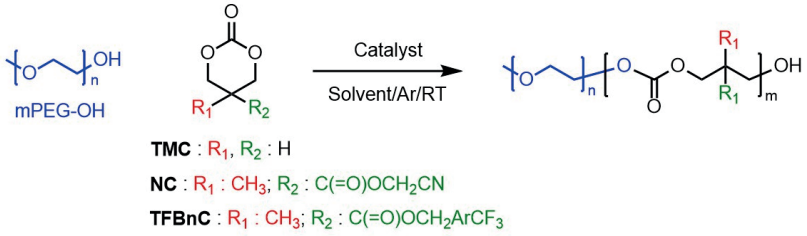

Figure 1. Ring-opening polymerization of trimethyleen carbonate derivatives using PEO as initiator.

References

[1] T. Morioka, K. Ota, Y. Tominaga, *Polymer* **84** (2016) 21–26
[2] Y. Tominaga, K. Nakano, T. Morioka, *Electrochim. Acta* **312** (2019) 342-348
[3] W. Hea, Z. Cui, X. Liua, Y. Cui, J. Chai, X. Zhou, Z. Liu, G. Cui ; *Electrochim. Acta* **225** (2017) 151–159
[4] L.C. Rodrigues, M.M. Silva , M.J. Smith, *Electrochim. Acta* **86** (2012) 339– 345

Towards high-energy density lithium metal batteries: from hybrid solid electrolytes to in-situ cross-linked gel polymer electrolytes

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Compared with conventional Li-ion batteries, lithium metal batteries offer substantially higher energy density and specific energy. However, safety and stability concerns associated with commercial liquid electrolytes continue to impede their widespread deployment. Solid-state electrolytes, including polymer electrolytes, provide a safer alternative, but their relatively low ionic conductivity limits electrochemical performance, particularly with respect to operating temperature, C-rate capability, and achievable cathode areal loading. As a result, the practical applicability of fully solid-state battery technologies remains under debate [1].

In this contribution, we report several strategies that we have explored to enable practical, scalable, high-energy lithium metal batteries employing Ni-rich layered oxide cathodes for automotive applications. Separator-supported hybrid solid electrolytes, plasticized with PEGDME, delivered a specific energy of up to 358 Wh kg^{-1}, exceeding that of state-of-the-art Li-ion batteries, albeit at an operating temperature of 60 °C and at low C-rate of C/20 [2,3]. Transitioning to gel polymer electrolytes (GPEs) plasticized with ethylene carbonate significantly enhanced ionic conductivity, enabling room-temperature operation (25 °C) and higher C-rate capability. Also in this case, electrolyte support on a microporous separator was essential to sustain cycling at high areal capacities of 2.5 mAh cm^{-2} [4]. Nevertheless, the scalability of separator-supported GPEs proved challenging and poorly compatible with existing coating lines.

Considering these limitations, we eventually shifted our attention to in-situ polymerized gel polymer electrolytes, which are readily scalable and compatible with current battery manufacturing lines. By combining the in-situ polymerization approach with the localized high concentration electrolyte concept, we demonstrate promising performance, with nearly 200 stable cycles at room temperature and a C-rate of C/3 in NMC-811||Li cells operating at an areal capacity of 3 mAh cm^{-2}.

In summary, our results suggest that achieving practical high-energy lithium metal batteries for automotive application, with competitive performance, will require moving beyond fully solid polymer electrolytes. Semi-solid electrolyte systems represent an effective pathway to combining safety, performance, and scalability, offering a realistic route to liquid-electrolyte-like performance while preserving manufacturing compatibility.

References

[1] M.A. Cabañero Martínez, N. Boaretto, A.J. Naylor, F. Alcaide, G.D. Salian, F. Palombarini, E. Ayerbe, M. Borras, M. Casas-Cabanas, *Adv. Energy Mater.,* **12** (2022) 2201264.

[2] N. Boaretto, H. Perez-Furundarena, L. Meabe, S. Lindberg, I. Combarro, A. Kvasha, E. Esen, D. Del Olmo, E. Ayerbe, H. Demers, A. Guerfi, R. Vincent, J. Cognard, S. Genies, M. Lechartier, L. Daniel, A. Mery, L. Castro, A. Guéguen, M. Martínez-Ibañez, *J. Power Sources,* **653** (2025) 237671.

[3] N. Boaretto, L. Meabe, S. Lindberg, H. Perez-Furundarena, I. Aldalur, E. Lobato, F. Bonilla, I. Combarro, A. Gutiérrez-Pardo, A. Kvasha, M. Lechartier, R. Vincent, J. Cognard, S. Genies, L. Daniel, M. Martinez-Ibañez, *Adv. Funct. Mater.,* **34** (2024) 2404564.

[4] N. Boaretto, O. Garcia-Calvo, M. Cobos, A. Fernandez de Añastro, M. Diez Viera, M. Al Sammarraie Shakir, S. Lindberg, R. Cid Barreno, G. Godillot, L.O. Jøsang, A. Kvasha, M. Martínez-Ibañez, *Energy Mater.,* **5** (2025) 500040.

Interface Stability in High-Voltage LNMO - Lithium Metal Batteries with a Polyester Solid Polymer Electrolyte

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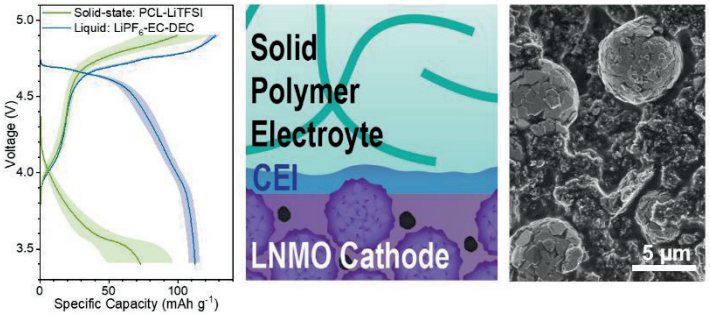
Solid polymer electrolytes (SPEs) are a compelling material for high-energy lithium metal batteries. Compared to conventional liquid electrolytes, SPEs offer improved safety, sustainability and cost, in addition to the ability to suppress lithium dendrite growth. Furthermore, polymer-based electrolytes exhibit superior mechanical properties to inorganic solid-state electrolytes to enable good interfacial contact and withstand strain. Pairing a lithium metal anode with a high-voltage cathode promises a high energy density battery, but to date development has focused most significantly on lithium nickel manganese cobalt oxide (NMC) and less progress has been made on the use of lithium nickel manganese oxide ($LiNi_{0.5}Mn_{1.5}O_4$; LNMO) cathodes, free of environmentally and ethically harmful cobalt. However, the oxidative stability of many polymers has limited their application and few suitable materials have been reported to enable cycling above 5 V vs Li/Li^+. Commonly utilized polyethylene oxide (PEO) is well understood to be unstable at high voltages, and so far only few PVdF-based electrolytes have demonstrated cycling with LNMO,[1] the majority of which have been implemented as gel electrolytes.[2] Consequently, it is highly relevant to explore further polymer electrolyte materials in fully solid-state systems and to characterize the challenging hidden electrode – electrolyte interface to understand their stability with LNMO.

Polycaprolactone (PCL), a biodegradable polyester, is a promising material for application with LNMO due to its high oxidative stability (up to 5.4 V vs Li/Li^+ with LiTFSI)[3] and low glass transition temperature (~ - 60 °C). Here, PCL-based SPEs and composite polymer electrolytes (CPEs) were studied in full cell LNMO – Li metal batteries and have successfully achieved charging to > 5 V. However, significant instability identified in subsequent cycling has motivated a detailed investigation of the system's interfaces. With focus on the cathode – electrolyte interphase, a combination of electrochemical, spectroscopy, and microscopy techniques has been employed to evaluate battery performance and to probe the buried interface. In particular, a relatively straight-forward sample preparation method was developed to enable lab-scale and synchrotron characterization of the polymer - LNMO interface by X-ray photoelectron spectroscopy (XPS) and hard X-ray photoelectron spectroscopy (HAXPES) to different depths within the interfacial region.

Aiming to expand the application of SPEs for high-energy LNMO – Li metal batteries, the stability of the hidden cathode-electrolyte interface was explored to reveal a combination of polymer degradation, mechanical contact, and active material dissolution controlling the performance and lifetime of the high-voltage battery.



References

[1] Y. Hou *et al.*, *Angew Chem Int Ed,* **64**, 29 (2025) e202505147.
[2] J. Yu *et al.*, *J. Mater. Chem. A,* **10**, 24 (2022) 12842–12855.
[3] C. F. N. Marchiori, R. P. Carvalho, M. Ebadi, D. Brandell, and C. M. Araujo, *Chem. Mater.,* **32**, 17 (2020) 7237–7246.

Mechanical Analysis of the Lithium/Electrolyte Interface via 180° Peel Tests for Solid-State Batteries

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The European SEATBELT project[1] aims to develop a solid-state electrolyte, moving from a polymer electrolyte to a new configuration of hybrid electrolyte. One critical issue using a solid-state electrolyte is the contact failure with the anode interface which can have a significant impact of the electrochemical Coulombic efficiency and on the mechanical stability of the battery[2]. Therefore, mechanical studies of this interface are key to having a better understanding of the interface properties and to link the mechanical behaviour with physico-chemical characterisations.

A methodology has been recently developed to do so*. It consists of a 180° peel test[3] enabling the quantification of the mechanical adhesion level between a lithium electrode and a solid polymer electrolyte based on poly(ethylene oxide) (PEO-LiTFSI) through the determination of a critical peeling force. It was found that the annealing time at 80 °C under a 1 bar pressure affected the interfacial adhesion properties, highlighting two adhesive regimes before and after 6 h of annealing as presented in **Figure 1**. To understand the origin of this variation, there is a need to use other characterization methods. Firstly, X-ray photoelectron spectroscopy (XPS) was used to analyze the lithium metal anode surface and detect (or not) presence of PEO-LiTFSI. Then, scanning electron microscopy (SEM) was used to visualize any evolution of the materials morphologies after peel tests. This revealed the replication of the lithium grain boundaries on the PEO-based electrolyte surface and how they could be the location of the physico-chemical reactions inducing a better adhesion[4]. Variations in peel strength as a function of annealing time are also linked with the anode/electrolyte interface resistance as determined by electrochemical impedance spectroscopy (EIS). Two regimes were also observed before and after 6 h annealing time which can be link to the development of the passivation layer at the interface[5]. This peel test methodology is therefore a reliable and robust tool for characterizing interfaces and their evolution over time under different parameters.

The aim of this study is to revisit this recent methodology to characterize new materials such as a lithium indium chlorine (Li_3InCl_6) inorganic dispersed in an organic polyethylene (PE) matrix, forming thus a hybrid LIC-PE electrolyte material. Its intrinsic properties are firstly characterized (tensile properties, topography by confocal microscopy, …). Then the same annealing parameters (80°C under a 1 bar pressure) were applied on LIC-PE and Li-metal which led to peel tests with very different results from PEO-LiTFSI polymer electrolyte.

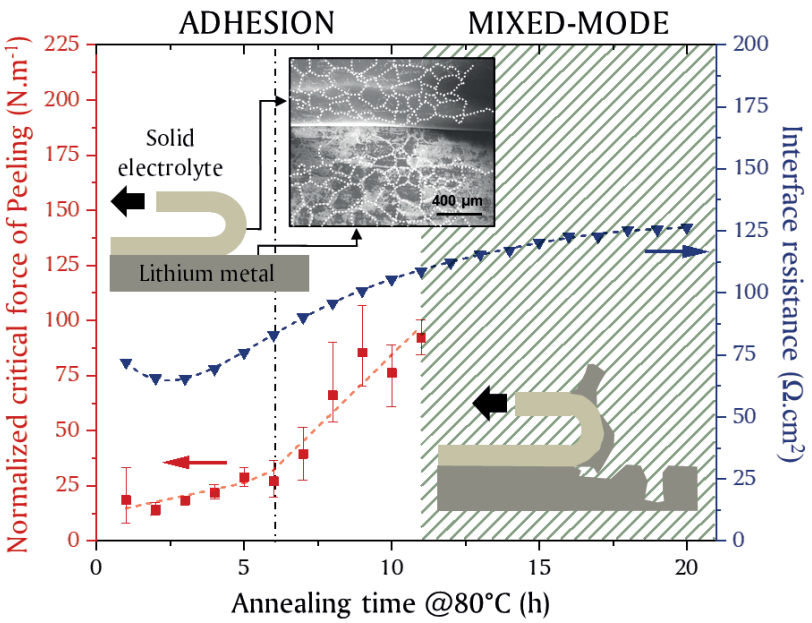

Figure 1. Normalized critical force of peeling ($N.m^{-1}$) and Interface resistance ($\Omega.cm^2$) of the lithium metal/PEO-LiTFSI interface as a function of annealing time (@80°C under a 1 bar pressure) using a 180° peel test methodology

References

[1] « SEATBELT-project-eu ». Consulté le: 31 mars 2026. [En ligne]. Disponible sur: https://seatbelt-project.eu/

[2] S. Kalnaus, N. J. Dudney, A. S. Westover, E. Herbert, et S. Hackney, « Solid-state batteries: The critical role of mechanics », *Science*, vol. 381, nº 6664, p. eabg5998, sept. 2023, doi: 10.1126/science.abg5998.
* D. Molé et al., « Mechanical Analysis of the Lithium/Electrolyte Interface via 180 Peel Tests for Solid-State Batteries », ACS Appl. Energy Materials, 2026, accepted (in press), https://doi.org/10.1021/acsaem.5c04055

[3] M. D. Bartlett, S. W. Case, A. J. Kinloch, et D. A. Dillard, « Peel tests for quantifying adhesion and toughness: A review », *Progress in Materials Science*, vol. 137, p. 101086, août 2023, doi: 10.1016/j.pmatsci.2023.101086.

[4] E. Milan et M. Pasta, « The role of grain boundaries in solid-state Li-metal batteries », *Mater. Futures*, vol. 2, nº 1, p. 013501, déc. 2022, doi: 10.1088/2752-5724/aca703.

[5] S.-K. Otto *et al.*, « Storage of Lithium Metal: The Role of the Native Passivation Layer for the Anode Interface Resistance in Solid State Batteries », *ACS Appl. Energy Mater.*, vol. 4, nº 11, p. 12798-12807, nov. 2021, doi: 10.1021/acsaem.1c02481.

Poster Session 1

(Monday, June 1st)

Poster Session 1, Monday, June 1st, 17:35-19:30

No	Presenter	Poster Title
P1-1	**Lixin Qiao**	Asymmetric Cyano-Functionalized Lithium Sulfonimide Salt Facilitates Fast Lithium-ion Transport and Enables High-Voltage All-Solid-State Lithium Metal Polymer Batteries
P1-2	**Huanrui Zhang**	Functional polymer electrolytes enable high-safety and high-energy-density lithium batteries
P1-3	**Shanmu Dong**	Regulating intermolecular bonding towards rechargeable Li-SOCl 2 battery
P1-4	**Poongodi Ayyanusamy**	Rapid Synthesis of Hybrid Li3InCl6 Halide Electrolytes: An Operando WAXS/EIS Study of Phase Formation
P1-5	**Lorenzo Pirollo**	In Situ photopolymerized ionogels for safe high-Voltage lithium metal batteries
P1-6	**Federica Tidona**	Wet vs Dry Processing of PEO and Single Ion Conductive Polymer Electrolytes for Lithium Metal Batteries
P1-7	**Nana Terasoba**	Conductivity and Stability Prediction of Anion Exchange Membranes Using Small Data Machine Learning
P1-8	**Satendra Kumar**	Correlating Deposition Kinetics and Morphology to Optimize Binder-free Electroactive Interfaces
P1-9	**Lukas Metzler**	Synthesis and Characterization of Ladder Ionomers via CANAL Polymerization for AEM Fuel Cell and Water Electrolyzer Applications
P1-10	**Leire Unanue**	Nanostructured ion-conducting materials based on polymer nanoparticles and diblock copolymers
P1-11	**Toru Ishikawa**	Enhancement of Ionic Conductivity in Solid Polymer Electrolytes via Incorporation of a Pyrrolidinium-Based Ionic Plastic Crystal
P1-12	**Qaphelani Ngulube**	4D imaging of SPE-based all-solid-state lithium batteries
P1-13	**Shinji Kondou**	Computational Co-Design of Poly(ionic liquid)s-in-Salt Electrolytes Toward Enhanced Alkali Metal Ion Transport
P1-14	**Killian Delvalle**	Molten Salt Electrolytes – A Combined Experimental and Computational Approach
P1-15	**Kota Ogino**	Ion Transport Properties of Composite Materials Based on Deeply Supercooled Li-Salt (Li-DSS)
P1-16	**Fukao Yuma**	Ga-based Liquid Metal−Ionic Liquid Composite Gels for Stretchable Li-ion Battery Electrodes
P1-17	**Hisamune Ayaka**	Thermal and Electrochemical Properties of Oligoether-Functionalized Asymmetric Lithium Imide Salts for Molten Salt Electrolytes
P1-18	**Iguchi Sayaka**	Poly(Solvate Ionic Liquids) Electrolytes Based on Polymeric Lithium Salts

P1-19	**Wansu Bae**	Sulfur-Substituted Polymer Electrolytes with Weak Li–S Coordination for Fast Lithium-Ion Transport
P1-20	**Doyul Lee**	In Situ Polymerized Sulfur–Containing Allyl Thiirane Electrolytes Initiated by LiFSI for High Conductivity and Interface Stability
P1-21	**Aya Kato**	Electrochemical CO_2 Absorption Behavior Properties of Anthraquinone Bearing Ionic Liquid Moiety
P1-22	**Marissa Cavallotti**	Interfacial Behaviour of Li-Metal Anodes and Polymer Electrolytes in Solid-State Cell Architectures
P1-23	**Nadine Tänzer**	Impact of Particle Sizes in Hybrid-Polymer Electrolytes on Lithium Inventory Homogeneity in Lithium Metal Batteries
P1-24	**Felix Nordgren**	Topology-dependent transport mechanism(s) in lithium-based molten salt electrolytes
P1-25	**Mina Nakazawa**	New Design Principle for Proton Conductive Organic Electrolytes Based on Activation of Surface Proton Hopping Conduction Mechanism
P1-26	**Deborah Feld**	Dynamics & Mechanical Properties of Double-Network Ionogels
P1-27	**Leire Meabe**	Designing Stable 5 V Cathode Compositions for Safe Solid-State Lithium Metal Batteries
P1-28	**Iker Varona**	Optimization of Inorganic-Polymer Hybrid Electrolytes by Rheology-Guided Extrusion for Solid-State Batteries
P1-29	**Saori Yamaguchi**	Temperature dependence of molecular dynamics and ionic conductivity in hydrated ionic liquids showing LCST-type phase transition behavior
P1-30	**Takahiro Ichikawa**	Gyroid Nanostructured Proton Conductive Polymer Membranes Transporting Proton Through Surface Hopping Conduction Mechanism
P1-31	**Clément Pechberty**	Safer sodium-ion batteries by non-flammable deep eutectic electrolytes
P1-32	**Valentina Colli**	Interpenetrating Gel Polymer Electrolytes: Effects of PEO Structural Design

Asymmetric Cyano-Functionalized Lithium Sulfonimide Salt Facilitates Fast Lithium-ion Transport and Enables High-Voltage All-Solid-State Lithium Metal Polymer Batteries

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All-solid-state polymer electrolytes (ASSPEs) have attracted considerable interest from both academic and industrial settings due to their remarkable flexibility, excellent processability and enhanced safety.[1] However, the widespread application of conventional lithium bis(trifluoromethanesulfonyl)imide/poly(ethylene oxide) (LiTFSI/PEO)-based ASSPEs is limited by sluggish Li^+ transport kinetics and insufficient anodic stability.[1] In this presentation, we introduce an asymmetric cyano-functionalized lithium salt, lithium cyano(trifluoromethanesulfonyl)imide (LiCTFSI), which effectively modulates the Li^+ solvation structure and interfacial chemistry to enable rapid Li^+ conduction and enhance compatibility between PEO and high-voltage cathodes. The incorporation of cyano groups in the anion not only weakens the Li^+–EO interaction and promotes Li^+–anion coordination, establishing a coupled Li^+–anion transport pathway, but also suppresses the catalytical decomposition of PEO matrices by aggressive cathode materials. Furthermore, the reinforced Li^+–anion solvation structure promotes the formation of compact and conductive solid electrolyte interphase enriched with LiF and Li_3N, significantly improving interfacial stability and cell performance. The Li/$LiCoO_2$ cells with LiCTFSI-based ASSPEs without any fillers/plasticizers could operate >100 cycles with >80% capacity retention under high-voltage of 4.3 V vs. Li/Li^+ (Figure 1). This work opens up a new avenue to simultaneously achieve fast Li^+ transport and excellent compatibility of PEO-based ASSPEs with high-voltage cathodes.

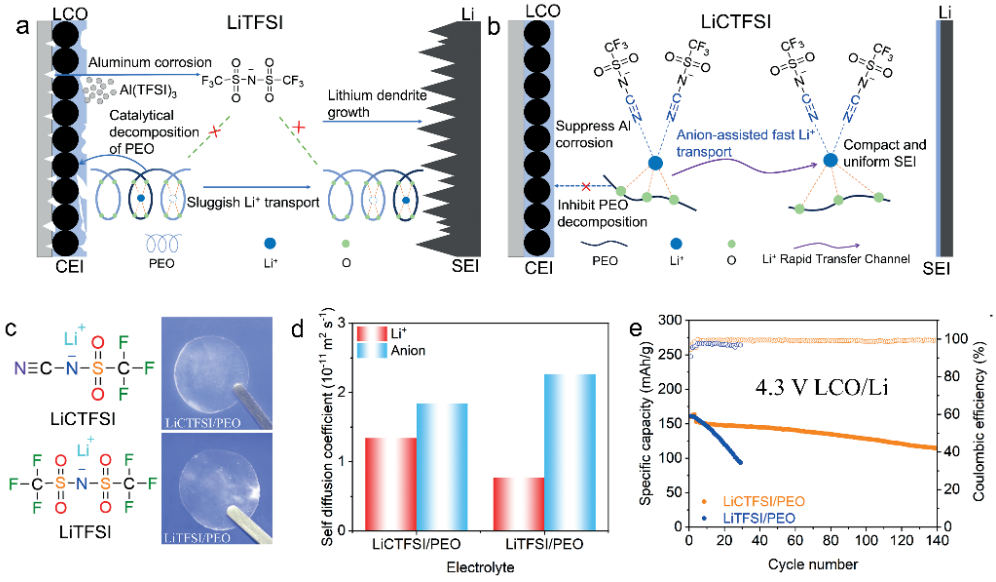

Figure 1. Schematic illustration of the roles of (a) LiTFSI and (b) LiCTFSI. (c) Structure of lithium salts and the corresponding digital images of the membranes. (d) Summary of the Li^+ and anion diffusion coefficients obtained from PFG-NMR. (e) Long-term cycling performance of the LCO/Li cells.

References

[1] Qiao, L.; Rodriguez Peña, S.; Martínez-Ibañez, M.; Santiago, A.; Aldalur, I.; Lobato, E.; Sanchez-Diez, E.; Zhang, Y.; Manzano, H.; Zhu, H.; Forsyth, M.; Armand, M.; Carrasco, J.; Zhang, H., *J. Am. Chem. Soc.* 2022, 144, 9806–9816.

Functional polymer electrolytes enable high-safety and high-energy-density lithium batteries

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Strong demand for consumer electronics and electric vehicles has propelled the development of high-energy-density lithium batteries operating at high voltages. However, increasing the operating voltage accelerates capacity fading and exacerbates safety risks in lithium batteries using conventional liquid electrolytes. To tackle these challenges, the development of functional polymer electrolytes represents one of the most effective strategies. Recently, we have fabricated a series of in-situ formed functional polymer electrolytes [1-5], which significantly enhance both the electrochemical performance and thermal safety of high-voltage lithium-ion batteries. For instance, we first developed thermal-shutdown type in-situ polymerized electrolytes based on the nucleophilic addition between carbamate and isocyanate groups, or the re-polymerization of residual acrylate moieties within the polymer matrix under thermal abuse conditions (Figure 1a–b). In 4.3 and 4.4 V-class $LiCoO_2$ cathode-based full batteries, the as-developed electrolyte can deliver superior long-term cycling stability compared with liquid electrolytes (Figure 1c), owing to the formation of more compatible electrode/electrolyte interphases. Furthermore, improved battery thermal safety is achieved through the thermally induced crosslinking reaction of the polymer matrix, which effectively suppresses electrode–anode chemical crosstalk and enables battery thermal shutdown under thermal abuse (Figure 1d).

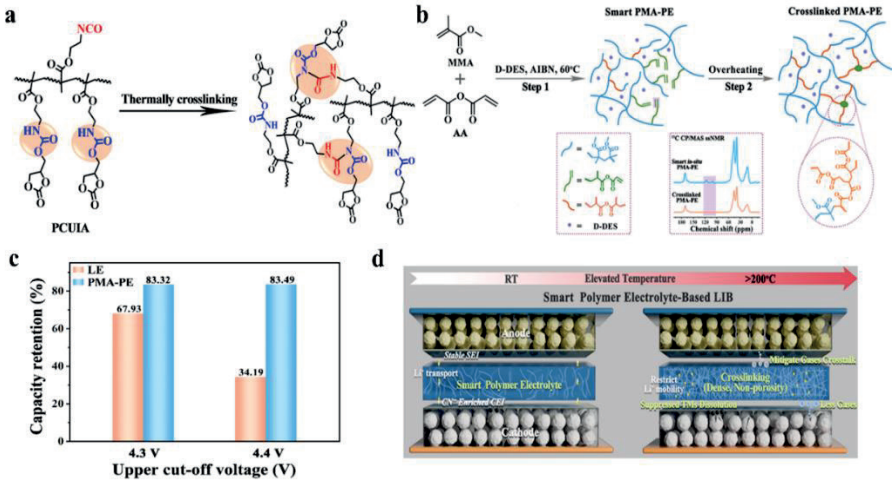

Figure 1. (a) Nucleophilic addition of carbamate to isocyanate units in PCUIA; (b) Re-polymerization of residue acrylate motifs in PMA; (c) Capacity retentions of $LiCoO_2$/graphite full-cells (cathode active mass loading of 15.7 mg cm^{-2}) employing $LiPF_6$/EC-DMC liquid electrolyte and smart PMA-PE after 500 cycles at 0.5 C under 30 ºC. (d) Electrode-anode crosstalk suppression and battery thermal shutdown behaviors under thermal abuse.

References

[1] Zhang, H.; Cui, G. et al., *Energy Storage Mater.* **2022**, *50*, 525.
[1] Zhang, H.; Cui, G. et al., *eScience*, **2022**, *2*, 201
[2] Zhang, H.; Cui, G. et al., *Adv. Mater.* **2024**, *36*, 2400737.
[3] Zhang, H.; Cui, G. et al., *Adv. Mater.* **2025**,*38,* e14210.
[4] Zhang, H.; Cui, G. et al., *Nat. Commun.* **2025**, *16*, 2979.

Regulating intermolecular bonding towards rechargeable Li-$SOCl_2$ battery

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Li-$SOCl_2$ secondary batteries are promising energy storage systems due to their exceptional energy density and robust safety performance under wide-range temperatures. However, their practical application is hindered by the crosstalk of cathode-derived species and electrolyte volatility, which leads to rapid anode deterioration, poor reversibility, and limited cell life. To address these issues, we present an in situ formed quasi-solid-state gel electrolyte mediated through lithium bond (Li-bond), which effectively suppresses the detrimental crosstalk and electrolyte volatilization. This design is achieved through the limited dissociation of fluorinated lithium salts in $SOCl_2$, where the highly polarized additives promote the self-assembly Li-bonding networks. Built upon the solvated chain-like structure of the conventional $LiAlCl_4$-$SOCl_2$ electrolyte, the Li-bonding promotes the formation of a stable lithio-gel electrolyte (LGE). Electrochemical tests demonstrate that the LGE mitigates the detrimental byproducts on Li deposits through restraining the crosstalk of cathode-derived species and significantly extends cycle life of limited Li anode (100μm) at high areal capacities, maintaining a high areal capacity of 6 mAh cm^{-2} at a current density of 5 mA cm^{-2} for 60 cycles, and achieving an active material capacity utilization of over 80% with 3000 mAh g^{-1} capacity for 80 stable cycles, which further enhanced the practicality (minimize Li anode excess). Then we discover a polymer that can sustain the strong Lewis acidity of $SOCl_2$ based systems. This polymer based $SOCl_2$ system can improve the reversibility of plating/stripping performance on anode side and reduce the polarization of cathode reaction at the same time.

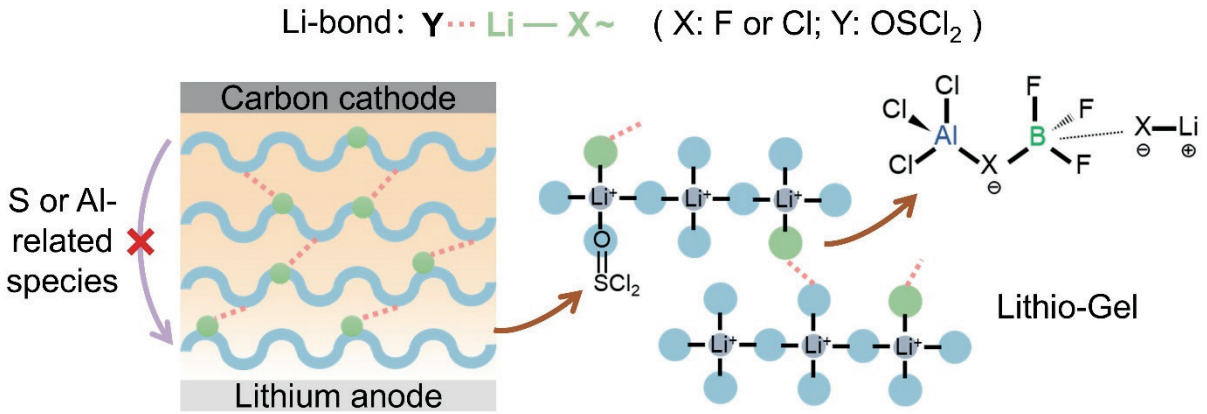

Figure 1. Schematic illustration of Lithio-gel-electrolyte.

References

[1]. G. Chen, X. Liu, H. Hou, X. Gao, L. Hu, S. Dong, G. Cui, *Sci. Bull.,***70** (2025) 140-143.
[2]. G. Chen, W. Li, X. Du, C. Wang, X. Qu, X. Gao, S. Dong, G. Cui, L. Chen, *J. Am. Chem. Soc.* **145** (2023) 22158−22167.

Rapid Synthesis of Hybrid Li_3InCl_6 Halide Electrolytes: An Operando WAXS/EIS Study of Phase Formation

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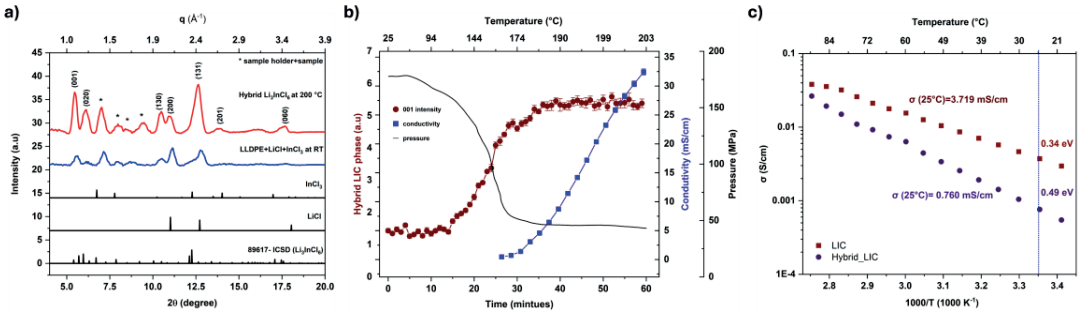
Halide solid-state electrolytes have emerged as promising candidates for next-generation all-solid-state lithium batteries due to their high ionic conductivity, oxidation stability, and simple synthesis routes[1,2]. However, the fundamental mechanisms governing the precursor transformation into the final functional phases are still not well understood, and rapid, scalable synthesis strategies are highly desirable. In this work, we present an *operando* investigation of the in-situ synthesis and rapid formation of Li_3InCl_6 (LIC) and a hybrid LIC electrolyte incorporating linear low-density polyethylene (LLDPE) as a function of temperature. A laboratory-based high-energy wide-angle X-ray scattering (WAXS) setup was developed and integrated with electrochemical impedance spectroscopy (EIS) and pressure monitoring, enabling simultaneous structural, electrochemical, and mechanical characterization during synthesis. Real-time measurements reveal direct conversion of precursors to phase-pure LIC within one hour, with ionic conductivity reaching 3.72 mS/cm for LIC and 0.76 mS/cm for the hybrid LIC at room temperature. Furthermore, this work demonstrates that polymer integration can simultaneously enhance mechanical properties while maintaining high conductivity, establishing *operando* WAXS/EIS as a powerful approach for accelerating the development and scalable manufacturing of solid-state battery materials.



a) XRD patterns of Hybrid LIC, b) Evolution of hybrid LIC phases during temperature increase, c) Arrhenius plots for LIC and hybrid LIC during cooling

References

1. Stankiewicz, N. *et al.* Unveiling the crucial morphological effect of non-conducting polymer binders on inorganic-rich hybrid electrolytes. *J. Mater. Chem. A Mater.* **13**, 20812–20824 (2025).

In Situ photopolymerized ionogels for safe high-Voltage lithium metal batteries

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The increasing demand for high-energy-density storage systems has accelerated research on lithium metal batteries (LMBs) coupled with high-voltage cathodes such as LMNO and LMFP. However, the practical implementation of these systems remains impeded from one side by the high reactivity of metallic lithium leading to uncontrolled dendrite growth, and to the other side from the limited oxidative stability of most common electrolytes. Two possible answers to these challenges are the use of a polymer based electrolyte to mechanically limit dendrite nucleation and growth and the addition of an ionic liquid to enhance the ionic conductivity and the interfacial contact, while taking advantage of the intrinsically high oxidative stability and low vapor pressure. Hence, we report a novel gel polymer electrolyte (GPE) synthesized via in situ UV-induced radical photopolymerization of butyl methacrylate (BMA) and poly(ethylene glycol) diacrylate (PEGDA) matrix incorporating the ionic liquid 1-ethyl-3-methylimidazolium bis(fluorosulfonyl)imide (EMIMFSI) and lithium bis(trifluoromethylsulfonyl)imide (LiTFSI). Electrochemical characterization demonstrates that the optimized GPE exhibits an electrochemical stability window exceeding 4.5 V vs. Li/Li^+, rendering it suitable for high-voltage operation. The GPE shows enhanced interfacial stability, slightly higher ionic conductivity compared to the neat ionic liquid electrolyte, and effective suppression of lithium dendrite growth during prolonged plating/stripping tests. Li/GPE/LMFP full cells deliver an initial discharge capacity of 120 mAh g^{-1} with a Coulombic efficiency of 90% over 50 cycles, and retain a capacity of 100 mAh g^{-1} after 100 cycles. These results highlight the potential of ionogel-based electrolytes as safe and stable candidates for next-generation lithium metal batteries.

Wet vs Dry Processing of PEO and Single Ion Conductive Polymer Electrolytes for Lithium Metal Batteries

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Solid polymer electrolytes (SPEs), composed of lithium salts dispersed in a polymer matrix, have attracted considerable attention for lithium metal batteries due to their enhanced safety, mechanical flexibility, and ability to suppress dendrite formation compared to conventional liquid electrolytes. Conventional fabrication of SPE membranes relies on solution casting and solvent evaporation, which can leave residual solvent traces that negatively affect electrolyte structure and ionic conductivity, while also increasing processing time, energy consumption, and environmental impact. To overcome these limitations, solvent-free dry processing methods have gained interest. In particular, melt compounding offers a scalable route to produce SPEs without solvents, reducing environmental impact and simplifying manufacturing process.

In this work, we directly compare wet (solution-cast) and dry (melt-compounded) SPEs to assess how the fabrication route influences their physicochemical and electrochemical properties. Two systems are investigated: a conventional PEO-based composite electrolyte and a single-ion hybrid polymer electrolyte based on PMTFSILi. All electrochemical and physicochemical tests were performed on both electrolytes and both processing methods.

Li|SPE|Li pouch cells were assembled and cycled at 60 °C to assess interfacial stability and lithium plating/stripping behaviour. Ionic conductivity was measured over a temperature range of 20–80 °C under heating–cooling cycles to evaluate bulk ion transport properties, and the lithium transference number was determined at 60 °C. Cyclic voltammetry (CV) and linear sweep voltammetry (LSV) were conducted at 60 °C to determine the electrochemical stability window. Preliminary results indicate that wet- and dry-processed PEO-based electrolytes exhibit comparable electrochemical performance. Complementary physicochemical characterization was carried out using X-ray photoelectron spectroscopy (XPS) to probe interfacial chemistry and differential scanning calorimetry (DSC) to investigate thermal properties and polymer structure, and optical microscopy to assess film morphology and interfacial uniformity.

These results reveal how wet and dry processing distinctly affect polymer electrolyte performance, highlighting the importance of fabrication choice for optimizing ion transport, interfacial stability, and overall suitability of SPEs in solid-state lithium batteries.

References

[1] Z. Song, et al., *Nature Commun.*, **14** (2023) 4884.
[2] J. Mu, et al., *Polymer Chem.*, **15** (2024) 473-499.
[3] Y. Lu, et al., *Matter*, **5** (2022) 876-898.
[4] J. Yang, et al., *ACS Nano*, **17** (2023) 19903-19913.

Conductivity and Stability Prediction of Anion Exchange Membranes Using Small Data Machine Learning

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Anion exchange membranes (AEMs) are core components of electrochemical energy conversion devices, such as water electrolyzers and fuel cells [1, 2]. Achieving both high hydroxide ion conductivity and sufficient alkaline stability simultaneously remains a central challenge in AEM material design. The performance of AEMs is strongly influenced not only by the polymer backbone but also by the chemical structures of the ion exchange groups (IEGs), which are the structural units that directly govern OH^- transport properties and chemical degradation pathways. Our previously reported machine learning (ML) approach for AEMs represented full polymer structures as molecular descriptor vectors for data-driven property prediction [3]. However, such representations, which are based on global molecular descriptors, struggle to capture the local physicochemical properties of IEGs that are most relevant to conductivity and degradation behavior. The present study adopts a localized perspective by extracting substructures around ion exchange groups (sIEGs) as the analytical units (Fig. 1). A database of 50 anion conductive polymers was constructed from 28 literature sources. Hydroxide ion conductivity (σ) and alkaline stability (Tc10: the elapsed time for conductivity to decrease to 90% of its initial value during accelerated alkaline stability testing) were treated as binary classification targets using median thresholds ($\sigma \geq 0.0765$ S cm^{-1}; Tc10 ≥ 558.5 h), yielding 25 samples in each class. Two types of descriptor sets were computed for each sIEG: structural descriptors (9 features) calculated using RDKit, and electronic/thermodynamic descriptors (10 features) derived from Gaussian quantum chemical calculations, including HOMO/LUMO energies, HOMO–LUMO gap, Gibbs free energy, dipole moment, and logP. To the best of our knowledge, this is the first ML study on AEMs to explicitly incorporate quantum chemically computed electronic descriptors of sIEGs as model features. A Random Forest classifier with stratified 3-fold cross-validation and SHAP analysis was applied to both targets. SHAP analysis revealed that the HOMO–LUMO gap and the sIEG weight fraction (molecular weight of the sIEG unit / equivalent weight) are the dominant features for both targets. Alkaline stability showed higher classification performance than conductivity, suggesting that degradation resistance is more directly encoded in the electronic structure of sIEGs. A larger HOMO–LUMO gap was consistently associated with higher alkaline stability, providing a quantitative link between electronic structure and chemical durability. Conversely, conductivity was found to be governed by a combination of polarity-related descriptors and structural density, suggesting a more complex interplay of factors. These findings offer a physically interpretable pre-screening framework for rational sIEG design prior to synthesis, thereby accelerating the development of high-performance AEMs for hydrogen energy applications.

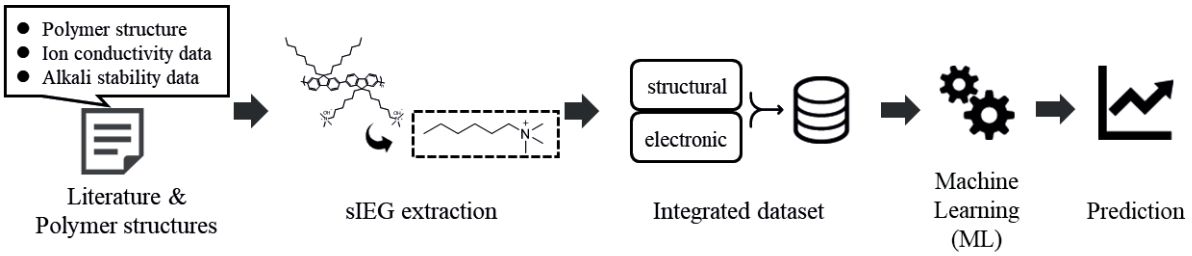

Figure 1. Schematic overview of the proposed framework: sIEG extraction, descriptor calculation (structural and electronic), machine learning-based classification, and property prediction.

References

[1] J. R. Varcoe, *et al.*, *Energy Environ. Sci.*, **7**, 3135–3191 (2014).
[2] C. Santoro, *et al.*, *ChemSusChem*, **15**, e202200027 (2022).
[3] Y. K. Phua, *et al.*, *ChemElectroChem*, **11**, e202400252 (2024).

Correlating Deposition Kinetics and Morphology to Optimize Binder-free Electroactive Interfaces

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Binder-free organic electrodes are attractive for next-generation batteries because they eliminate inactive components, reduce interfacial resistance, and enable efficient utilization of redox-active frameworks. However, their performance is often limited by poor control over deposition kinetics and the resulting ion-transport pathways within the active layer. Here, we establish a direct correlation between nucleation/growth dynamics, mesoscale morphology, and electrochemical transport in pyrene-derived interfaces fabricated via intramolecular electrochemical oxidative cyclodehydrogenation (i EoC), benchmarking cyclic voltammetry, chronoamperometry, and pulse reverse electrodeposition (PRE). Conventional deposition yields either compact films with limited electrochemical accessibility or overgrown dense layers with severely hindered ionic mobility, whereas PRE—by alternating anodic and cathodic polarization—enables dynamic interfacial reorganization and controlled nucleation to build continuous, electrically connected, porous oligopyrene networks directly anchored to the current collector. The optimized PRE condition (pPy 90) exhibits a favourable balance between electrochemically active surface area (0.45 cm2) and apparent ion diffusion (2.01×10^{-6} cm^2/s), lowered charge-transfer resistance, and robust interfacial stability upon extended cycling, demonstrating that performance gains arise primarily from morphological/transport optimization rather than increased molecular weight. These findings establish pulse reverse synthesis as a scalable strategy to engineer binder-free organic battery electrodes with tailored tortuosity and ionic accessibility, offering a clear design route for sodium-ion batteries where larger Na^+ requires open, low-tortuosity architectures for high-rate capability and stable cycling.

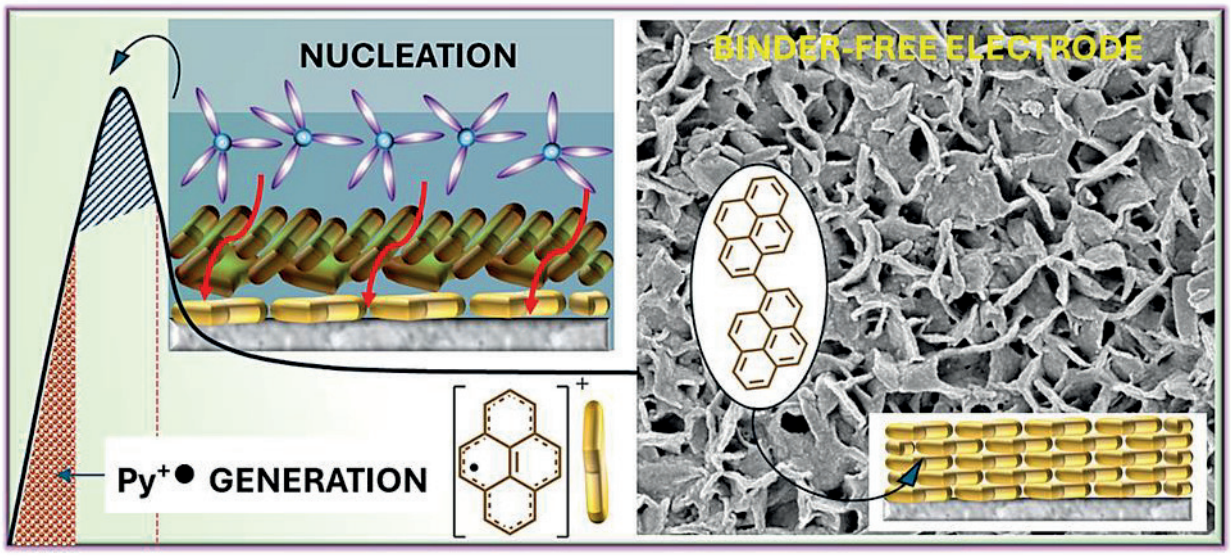

Figure: A schematic for nucleation and growth of a binder-free organic electrode with a morphological view.

References

[1] J. S. M. Lee, A. I. Cooper, *Chem. Rev*. **120** (2020) 2171.
[2] R. Zuzak, J. Castro-Esteban, P. Brandimarte, M. Engelund, A. Cobas, P. Piątkowski, M. Kolmer, D. Pérez, E. Guitián, M. Szymonski, D. Sánchez-Portal, S. Godlewski, D. Peña, *Chem. Commun.* 54 (2018) 10256.

Synthesis and Characterization of Ladder Ionomers via CANAL Polymerization for AEM Fuel Cell and Water Electrolyzer Applications

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Anion Exchange Membrane Water Electolyzers (AEM-WE) can be operated using abundant non-PGM (Platinum Group Metals) catalysts.[1-3] This represents both an economic and scalability advantage compared to proton exchange membrane (PEM) WE, which strongly rely on rare precious metals like iridium and platinum.[1,3] Anion exchange polymers serve as both the solid-electrolyte membrane and the ionomer binder in the electrodes. Although very similar, the requirement of Anion Exchange Ionomers (AEIs) slightly differ from AEM when it comes to gas permeability and film forming properties (Table 1).

Property/requirement	***Binder***	***Membrane***
Anion conductivity	++	+++
OH^- stability	+++	+++
Gas permeability	+++	---
Radical resistance	++	--
Film-forming properties	-	+++
Mechanical properties	+	+++

Table 1 *comparison of the properties required in electrode binders and anion exchange membranes.*

In this study, we present the strategy of using ladder polymers for the synthesis of intrinsically porous AEI for AEM-WE applications (Figure 1). Ladder polymers have been synthesized via Catalytic Arene-Norbornene Annulation (CANAL) polymerization and have been shown to have high rigidity and gas permeability.[4] Quaternization reaction of amine groups using various reagents is a key step in yielding polymer electrolytes.

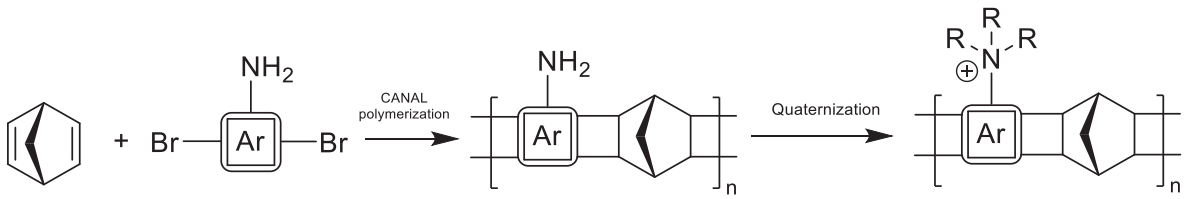

Figure 1. Synthesis of intrinsically porous CANAL-Ionomers.

References

[1] Santoro, C.et al. *ChemSusChem 15*, e202200027 (2022).

2 Heinius, L. , [...] Metzler, L., [...] *et al.. Electrochemical Science Advances* **5**, e202400027 (2025).

3 Klingenhof, M., [...] Metzler, L., [...] et al. Nat Catal 1–10 (2024).

[4] Teo, Y. C.; Lai, H. W. H.; Xia, Y. *Chem. Eur. J. 23*, 14101 (2017).

Nanostructured ion-conducting materials based on polymer nanoparticles and diblock copolymers

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In this work, we investigated the effect of nanoparticle incorporation on the morphology and ion transport in solid electrolytes based on block copolymers. This understanding is key to developing potential SPE materials that are both mechanically robust and exhibit improved ion-transport properties. Thus, the selective placement of lithium sulfonamide functionalized polymer nanoparticles (LiNPs) into a microphase-separated block copolymer, poly(vinyl benzyl methoxy poly(ethylene oxide) ether)-*block*-polystyrene, is investigated here. Different characterization techniques, i.e., transmission electron microscopy (TEM), atomic force microscopy (AFM), and small-angle X-ray scattering (SAXS), showed that the block copolymer morphology is lamellar and is maintained after nanoparticle incorporation. The LiNPs localize selectively within the poly(ethylene oxide) lamellae, leading to complex hierarchical materials, aided by the self-assembly of the block copolymer, the selective ordering of the nanoparticles, and the crystallization of the poly(ethylene oxide). Differential scanning calorimetry (DSC) revealed changes in the crystallinity of the poly(ethylene oxide)- type phase upon addition of nanoparticles and a sulfolane plasticizer. The hierarchical materials exhibited different ionic conductivities depending on nanoparticle content, which could be further improved by adding a plasticizer.

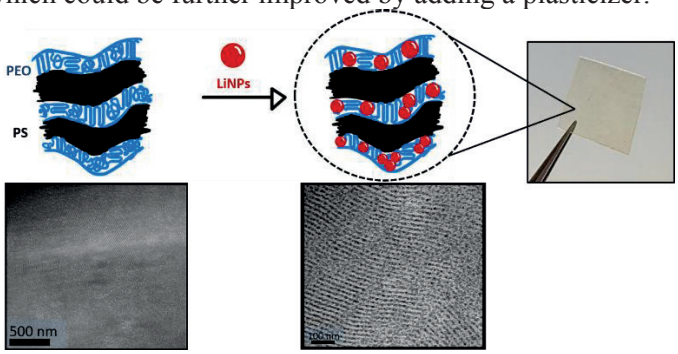

Figure 1 Schematic representation of the selective ordering of the LiNPs within the block copolymer along with TEM micrographs.

References

[1] J.M. Tarascon, M. Armand, *Nature,* **414** (2001) 359-367.
[2] D.T. Hallinan, N.P. Balsara, *Annu. Rev. Mater. Res.*, **43** (2013) 503-525.
[3] A. Herranz Berzosa, A. Gallastegui, et al., *ACS Appl. Polym. Mater.*, **7** (2025) 2286-2296.
[4] A.J. Butzelaar, P. Röring, et al., *ACS Appl. Mater. Interfaces*, **13** (2021) 39257-39270.

Enhancement of Ionic Conductivity in Solid Polymer Electrolytes via Incorporation of a Pyrrolidinium-Based Ionic Plastic Crystal

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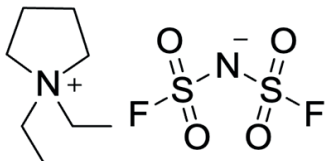
Solid polymer electrolytes have attracted considerable attention as promising electrolytes for next-generation lithium secondary batteries because of their superior safety and mechanical stability compared with conventional organic liquid electrolytes. However, their low ionic conductivity at room temperature remains a major obstacle to practical application. In recent years, ionic plastic crystals (IPCs) have been considered promising solid electrolyte materials because they possess high molecular mobility despite being in the solid state and can exhibit relatively high ionic conductivity when combined with lithium salts. Among them, *N*,*N*-diethylpyrrolidinium bis(fluorosulfonyl)amide ([P22][FSA]) is known as a pyrrolidinium-based ionic plastic crystal, and it has been reported to exhibit a plastic crystal phase over a wide temperature range.[1]



[P22][FSA]

Fig. 1 Chemical structure of [P22][FSA].

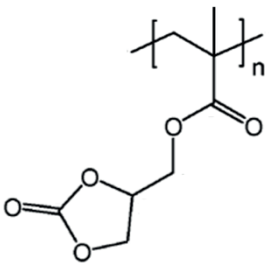


Fig. 2 Chemical structure of polyDOMA.

In this study, we aimed to improve the ionic transport properties of solid polymer electrolytes by introducing the pyrrolidinium-based ionic plastic crystal [P22][FSA]. For this purpose, composite polymer electrolytes were prepared by mixing polyDOMA,[2] LiFSA, and [P22][FSA] at various compositions, and the effect of the [P22][FSA] content on ionic conduction behavior was investigated.

Composite polymer electrolytes were prepared by mixing polyDOMA, LiFSA, and [P22][FSA]. The polyDOMA/LiFSA mixtures are denoted as SPE*x*, where *x* indicates LiFSA concentration (mol% to the repeating unit). The polyDOMA/LiFSA/[P22][FSA] mixtures are denoted as SPE*x*/P22*y*, where *y* indicates the weight ratio (wt%) of [P22][FSA] to the polyDOMA/LiFSA mixtures. Their ionic conductivity was evaluated over a wide temperature range by AC impedance spectroscopy. Based on the obtained Arrhenius plots, the effect of the [P22][FSA] content on ionic conduction behavior was compared.

In the SPE*x*, although the ionic conductivity increased with increasing LiFSA content, the ionic conductivity values around room temperature remained low. In contrast, the incorporation of [P22][FSA] greatly improved the ionic conductivity. The samples containing 70 wt% [P22][FSA] showed the highest ionic conductivity over a wide temperature range, indicating that there is an optimal [P22][FSA] content.

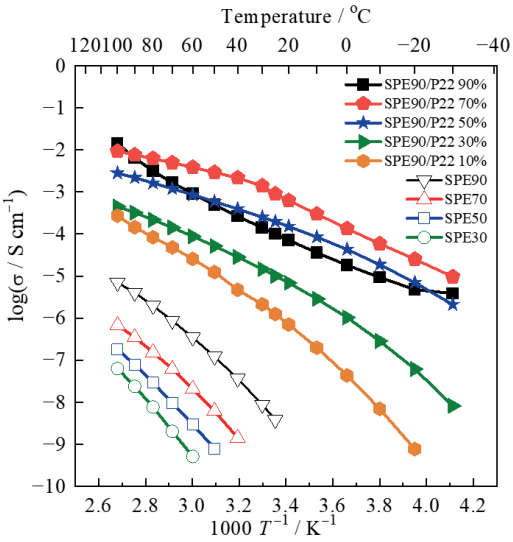


Fig. 3 Arrhenius plots of ionic conductivities for SPE and SPE/[P22][FSA] composite electrolytes.

References

1. H. Yamada, Y. Miyachi, Y. Takeoka, M. Rikukawa, M. Yoshizawa-Fujita, Electrochim. Acta, 303 (2019) 293–298.
2. F. Camara, S. Caillol, B. Boutevin, Eur. Polym. J., 61 (2014) 133–144.

4D imaging of SPE-based all-solid-state lithium batteries

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Lithium-ion batteries currently dominate the modern battery applications. These batteries contain highly flammable organic liquid electrolytes which give rise to safety concerns [1]. The current efforts to pivot towards all-solid-state batteries are partly to address this challenge. All-solid-state lithium batteries also open up the possibility of using the lithium metal anode and high voltage cathodes, to significantly increase the energy density [2]. Solid polymer electrolytes (SPEs) are promising candidates for all-solid-state batteries. However, their full-scale implementation has not been realized because of issues which include early cycling failure and poor cyclability. These issues are mostly attributed to dendrite formation and electrolyte oxidation [3],[4]. One technique that can make a good assessment of this attribution is computed tomography (CT). CT enables non-invasive and non-destructive visualisation of the internal structure of objects [5], making it an attractive technique to explore the morphology and microstructure of battery components without the challenges associated with ex-situ measurements. In our work, we develop and demonstrate a methodology for controlled-temperature *operando* X-ray microtomography for solid polymer electrolyte-based all-solid-state lithium batteries. We performed *operando* 4D imaging of a Li -NMC cell with a PEO/LiTFSI electrolyte at an elevated temperature of 60 °C. We show the surface evolution of the lithium anode and track the changes in the electrolyte thickness, to demonstrate the applicability of the methodology. Figure 1 shows 2D cross-sectional slices of the cell before and after cycling, showing notable changes even to the naked eye.

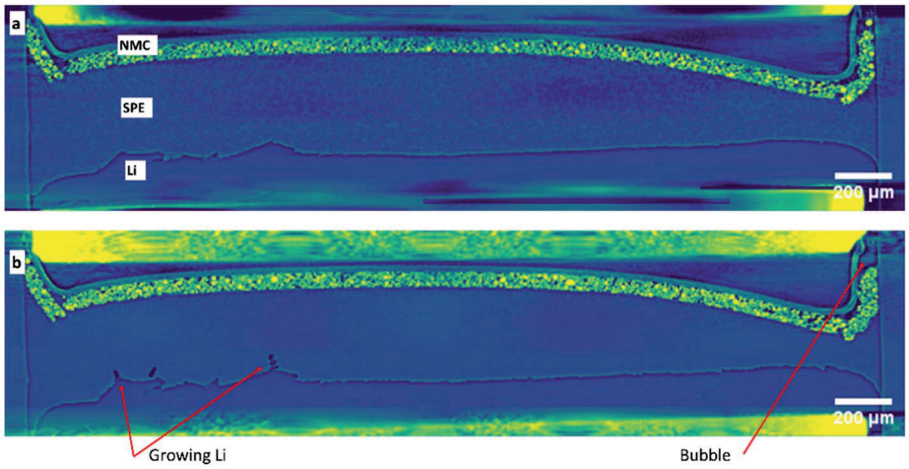

Figure 1. 2D vertical slices of tomograms for a lithium SSB: (A) before cycling, and (B) after cycling.

References

[1] J. Popovic, *Macromolecular Chemistry and Physics*, **223**(2022) 2100344.
[2] M. Ma, M. Zhang, B. Jiang, Y. Du, B. Hu, C. Sun, *Materials Chemistry Frontiers*, **7** (2023) 1268-1297.
[3] L. Stolz, G. Homann, M. Winter, J. Kasnatscheew, *Materials Advances*, **2**(2021) 3251-3256.
[4] I. J. Johansson, C. Sangeland, T. Uemiya, F. Iwasaki, M. Yoshizawa-Fujita, D. Brandell, J. Mindemark, *ACS Applied Energy Materials*, **5**(2022) 10002-10012.
[5] S. R. Stock, *International materials reviews,* **53** (2008) 129-181.

Computational Co-Design of Poly(ionic liquid)s-in-Salt Electrolytes Toward Enhanced Alkali Metal Ion Transport

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Poly(ionic liquid)s (polyILs) combined with high concentrations of alkali metal salts represent a promising class of solvent-free solid polymer electrolytes for next-generation lithium (Li) and sodium (Na) metal batteries. This contribution presents our recent integrated computational–experimental efforts to understand and optimize ion transport in polyILs-in-salt systems based on poly(diallyldimethylammonium) (PDADMA) and its structural analogues, combined with a range of sulfonylimide-type and other counter-anions. [1-5]

Through molecular dynamics (MD) simulations and density functional theory (DFT) calculations, we identified a co-coordination environment in which anions bridge the polycation backbone and the metal cation, facilitating the decoupling of Li^+ diffusion from polymer segmental dynamics (Fig. 1).[2] These findings suggest that appropriate control of the ion–ion interactions within the co-coordination environment is key to further promoting decoupled ion transport and, ultimately, achieving higher ionic conductivity. PolyILs offer an ideal platform for such systematic tuning owing to the inherent modularity of their monomer and anion chemistry. We have previously shown that replacing one methyl substituent on the PDADMA nitrogen center with methoxyethyl, butyl, or isobutyl groups modulates the polycation–anion binding energy and, in turn, alters Li^+ diffusivity.[3] It should be noted, however, that the observed changes in diffusion are not governed solely by ion–ion interactions but are also closely linked to shifts in the glass transition temperature (T_g) of the neat polymer and to variations in the local coordination structure around the metal cation, which remain to be elucidated.

In this work, we extend our exploration to several new polycation architectures such as poly(diallyl-*N,N*-dimethylformamidinium), which features a resonance-stabilized cationic center designed to weaken polycation–anion interactions and discuss the implications for rational molecular design of polyIL-in-salt electrolytes with enhanced ion transport.

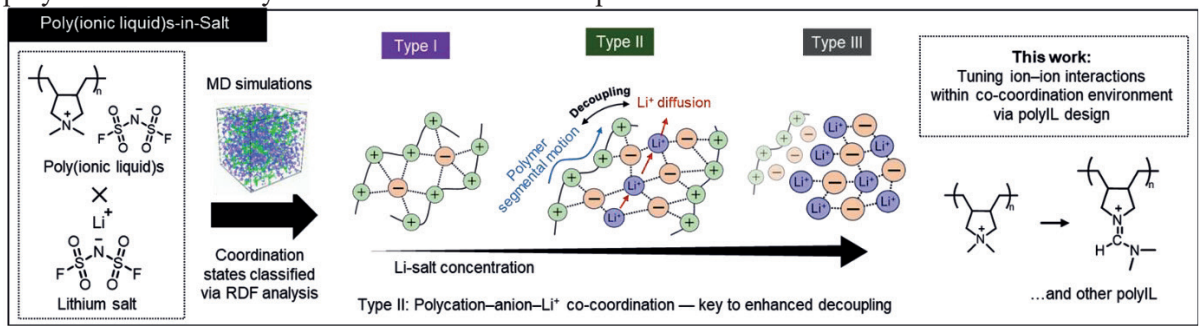

Fig. 1 Overview of the design strategy for polyILs-in-salt electrolytes toward enhanced ion transport.

References

[1] F. Chen *et al.*, *Nat. Mater.*, **2022**, 21, 1175-1182. [2] S. Kondou *et al.*, *J. Am. Chem. Soc.*, **2024**, 146, 33169-33178. [3] K. Cai *et al.*, *Batteries Supercaps.*, **2025**, 8, e202500484. [4] F. Chen *et al.*, *ACS Appl. Polym. Mater.*, **2025**, 7, 15420-15428. [5] G. Lingua *et al.*, *Acc. Mater. Res.*, **2025**, 6, 1368-1378.

Molten Salt Electrolytes – A Combined Experimental and Computational Approach

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Aiming to combine the strengths of both liquid and solid-state lithium-based electrolytes, semi-solid electrolytes (SeSEs) have recently gained popularity [1,2]. Within the various SeSEs, molten salt electrolytes (MSEs) are promising due to their, in principle, compatibility with high-voltage cathodes, such as NMC811, and intrinsic non-flammability, as there are no solvent or other organic components present. While originally only high-temperature applications were targeted [3], several adaptations towards ambient temperature applications have been made by mixing lithium salts with varying anion or cation to form eutectic compositions. Mixtures with varying cations are more common [3,4], whilst approaches considering a variation in the anion are much scarcer with no systematic study being reported. A fully lithium-based, binary MSE was though designed by Y. Ito *et al*. [5] and successfully employed in a graphite-lithium cell at 80 °C.

Here, a systematic screening of binary MSEs is presented, which relies on thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) to evaluate the thermal properties, along with rheology and pulse-gradient spin-echo nuclear magnetic resonance (PGSE-NMR) to assess the dynamics. Additionally, Raman spectroscopy allows us to probe local structures, which will be rationalized with the support of DFT calculations. Moreover, statistical thermodynamic data will be computed using COSMO-RS. This combined computational and experimental approach will allow to construct a holistic and robust insight in the promises of binary MSEs for application in proof-of-concept cells.

References

[1] A.M. Navarro-Suárez, P. Johansson, *J. Electrochem. Soc.*, **167** (2020) 070511
[2] Y. Yang, Y. Ma, K. Feng, S. Zheng, Z. Wang, *J. Coll. and Int. Sci.*, **700** (2025) 138323
[3] S. Yan et *al.*, *Energy Environ. Sci.*, **18** (2025) 1696-1706.
[4] J. Timhagen, C. Cruz, J. Weidow, P. Johansson, *Batt. & Supercaps*, **7** (2024) e202400297
[5] Y. Ito, K. Kubota, Y. Maeyoshi, T. Okumura, K. Yoshii, *J. Phys. Chem. C*, **129** (2025) 9656-9661.

Ion Transport Properties of Composite Materials Based on Deeply Supercooled Li-Salt (Li-DSS)

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Improving the charge–discharge performance of Li-based secondary batteries requires the development of electrolytes that exhibit not only high ionic conductivity (σ_{ion}) but also a high lithium-ion transference number (t_{Li}). In particular, next-generation electrolytes with single lithium-ion conduction ($t_{Li} \approx 1$) are highly desirable [1]. One representative example is all-solid-state batteries employing inorganic solid electrolytes [2]. However, challenges in the design of stable electrolyte–electrode interfaces remain. On the other hand, in liquid electrolytes that are advantageous for interface design, one of the simplest approaches to achieve $t_{Li} \approx 1$ is the use of Li molten salt, which contain no solvent and cannot form a concentration gradient of lithium salts under anion-blocking conditions [3],[4]. However, many lithium salts possess both high melting points and high crystallinity, leading to crystallization near room temperature.

To address this issue, we have recently demonstrated that the addition of a trace amount of polymer to lithium salts effectively suppresses crystallization, enabling the formation of a long-term stable supercooled liquid at ambient temperature. We refer to this system as a deeply supercooled Li-salt (Li-DSS), which exhibits near single lithium-ion conduction ($t_{Li} \approx 1$). However, the σ_{ion} of Li-DSS at room temperature remains insufficient for practical applications.

In this study, we investigated the ion transport properties of Li-DSS. Fig. 1 shows the temperature dependence of σ_{ion} for Li-DSS composed of poly(methyl methacrylate) (PMMA, $M_n \approx$ 48,000), lithium (fluorosulfonyl)(trifluoromethanesulfonyl)amide (Li[FTA]), and lithium bis(fluorosulfonyl)amide (Li[FSA]) with the corresponding molten salt mixture ($Li[(FTA)_{0.5}(FSA)_{0.5}]$) as a reference. Li-DSS exhibited higher σ_{ion} than the reference molten salt, particularly near ambient temperature. This enhancement is attributed to the lower glass transition temperature of Li-DSS ($T_{g,\ Li\text{-}DSS}$: −46.1 °C) compared to that of the molten salt ($T_{g,\ molten\ salt}$: −22.7°C), which enhances ionic mobility. In this presentation, we will discuss strategies to further reduce T_g by incorporating additional components into the Li-DSS system.

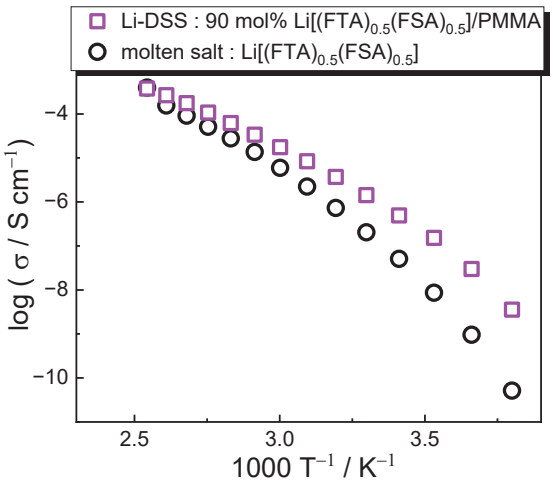

Fig. 1. Temperature dependence of ionic conductivity (σ) in Li-DSS (pink) and molten salt (black) [5].

References

[1] K. M. Diederichsen *et al*., *ACS Energy Lett.,* 2017, **2**, 2563-2575
[2] Y. Kato, *et al*., *Nat. Energy*, 2016, **1**, 16030.
[3] K. Kubota *et al*., *J. Phys. Chem. C*, 2013, **117**, 18829-18836.
[4] K. Shigenobu *et al*., *Phys. Chem. Chem. Phys.*, 2023, **25**, 6970-6978.
[5] T. Sudoh *et al*., *Adv. Energy Mater.,* 2026, **16**, e05229

Ga-based Liquid Metal−Ionic Liquid Composite Gels for Stretchable Li-ion Battery Electrodes

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Liquid metals (LMs) exhibit low melting points, high electronic conductivity, and excellent fluidity. Although mercury is a representative LM, its toxicity limits its practical applications. In contrast, Ga-based alloys form LMs with melting points below room temperature and relatively low toxicity [1]. In this study, we explore stretchable materials with mixed electronic-ionic conductivity by utilizing the high deformability of Ga-based LMs and room-temperature ionic liquids (ILs) in composite gel membranes. ILs have attracted considerable attention owing to their high ionic conductivity, low volatility, and thermal stability. Ion gels, composed of polymer networks swollen with ILs, not only retain these properties but also exhibit self-supporting and elastic characteristics [2]. In our previous work, we reported LM-IL composite gels formed by incorporating a Ga-In eutectic LM into an ion gel, which exhibits both electronic and ionic conductivity along with flexibility and stretchability [3]. In this study, we aim to develop stretchable Li-ion battery electrodes with high electronic/ionic conductivity as well as mechanical flexibility and stretchability (**Fig. 1**) by incorporating Ga-based LMs and active materials such as $Li_4Ti_5O_{12}$ (LTO) and $LiFePO_4$ (LFP). To further enhance electronic conductivity, metallic solid filler and their content were also optimized. In addition, the polymer matrix of the composite gel membrane was varied to improve stretchability. The LM-IL composite gel electrode containing LTO demonstrated stable charge-discharge performance even under stretching conditions (**Fig. 2**). In contrast, for LFP-based electrode, Ga-In undergoes corrosion at around 2.7 V vs. Li/Li^+, limiting its applicability as a cathode component. To address this issue, Ga-In was replaced with Ga-Sn, which has superior electrochemical stability, and electrolyte additives were further investigated to suppress corrosion at higher potentials. In the presentation, we will also report the corrosion suppression effect of additives, as well as the electrochemical performance of the composite gel electrodes.

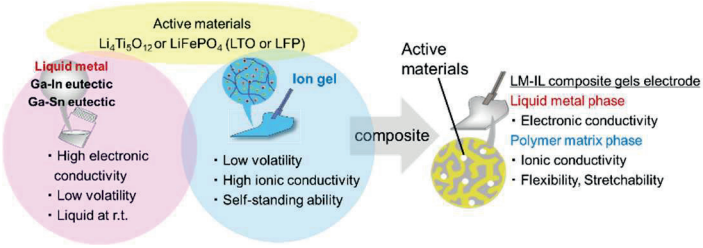

Fig. 1 Preparation of LM-IL composite gels consisting of Ga-based LM, IL and active materials (LTO or LFP) of Li-ion batteries.

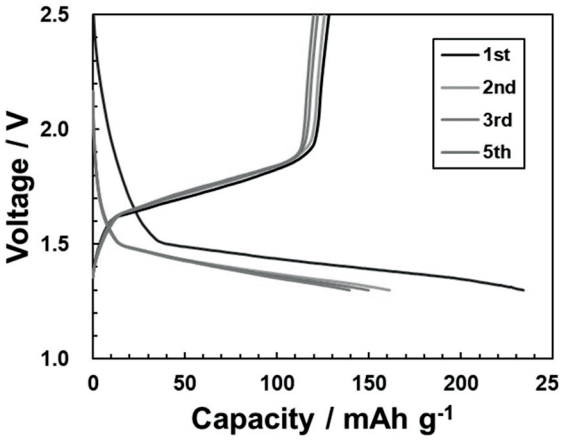

Fig. 2 Charge-discharge curves of the LM-IL composite gel electrode containing LTO under 50% stretching.

References
[1] N. Kazem, et al, Adv. Mater., **2017**, 29, 1-14.
[2] M. A. B. H. Susan, et al, J. Am. Chem. Soc., **2005**, 127, 4976.
[3] J. Asada, *et al*., *Macromol. Chem. Phys.*, **2022**, 223, 2100319.

Thermal and Electrochemical Properties of Oligoether-Functionalized Asymmetric Lithium Imide Salts for Molten Salt Electrolytes

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Fast charge-discharge performance in Li secondary batteries requires electrolyte materials with both high ionic conductivity and a high Li ion transference number (t_{Li}) [1]. Solvent-free molten Li salts are promising candidates for achieving high Li ion transference numbers ($t_{\mathrm{Li}} \approx 1$) in the liquid state. However, conventional molten Li salts typically require high-temperature operation due to their high melting points (T_{m}), high viscosities and low ionic conductivities [2]. Inspired by advances in ionic liquid research [3], the introduction of ether-functionalized ions has been shown to effectively reduce melting points and viscosities by enhancing conformational flexibility and disrupting efficient molecular packing. Based on this concept, we explored asymmetric Li imide salts as candidates for low-melting molten salt electrolytes. In this study, we investigated a series of Li imide salts with controlled variations in the number and positioning of ether oxygen atoms, as well as anion geometry and architecture, to elucidate how anion size, flexibility, and branching influence melting behavior and electrochemical properties. Among the studied salts, those containing two ether oxygen atoms within linear oligoether segments, such as Li[TfN2O2O1] exhibit relatively low T_{m} (Fig. 1a) [4]. In contrast, Li[DMESI] (Fig. 1a), a structural analogue with a branched oligoether chain, shows a higher T_{m} (>150 °C), highlighting key molecular design principles for low-melting Li imide salts. Furthermore, a Li imide salt incorporating a poly(propylene oxide)-type oligomer side chain remains liquid at room temperature, demonstrating the effectiveness of steric disruption in suppressing crystallization. Electrochemical measurements were also conducted for selected salts with relatively low melting points, including ionic conductivity (Fig. 1b), t_{Li}, and electrochemical stability. These properties are discussed in relation to Li^+ coordination structures revealed by both experimental and computational analyses in molten salt electrolytes.

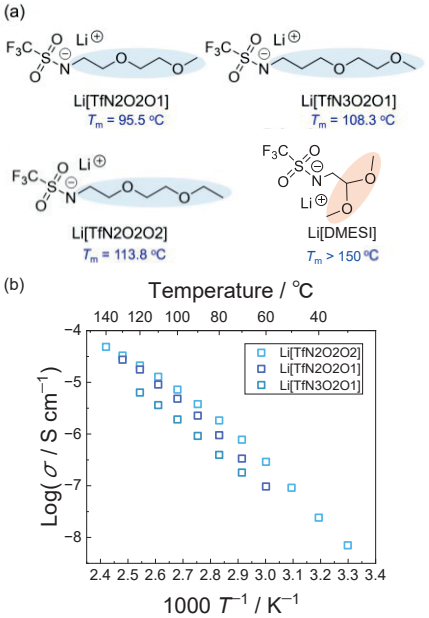

Fig. 1 (a) Chemical structures and T_{m} and (b) ionic conductivities of oligoether-functionalized asymmetric Li imide salts.

References

[1] K. M. Diederichsen *et al., ACS Energy Letters*, **2**, 2017, 2563-2575.
[2] K. Shigenobu *et al.*, *Phys. Chem. Chem. Phys.*, **25**, 2023, 6970-6978.
[3] S. Tang *et al.*, *Chem. Soc. Rev.*, **41**, 2012, 4030.
[4] Y. Matsuyama *et al., Energy Adv.*, **4**, 2025, 1438.

Poly(Solvate Ionic Liquids) Electrolytes Based on Polymeric Lithium Salts

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To improve the performance and safety of Li-ion batteries, it is essential to develop electrolyte materials that exhibit not only high ionic conductivity (σ) and Li ion transference number (t_{Li}), but also high thermal and electrochemical stability, along with the ability to form robust electrode/electrolyte interface [1]. Our research group has previously investigated liquid electrolytes based on polymeric Li salts dissolved in organic solvents, where anionic functional groups are immobilized on polymer chains [2]. While this approach achieves a high t_{Li} (~ 0.8) by effectively suppressing anionic mobility, their thermal stability remains insufficient due to the presence of free flammable solvents. In contrast, equimolar complexes composed of oligoethers (glymes) and Li salts (e.g., LiTFSA) form solvate ionic liquids (SILs) with negligible free solvent content, exhibiting excellent thermal and electrochemical stability [3]. However, these systems typically show a significantly low t_{Li} (< 0.1) [4]. In this study, we studied a new class of polymer-based electrolytes, termed poly(solvate ionic liquids), which integrate the advantages of polymeric Li salts and solvate ionic liquids. This hybrid approach is expected to simultaneously achieve high t_{Li} and improved thermal stability [5].

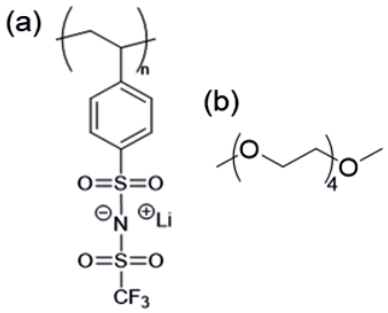

Fig. 1 Chemical structures of (a) PLiSTFSA and (b) tetraglyme.

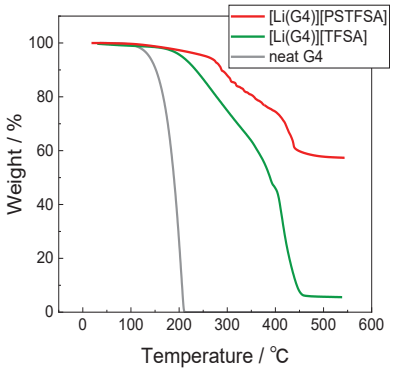

Fig. 2 TG curves of [Li(G4)]X and neat G4 at a heating rate of 10 °C min^{-1} under nitrogen atmosphere. [3]

A polymeric Li salt, PLiSTFSA (Fig. 1a) was synthesized via radical polymerization of 4-styrenesulfonyl (trifluoromethanesulfonyl)amide (LiSTFSA). An equimolar mixture, [Li(G4)][PSTFSA], was prepared using tetraglyme (G4) (Fig. 1b). Thermal stability was evaluated by thermogravimetric analysis (TGA) (Fig. 2). [Li(G4)][PSTFSA] exhibited significantly higher thermal stability than neat G4, indicating that the strong interaction between Li ions and G4 effectively suppresses the volatilization of the solvent. Furthermore, the 5 % weight loss temperature (T_d) of [Li(G4)][PSTFSA] (T_d = 257 °C) was higher than that of the conventional SIL, [Li(G4)][TFSA] (T_d = 204 °C), suggesting the formation of stable complex cations ($[Li(G4)]^+$) and a near absence of free G4 molecules. Furthermore, the t_{Li} measured under anion-blocking conditions using Li symmetric cells reached 0.64 at 80 °C. These results demonstrate that [Li(G4)][PSTFSA] forms a poly(solvate ionic liquid) despite the relatively strong Li^+-anion interactions, likely due to counterion condensation in polyelectrolyte system.

References

[1] K. M. Diederichsen *et al.*, *ACS Energy Lett.* **2**, **11**, 2563 (2017).
[2] S. Kondou *et al.*, *ACS Appl. Mater. Interfaces*, **14**, 18324 (2022).
[3] K. Ueno *et al.*, *J. Phys. Chem. B*, **116**, 11323 (2012).
[4] K. Shigenobu *et al.*, *Phys. Chem. Chem. Phys.*, **22**, 15214 (2020).
[5] W. Zhang *et al.*, *Chem. Mater.*, **33**, 524 (2021).

Sulfur-Substituted Polymer Electrolytes with Weak Li–S Coordination for Fast Lithium-Ion Transport

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With the continued growth of the secondary battery market, increasing attention has been devoted to the development of electrolyte systems with improved thermal and electrochemical stability and reliability for lithium-ion batteries. Conventional liquid electrolytes provide high ionic conductivity and excellent electrode wettability; however, they also suffer from several inherent drawbacks, including leakage, flammability, and poor thermal stability. To overcome these limitations, polymer electrolytes have been widely regarded as promising alternatives.

Among them, poly(ethylene oxide) (PEO)-based polymer electrolytes have been extensively studied because they enable lithium-ion transport through the segmental motion of polymer chains while also offering relatively good lithium salt solubility. In addition, they exhibit superior mechanical and thermal stability compared with liquid electrolytes. Nevertheless, PEO-based electrolytes still suffer from several limitations. Their semi-crystalline nature at ambient and intermediate temperatures reduces the fraction of amorphous domains favorable for ion conduction, while the strong coordination interaction between ether oxygen atoms and lithium ions suppresses lithium-ion mobility.

Oxygen-to-sulfur substitution has been considered a possible structural strategy to weaken such lithium-ion coordination. Because sulfur possesses a larger electron shell than oxygen, the distance between the valence electrons and the nucleus increases, which can lead to weaker interactions with lithium ions. This weaker coordination is expected to contribute to enhanced lithium-ion mobility. However, sulfur substitution may simultaneously reduce lithium salt solubility and therefore requires careful consideration in electrolyte design.

From this perspective, poly(tetrahydrofuran) (PTHF)-based structures are particularly attractive. Compared with PEO, PTHF has a lower oxygen density and a higher carbon-to-oxygen ratio, which is expected to suppress crystallization. These structural characteristics may enhance polymer chain flexibility and thereby facilitate ion transport. Furthermore, PTHF-based systems may provide a more balanced environment by mitigating the decrease in lithium salt solubility that can arise in sulfur-containing structures. In addition, in situ polymerization offers further advantages, including intimate interfacial contact between the electrolyte and electrodes after cell assembly and the formation of a uniform electrolyte phase within the cell.

In this study, a thiirane-containing monomer was synthesized by converting the epoxy groups of 1,4-butanediol diglycidyl ether (BTDE) using potassium thiocyanate (KSCN). The resulting precursor was subsequently polymerized in situ with THF to prepare a polymer electrolyte. This study aims to propose a design strategy for polymer electrolytes for lithium-ion batteries by combining sulfur substitution, a PTHF-based structure, and in situ polymerization.

References

[1] Adisu Makeyaw et al., *Am. J. Quantum Chem. Mol. Sepctrosc*., **9** (2025) 12–30
[2] Kitalu Ricin Ngoy et al., *Renew. Sustaion. Energy. Rev.,* **223** (2025) 115971
[3] Mark A. Ratner et al., *MRS Bull.,* **25** (2000) 31–37
[4] P. Johansson, *Polymer*, **42** (2001) 4367–4373
[5] Deya Das et al., *Chem. Mater*. **30** (2018) 24, 8804–8810

In Situ Polymerized Sulfur–Containing Allyl Thiirane Electrolytes Initiated by LiFSI for High Conductivity and Interface Stability

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Due to their high energy density and excellent cycle characteristics, lithium-ion batteries are widely used in various fields such as portable electronic devices, electric vehicles, and energy storage systems. However, traditional carbonate-based liquid electrolytes are vulnerable to safety due to their high volatility and flammability, and can cause problems such as thermal runaway under high temperature and overcharging conditions. These problems have consistently raised the need for safer electrolyte systems.

In this situation, polymer electrolytes with excellent stability are attracting attention as next-generation electrolytes. In particular, solid and gel polymer electrolytes have the advantage of being able to form a stable interface with electrodes and provide improved mechanical stability. However, polymer electrolytes generally have limitations of low ion conductivity and high interfacial resistance, which is a major cause of electrochemical performance degradation when the battery is actually operated. In addition, the existing polymer matrix, poly(ethylene oxide) (PEO), has structural problems in which the movement of lithium ions is limited due to strong coordination bonds with lithium ions and high crystallinity.

To overcome the limitations of these polymer electrolytes, this study proposes a strategy to improve ion transport properties and interfacial stability by introducing a functional group in the polymer structure. The polymer containing sulfur has a lower electronegativity than the oxygen-based polymer, so its binding force with lithium ions is weak, which can promote ion dissociation and migration. In addition, unsaturated functional groups such as allyl can be reduced during the initial charging and discharging process to form a stable solid electrolyte interface (SEI), which can effectively protect the electrode interface.

Furthermore, by introducing the in situ polymerization process to form a polymer electrolyte directly on the electrode surface, it was intended to implement a close interface between the electrode and the electrolyte to reduce the interfacial resistance effectively. By combining this functional group design and in situ polymerization strategy, this study proposes a high-performance polymer electrolyte system that can improve ion conductivity, interfacial stability, and battery safety.

The synthesized polymer is characterized using FT-IR and thermal properties are analyzed using TGA and DSC. The electrochemical properties of the polymer electrolyte are investigated through EIS and charge/discharge studies.

References

[1] Khan, A.; Al Rashid, H.; Roy, P. K.; Chowdhury, S. I.; Sathi, S. A. Challenges and the Way to Improve Lithium-Ion Battery Technology for Next-Generation Energy Storage. *Energy & Environmental Materials* **2025**, *8* (6), e70088.

[2] Wang, L.; Yue, K.; Qiao, Q.; Zhao, Z.; Xu, Y.; Pan, L.; Liu, Y.; Li, H.; Zhu, B. In Situ Self-Polymerization of Thioctic Acid Enabled Interphase Engineering Towards High-Performance Lithium–Sulfur Battery. *Advanced Energy Materials* **2025**, *15* (1), 2402617.

[3] Fasulo, F.; Muñoz-García, A. B.; Massaro, A.; Crescenzi, O.; Huang, C.; Pavone, M. Vinylene carbonate reactivity at lithium metal surface: first-principles insights into the early steps of SEI formation. *Journal of Materials Chemistry A* **2023**, *11* (11), 5660–5669.

[4] Wagner-Henke, J.; Kuai, D.; Balbuena, P. B.; Krewer, U. Understanding the effect of vinylene carbonate on SEI formation, morphology and stability on lithium metal anodes. *Energy Storage Materials* **2025**, 104434.

Electrochemical CO_2 Absorption Behavior Properties of Anthraquinone Bearing Ionic Liquid Moiety

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CCUS (Carbon dioxide Capture, Utilization and Storage), is attracting significant attention as an effective method for reducing carbon dioxide (CO_2) emissions toward carbon neutrality by 2050. Currently, CO_2 capture is mainly achieved by chemical absorption using liquid amine followed by thermal desorption. However, this process requires a large amount of energy for CO_2 release, resulting in low overall energy efficiency. Electrochemical CO_2 capture has emerged as a promising alternative because it facilitates the use of renewable energy and offers high energy efficiency. Quinone-based systems are widely studied as CO_2 absorbents used in this method.[1] In non-aqueous solvents, reduced quinone species selectively capture CO_2 from a dilute source solution[2] via nucleophilic addition reactions and release CO_2 through electrochemical oxidation. However, the solubility of quinones in non-aqueous solvents is low, which limits the total CO_2 carrying capacity. To address solubility issue, our group has developed anthraquinones functionalized with ionic liquids moieties, namely ionic anthraquinones, which exhibit significantly enhanced solubility in non-aqueous solvents.[3]

In this study, we investigated the effects of solvents and supporting electrolytes on the CO_2 absorption behavior of an ionic quinone, tetrabutylammonium 9,10-anthoraquinone-2-sulfonate ([TBA][AQS]). Typical results of cyclic voltammograms under Ar and CO_2 are shown in **Fig. 1**. Under Ar, two reduction waves are observed, whereas under CO_2, they merge into a single broad wave due to a positive shift of the second reduction potential. Because one quinone molecule stoichiometrically captures two CO_2 molecules, the Coulombic ratio was evaluated by comparing the reduction charge under CO_2 with that of the first reduction wave under Ar. In this study, this Coulomb ratio was used as an indicator of CO_2 separation ability.

Cyclic voltammetry (CV) measurements were performed on a solution containing 0.2 M [TBA][AQS], 1 M [TBA][PF_6] as a supporting electrolyte, and 0.1 M H_2O, and the Coulombic ratios were calculated for each cycle. Although DMSO exhibits lower reduction currents than acetonitrile due to its higher viscosity, it shows a higher Coulombic ratio. This suggests that the high permitivity of DMSO effectively screens electrostatic interactions between the one-electron reduced AQS species and its counter cation, thereby enhancing the reactivity between the quinone and CO_2.

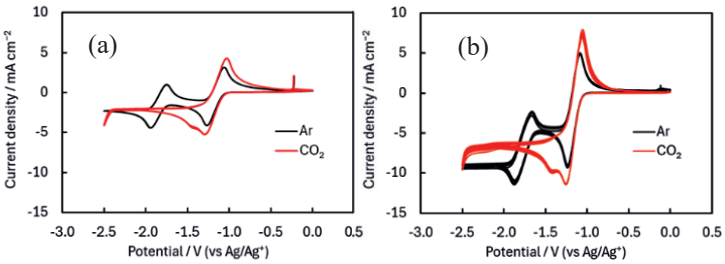

Fig. 1 CV of 0.2 M [TBA][AQS] in 1 M [TBA][PF_6]/ (a) DMSO and (b) Acetonitrile + 0.1 M H_2O under Ar (black) and CO_2 (red) with GC electrode.

References

[1] B. Gurkan, et al., *ACS Sustainable Chem. Eng.* **2015**, 3, 1394.
[2] P. Scovazzo, *et al*., *J. Electrochem. Soc*., **2003**, *150* , D91.
[3] H. Iida *et al.*, *J. Phys. Chem. C*, **2023**, *127*, 10077.

Interfacial Behaviour of Li-Metal Anodes and Polymer Electrolytes in Solid-State Cell Architectures

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Lithium (Li) metal is considered one of the most promising anode material choices for solid-state battery applications due to its high specific capacity (3860 mAh/g) and low electrochemical potential. However, Li metal faces significant challenges that limit its practical application and commercialization as anode material, for example lithium dendrite formation during cycling, leading to internal short circuits, capacity degradation and safety risks, highlighted in this study in the case of commercial copper lithium (Cu-Li) laminate. To mitigate these challenges, the interphase of the lithium anode and solid polymer electrolyte should be improved. [1]

One potential option for anode materials is pulsed laser deposited (PLD) lithium anodes [1] alloyed with either indium (In) [2] or magnesium (Mg) [3], thus chosen to be studied further in this research. The electrolytes chosen for the symmetrical coin cell assembly include a polyethylene oxide (PEO) based electrolyte and two single-ion conducting PVDF-HFP based electrolytes with different compositions. Initial characterization for anode materials includes SEM-EDS analysis of both surface and cross section to evaluate coating quality of the alloyed samples and elemental distribution on coating, grazing incidence XRD to detect phases and alloys present in the coatings as well as surface XPS to detect the atomic percentage of different elements. Anode materials and electrolytes are then cut and assembled as symmetrical coin cells (CR2032). Electrochemical impedance spectroscopy (EIS) is performed after conditioning and after every 10 cycles. Finally, cells are disassembled and cross-sectional SEM-EDS analysis is performed to investigate reasons for short circuits (e.g. dendrites, dead lithium, resistive SEI) and to tentatively evaluate failure mode. Additionally, this research offers a critical point of view to coin cells in solid state battery research, for example the critical effect of component assembly order and as well as hot lamination of active material stack and presents initial results from disc in pouch method as an alternative.

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References

[1] N. Zamperlin, R. Cid, V. Kekkonen, A. Pesce, M. Cheddadi, B. M. Gomes, Manuela C. Baptista, M. H. Braga, A. Orue, P. López-Aranguren, Advanced manufacturing of thin-film lithium metal anode by pulsed-laser deposition for next-generation solid-state batteries, *Journal of Power Sources*, Volume **655** (2025)

[2] W. Jing, K. Zou, X. Dai, J. Sun, Q. Tan, Y. Chen, Y. Liu, Li-Indium alloy anode for high-performance Li-metal batteries, *Journal of Alloys and Compounds*, Volume **924** (2022)

[3] Yan, R. Fan, H. Cui, Mg-Li-Cu alloy anode for highly reversible lithium metal batteries*, Journal of Magnesium and Alloys*, Volume **13**, Issue **5**, Pages 2144-2154 (2025)

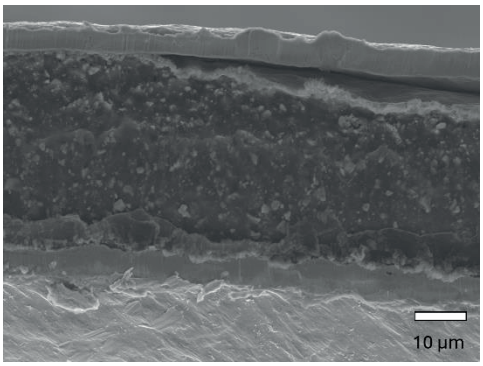

Figure 1: SEM cross section of cycled Cu-Li laminate + PEO symmetrical cell

Impact of Particle Sizes in Hybrid-Polymer Electrolytes on Lithium Inventory Homogeneity in Lithium Metal Batteries

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Motivated by the increasing demand for batteries with both high performance and high energy density, significant research efforts are currently in progress. Lithium metal anodes, in particular, are considered highly promising due to their substantially larger energy density compared to conventional anode materials.[1] A key challenge in their practical application comprises retention of the lithium inventory throughout electrochemical cycling. Insufficient preservation of lithium results in the formation of high-surface-area lithium, promoting dendrite growth and consequently raises significant safety risks.[2]

Grafted hybrid-polymer electrolytes have demonstrated significant effectiveness in suppressing dendrite formation while simultaneously preventing leakage of hazardous solvents. These electrolytes exhibit excellent cell longevity, indicating the effectiveness of dendrite suppression.[3] However, theoretical work indicated that mixing particles and polymers may cause irregular current flows and thus inhomogeneous lithium inventory.[4] Therefore, this study analyzes the impact of particle sizes and their distribution in hybrid-polymer electrolytes (Al_2O_3-PCL[3]) in view of their mechanical properties, ionic conductivity and especially, lithium plating in NMC||Li cells *via* SEM analysis.

The resulting 3D-SEM reconstructions demonstrate variations in the particle size distribution and aggregation of Al_2O_3 particles. Although particle size distribution reflects gaussian behavior, still the largest agglomerate contributes with 3 vol.% to the overall particle volume inside the hybrid polymer electrolyte, thus raising concerns for maintaining homogeneous lithium plating. Here, 3D-SEM reconstructions are evaluated as input structures for simulations of ionic conductivity and lithium concentration, respectively.[5]

In general, the present work shows trends of cell performance, demonstrating that particle size distribution is an important parameter. With larger average particle sizes, mechanical stability increases, while the achievable ionic conductivity is reduced. Also, SEM analysis of NMC||Li cells was performed after 100 cycles. The results illustrate the impact of particle size distribution in hybrid polymer electrolytes on the homogeneity and likely reversibility of lithium inventory. In this way, design criteria for 'ideal' hybrid polymer electrolytes can be obtained.[6]

Overall, this work clearly highlights the benefits of SEM imaging for achieving insights to cell designs, accounting for increasing complexity of cell constituents, while allowing for more comprehensive understanding of internal battery components and electrolyte|electrode interfaces. Especially, quantification of parameters such as particle distribution *via* SEM gives valuable insights in designing hybrid polymer electrolytes and multi-layer systems in future.

References:

[1] Brunklaus, G. et al, *Nature Reviews Electrical Engineering* **2024**, *1*, 79
[2] Lennartz, P. et al., *Joule* **2023**, *7*, 1471
[3] Scharf, F. et al., *Small* **2024**, *20*, 2404537
[4] Barai et. al., *ACS Appl. Energy Mater*. **2023**, *6*, 2160−2177
[5] Pleie, J., Tänzer, N., in preparation
[6] Tänzer, N., in preparation

Topology-dependent transport mechanism(s) in lithium-based molten salt electrolytes

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Molten salt electrolytes (MSEs) constitute an emerging class of solvent-free electrolytes suitable for several battery technologies [1]. They exhibit wide electrochemical stability windows [2,3] and enhanced safety, both due to the absence of solvents or other organic components [3]. The absence of solvent also causes the ion transport mechanism(s) to depend strongly on ion–ion correlations, percolating structural networks, and continuous dynamic restructuring. As a result, the ion transport differs markedly from the conventional vehicular model. These characteristics necessitate alternative approaches for analysing MSEs.

Here, classical molecular dynamics simulations of MSEs are combined with an analysis based on the CHAMPION framework [4] to investigate the time-dependent interactions between ions, enabling an assessment of how topology influences ion transport [5]. This is made possible by the hierarchical graph approach CHAMPION takes [4], identifying ions and molecules to be members of dynamic structures (Fig. 1). As an initial set of model systems, we consider binary mixtures of lithium salts such as LiTFSI, $LiPF_6$, LiTFA, $LiBF_4$, and LiFSI. Using the graph-theory analysis implemented in CHAMPION, we identify the average proportions of different ionic species together with their life-times, which allows us to determine the structures contributing more—or less—to the ion transport. Ultimately the aim is to establish a robust and physically grounded description of the ion transport mechanism(s), to support future MSE design and optimization.

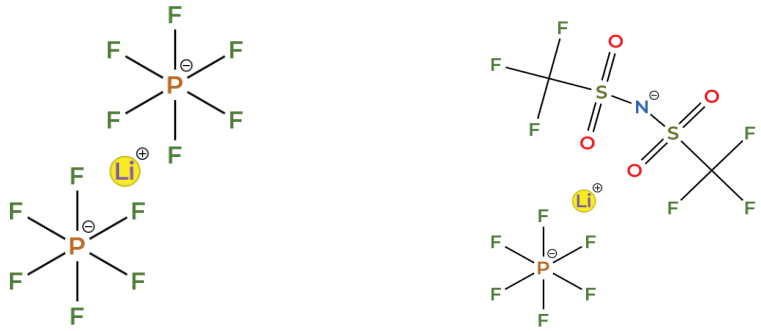

Figure 1. Two examples of dynamic structures in a binary MSE (1:1 LiTFSI:$LiPF_6$), a typical output from a CHAMPION analysis.

References

[1] H. Liu, X. Zhang, S. He, D. He, Y. Shang, H. Yu, *Materials Today.* (2022) 128-157.
[2] X. Tang, S. Lv, K. Jiang, G. Zhou, X. Liu, *J. Power Sources*, 542 (2022) 231792.
[3] M. Vu, P. Mirmira, R. P. Gomes, E. Doyle, H. Srinivasan, C. Amanchukwu, *Matter* (2023) 4357-4375.
[4] R. Andersson, F. Årén, A. A. Franco, P. Johansson, *J. Comp. Chem.*, 42 (2021) 1632-1642.
[5] F. Årén, R. Andersson, A. A. Franco, P. Johansson, *J. Electrochem. Soc.*, 170 (2023) 060506.

New Design Principle for Proton Conductive Organic Electrolytes Based on Activation of Surface Proton Hopping Conduction Mechanism

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Proton conductive polymer electrolytes are key materials for realizing a sustainable world owing to their versatile applications in fuel cells, bio-devices, sensors, catalysts, and other areas. The proton conductivity of these electrolytes influences device performance, so developing membranes with higher proton conductivity are essential. Generally, proton transport in polymer electrolytes can be explained using 1D or 2D water nanochannel models surrounded by hydrophobic walls. Generally, the Grotthuss mechanism in which proton migrate via cleavage and formation of hydrogen bonding networks of water molecules in the free water region is considered to be dominant in the water nanochannel. However, despite their high conductivity, their proton conductivity drastically decreases when free water freezes at low temperature and evaporates at high temperature. Moreover, their fluoropolymer-based composition poses environmental compatibility concerns. To develop the polymer electrolyte that does not rely on the Grotthuss mechanism, we have focused on the surface proton hopping conduction (SPHC) mechanism, in which a proton hops from a sulfonate group to its neighboring one on the surface of the hydrophobic walls. This mechanism has typically been regarded as trivial mechanism due to its extremely small diffusion coefficient.[1] Our idea is that a high-density alignment of sulfonate groups should enable fast proton conduction via an extremely-activated SPHC mechanism.[2] In this study, we have used the self-assembly of discotic liquid-crystalline (LC) molecules to form such an unusual situation where sulfonate groups form densely aligned on the surface.

We designed and synthesized a discotic LC molecule, named **TPES**, bearing sulfonate groups on the periphery of a tetraphenylethylene core (Figure 1a). **TPES** self-assembled into a hexagonal columnar structure in which the sulfonate groups are densely aligned with the distance of ca. 5 Å in the presence of appropriate amounts of water (Figure 1b). **TPES**/H_2O mixtures showed a maximum conductivity of 3.5 × 10^{-1} S cm^{-1} at 30 °C, which is extremely higher than that of the conventional polymer electrolytes, such as Nafion. Furthermore, the activation energy for proton conduction in the **TPES**/H_2O mixtures was estimated to be 6.0 kJ mol^{-1}, which is notably smaller than that of the Grotthuss mechanism. Analysis of the proton conduction mechanism indicates that the fast proton transport in the **TPES**/H_2O mixtures is achieved solely via the SPHC mechanism (Figure 1c).[3] The dynamics of water molecules involved in this mechanism was directly observed using quasi-elastic neutron scattering (QENS) measurements. This finding is expected to provide a foundation of a key strategy for creating next-generation proton conductive polymer electrolytes.

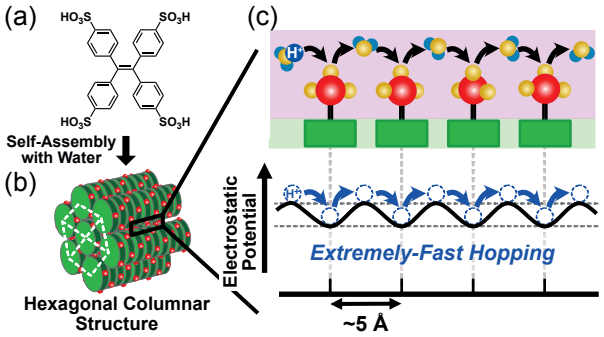

Figure 1. (a) Chemical structure of **TPES**. (b) Self-assembled structure of the **TPES**/H_2O mixtures. (c) Schematic image of the activated SPHC mechanism in the **TPES**/H_2O mixtueres.

References

[1] M. Eikerling, and A.A. Kornyshev, *J. Electroanal. Chem.*, **502** (2001), 1–14.
[2] T. Ichikawa, T. Yamada, N. Aoki, K. Suda, and T. Kobayashi, *Chem. Sci.*, **15** (2024) 7034–7040.
[3] M. Nakazawa, T. Yamada, Y. Tang, X. Zeng, and T. Ichikawa, *Under Revision*.

Dynamics & Mechanical Properties of Double-Network Ionogels

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Ionogels, i.e. gels based on ionic liquids (ILs), have been of significant research interest in recent years, since these materials show desirable properties for possible electrochemical applications, such as mechanical, electrochemical and thermal stability, as well as ionic conductivity. Furthermore, these properties are tunable by the chosen IL, the type of gelator and the overall composition of the gel. Possible host networks are of physical or chemical nature, the former describing gelation based on non-covalent interactions and the latter by covalent interactions. While physical networks demonstrate lower mechanical strength and a small impact on ion mobility, chemical networks increase the mechanical strength but also lower the mobility of ions.[1, 2, 3]

This work focusses on the combination of both gelation mechanisms to form double-network (DN) ionogels, hence combining the advantages of physical and chemical networks and confining the IL in a solid, but flexible matrix. As a physical gelator an oxamide-based low-molecular-weight gelator (LMWG) is used, while a polymer (either Bis-GMA:TEGDMA 60:40 or PEGDA) is utilized to form the chemical network. To investigate the influence of varying polymer concentrations as well as LMWG concentrations on the dynamical and mechanical properties of the ionogels, pulsed field gradient NMR (PFG-NMR) diffusion experiments and rheological measurements are employed. Further insight into the structure and electrochemical properties is gathered by scanning electron microscopy (SEM) and impedance spectroscopy (IS) measurements. Moreover, Li^+ is introduced to the ionogel matrices, leading to a significant impact on the structural and dynamic characteristics of the systems.

The results show that each ionogel component influences specific physicochemical properties, resulting in synergistic behaviour of the combined polymer and gelator networks. In the interpenetrating DN the LMWG concentration has only minor impact on ion transport. PEGDA-based DNs provide better stability than Bis-GMA:TEGDMA and yield stable ionogels. A lower polymer content is proven to be more beneficial, leading to gels with the best charge transport. Overall this study highlights the potential to tailor ionogels for specific functional applications.

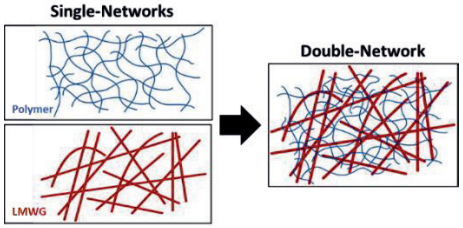

Figure 1. Illustration of different investigated host networks for ionogels. Two single network (SN) gelators are combined to form a double-network (DN) ionogel.

References

[1] A. Šantić, M. Brinkkötter, T. Portada, L. Frkanec, C. Cramer, M. Schönhoff, A. Moguš-Milanković, *RSC Adv.*, **10** (2020) 17070-17078.
[2] P. C. Marr, A. C. Marr, *Green Chem.*, **18** (2015), 105-128.
[3] X. Fan, S. Liu, Z. Jia,J. J. Koh, J. C. C. Yeo, C.-G. Wang, N. E. Surat'man, X. J. Loh, J. Le Bideau, C. He, Z. Li, T.-P. Loh, *Chem. Soc. Rev.*, **52** (2023) 2497-2527.

Designing Stable 5 V Cathode Compositions for Safe Solid-State Lithium Metal Batteries

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The development of safe and high-energy-density batteries is essential to meet the growing demands of the energy storage market. From a safety perspective, polymer electrolytes have been investigated for several decades as promising alternatives to conventional liquid electrolytes. However, when polymer electrolytes are used, the cathode formulation must be adapted to meet new requirements. In particular, the commonly used polyvinylidene fluoride (PVDF) binder in conventional cathodes must be replaced by an ionically conductive polymer composition. Nevertheless, most of the polymer systems proposed so far show limited electrochemical stability at high voltages (e.g., with $LiNi_{0.5}Mn_{1.5}O_4$, LNMO), and therefore most studies remain restricted to LFP or NMC-based chemistries.

To increase the energy density of solid-state batteries, the catholyte (ionically conductive binder[1]) composition has been optimized within the Horizon Europe project HyLiST (Grant Agreement No. 101147688[2]). The developed systems aim to simultaneously fulfill three critical requirements: stability at voltages up to 5 V, sufficient ionic conductivity, and adequate binding properties to ensure electrode integrity. Single-ion conducting polymer electrolytes have been selected to minimize cell polarization, as the anion is covalently anchored to the polymer backbone.[3] However, these polymers are typically rigid materials, which can limit both ionic conductivity and binding performance, making them challenging candidates for competitive catholytes.

In this dissemination the role played by various single-ion conductive polymer electrolytes and plasticizers will be discussed in meeting the three key parameters. Preliminary electrochemical cycling results using LNMO as the cathode active material will also be presented.

References

[1] A. Santiago, E. Sanchez-Diez, U. Oteo, I. Aldalur, M. Echeverría, M. Armand, M. Martinez-Ibañez, H. Zhang, **Small**, 18 (2022), 2202027

[2] https://hylist.eu/

[3] H. Zhang, C. Li, M. Piszcz, E. Coya, T. Rojo, L. M. Rodriguez-Martinez, M. Armand, Z. Zhou, ***Chem. Soc. Rev.***, (2017), 46, 797-815

Optimization of Inorganic-Polymer Hybrid Electrolytes by Rheology-Guided Extrusion for Solid-State Batteries

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Non-flammable, solid-state electrolytes have the potential to dramatically improve safety and performance of lithium batteries. The main objective in solid-state lithium batteries is to achieve high-performance electrolytes and electrodes at room temperature [1]. However, another concern in the battery community is the sustainability of the fabrication processes that are used to make these components. Extrusion processing is a sustainable process that saves cost, energy, and waste disposal in battery field. In electrolyte extrusion process, the ion transport properties are governed by microstructure and viscoelastic response, which will influence in the battery performance [2]. Nevertheless, the rheological effect of hybrid electrolytes on the extrusion parameters selection has never been studied so far, making a try and error approach their optimization. This work focuses on the development of a novel rheology-guided extrusion approach for high-performance inorganic-polymer hybrid electrolytes. Li_3InCl_6 (LIC) is used as inorganic part together with non-polar homopolymers (PIB and LDPE) to get solid electrolytes. The hybrid electrolytes are obtained by mixing the inorganic and the polymer in the extruder and the relationship between extrusion temperatures (from 130 to 200 °C), viscosities and ionic conductivities is studied.

References

[1] Daems, K.; Yadav, P.; Dermenci, K. B.; Van Mierlo, J.; Berecibar, M. Advances in Inorganic, Polymer and Composite Electrolytes: Mechanisms of Lithium-Ion Transport and Pathways to Enhanced Performance. *Renew. Sustain. Energy Rev.* **2024**, *191*, 114136. https://doi.org/10.1016/j.rser.2023.114136.

[2] Froboese, L.; Sichel, J. F. V. D.; Loellhoeffel, T.; Helmers, L.; Kwade, A. Effect of Microstructure on the Ionic Conductivity of an All Solid-State Battery Electrode. *J. Electrochem. Soc.* **2019**, *166* (2), A318–A328. https://doi.org/10.1149/2.0601902jes.

Temperature dependence of molecular dynamics and ionic conductivity in hydrated ionic liquids showing LCST-type phase transition behavior

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Ionic liquids (ILs) are organic salts that exist in liquid states below 100 °C. ILs have attracted significant attention owing to their unique physicochemical properties such as flame retardancy and non-volatility along with high ion density and ionic conductivity.[1] These characteristics can be changed by ion structures and combinations of cations and anions. We have found that a series of phosphonium-based ILs exhibit lower critical solution temperature (LCST) type phase transition behavior in water (Fig. 1).[2] Some phosphonium-based ILs (Fig. 2) are miscible with water at room temperature, however, they become immiscible with water upon heating, leading to the formation of two-phase separated states. This behavior can be applied for various applications, such as drug delivery and protein extraction. To control phase transition temperature, adjustments of the water content, modifications of the ionic structures, and the addition of organic compounds[3] have been employed. A leading theory regarding the mechanism of LCST-type phase transition behavior suggests that dehydration via the alkyl chains is involved. As the solution temperature rises, the mobility of the alkyl chains of the phosphonium cations increases. It leads to the decrease of hydration ability, inducing liquid-liquid phase separation. However, the detailed change in molecular dynamics and mechanism remain unclear. Therefore, in this study, we investigated ionic conductivity, diffusion coefficients and dynamics for aqueous solutions of **[P_{4444}][TsO]** (**[P_{4444}][TsO]$_{aq}$**) that exhibit LCST-type phase transition behavior. Furthermore, by examining the temperature dependence of these physicochemical properties, the relationship between phase transition and each of these parameters was investigated.

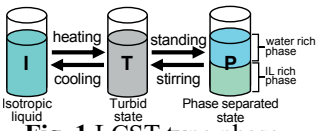

Fig. 1 LCST-type phase transition behavior of ILs in water.[1]

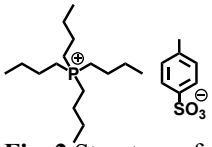

Fig. 2 Structure of phosphonium-based IL

[P_{4444}][TsO] was synthesized as previously reported[4]. A solution was prepared by mixing **[P_{4444}][TsO]** and water (100–X)/X (w/w). This solution is referred to as **[P_{4444}][TsO]$_{aq}$** (X wt%).

Ionic conductivity measurements were performed for **[P_{4444}][TsO]$_{aq}$** (X wt%). We found that ionic conductivity increased as the proportion of water in the solution increased (Fig. 3). Based on the ionic conductivity values, the diffusion coefficients of the component ions were estimated.

On the other hand, the diffusion coefficients of **[P_{4444}]** cation and **[TsO]** anion were separately evaluated using DOSY NMR. It was found that diffusion coefficients of **[TsO]** anion is larger than **[P_{4444}]** cation independent of water contents at room temperature. For example, for **[P_{4444}][TsO]$_{aq}$** (90 wt%), the diffusion coefficient of **[P_{4444}]** cation is 3.89 × 10^{-10} m^2/s while that of **[TsO]** anion is 5.48 × 10^{-10} m^2/s.

Dynamics of **[P_{4444}]** cation and **[TsO]** anion were exmined using quasi-elastic neutron scattering (QENS) measurement. QENS was performed on **[P_{4444}][TsO]$_{aq}$** (65 wt%) showing the lowest LCST phase transition temperature among the solutions with various water content X. Detailed results will be discussed in the presentation.

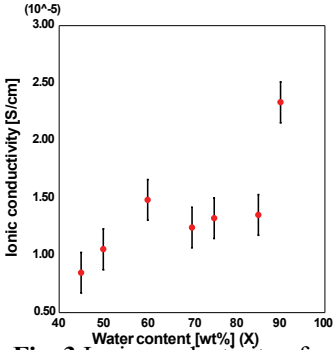

Fig. 3 Ionic conductivity of **[P_{4444}][TsO]$_{aq}$** (X wt%).

References

[1] M. Armand, F. Endres, D. R. MacFarlane, H. Ohno, B. Scrosati, *Nat. Mater.*, **2009**, *8*, 621–629.
[2] K. Fukumoto, H. Ohno, *Angew. Chem. Int. Ed.*, **2007**, *46*, 1852–1855.
[3] S. Yamaguchi, N. Nakamura, H. Ohno, T. Ichikawa, *Mol. Syst. Des. Eng.*, **2025**, *10*, 1024−1027.
[4] Y. Kohno, S. Saita, Y. Men, J. Yuan, H. Ohno, *Polym. Chem.*, **2015**, *6*, 2163–2178.

Gyroid Nanostructured Proton Conductive Polymer Membranes Transporting Proton Through Surface Hopping Conduction Mechanism

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Bicontinuous cubic (Cub_{bi}) phase is a class of nano-segregated liquid-crystalline phases in which both of two incompatible molecular parts form 3D continuous domains. To date, we have focused on ionic liquid crystals having zwitterionic headgroups.[1] For example, we designed and synthesized amphiphiles having pyridinium-type zwitterionic headgroups. It forms only layered smectic phases in the pristine states, however, it co-organized into Cub_{bi} liquid-crystalline phases in the presence of bis(trifluoromethane)sulfonimide (HTf_2N).[1] It is attributed to the formation of ion pairs between the pyridinium zwitterion part and HTf_2N through an ion exchange and the increase of the volume of the ionic parts. The Cub_{bi} liquid-crystalline assemblies have a hydrophilic gyroid minimal surface where sulfonate group sit on densely and periodically. When a suitable amount of water is incorporated into the Cub_{bi} liquid-crystalline assemblies, a 3D continuous water nanosheet is created, which function as proton conduction pathway.[1]

Based on the molecular design of the amphiphilic zwitterions, we have recently succeeded in the creation of a gemini-type amphiphilic zwitterion having polymerizable groups. It forms Cub_{bi} liquid-crystalline phases.[2,3] UV irradiation for the liquid-crystalline monomer in Cub_{bi} phases leads to the formation of self-standing and insoluble polymer membranes with preserving the gyroid nanostructures. The polymer membrane shows high proton conductivity in the order of 10^{-2} S cm^{-1} in the H_2O-absorbed condition. The proton conduction mechanism in the gyroid nanostructured polymer membranes has been found to be different from that in conventional proton conduction polymer membranes. The details of the proton conduction mechanism in the gyroid nanostructured polymer membranes will be discussed in the presentation.

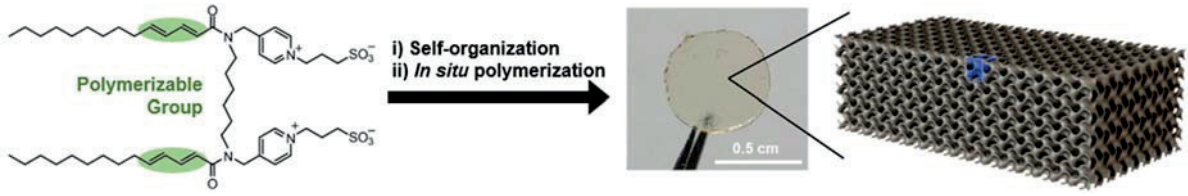

Figure 1. Creation of gyroid nanostructured polymer membranes through self-organization of polymerizable amphiphilic zwitterion and in situ polymerization.

References

[1] T. Ichikawa, T. Kato, H. Ohno, J. Am. Chem. Soc. 2012, 134, 11354.
[2] T. Kobayashi, Y. Li, A. Ono, X. Zeng, T. Ichikawa, Chem. Sci. 2019, 10, 6245.
[3] T. Ichikawa, T. Yamada et al., Chem. Sci. 2024, 15, 7034.

Safer sodium-ion batteries by non-flammable deep eutectic electrolytes

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As Europe continues its transition to renewable energy, the growing demand for energy storage raises significant concerns regarding safe supply of critical raw materials (CRMs), such as lithium, copper, and cobalt. Sodium-ion batteries (SIBs) offer a CRM-free alternative, but their electrolyte formulations still rely on highly flammable solvents and toxic components that also pose severe hazards should there be *e.g.* a thermal runaway.

Here, we present a comprehensive investigation of a non-flammable and non-toxic deep eutectic electrolyte (DEE) for SIBs utilizing urea and its derivatives combined with the salt sodium bis(fluorosulfonyl)imide (NaFSI). In addition, we use non-toxic additives and diluents to facilitate formation of stable electrolyte/electrode interfaces/interphases and to lower the electrolyte viscosity.[1-3]

First, the glass transition temperatures, ionic conductivities, and viscosities of the DEEs were determined to assess dynamic properties. Second, the electrochemical behavior of Na|DEE|Hard carbon half-cells and Hard carbon|DEE|Prussian White full cells was observed via galvanostatic cycling at ambient temperature. Finally, flammability tests were made to demonstrate the superior safety of DEEs *vs.* conventional electrolytes.

Overall, our findings indicate that DEEs offer a promising and inherently safer pathway for the development and operation of next-generation SIBs.

References

[1] N. Ito, T. Hosaka, M. Alhanash, T. Furuichi, R. Tatara, Z. T. Gossage, P. Johansson and S. Komaba, *ACS Appl. Energy Mater,* **9** (2026) 1377-1385.

[2] T. T. A. Dinh, T. T. K. Huynh, L. T. M. Le, T. T. T. Truong, O. H. Nguyen, K.T. T. Tran, M. V. Tran, P. H. Tran, W. Kaveevivitchai, and P. M. L. Le, *ACS Omega*, **5** (2020) 23843-23853.

[3] L. Mezzomo, N. Pianta, I. Ostroman, N. Aloni, D. Golodnitsky, E. Peled, P. Mustarelli, R. Ruffo, *J. Power Sources*, **561** (2023) 232746.

Interpenetrating Gel Polymer Electrolytes: Effects of PEO Structural Design

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Over the last decade, lithium-ion batteries (LIB) have expanded into high-performance applications such as electric vehicles and large-scale energy storage, owing to their high energy density, low weight, and long cycle life. However, the use of flammable organic liquid electrolytes presents significant safety concerns. In response, polymer electrolytes have emerged as safer alternatives, offering low flammability, enhanced thermal stability, and reduced leakage risk. These materials are generally classified into solid polymer electrolytes (SPE), gel polymer electrolytes (GPE), and composite polymer electrolytes (CPE). Particularly noteworthy are GPE which consist of a polymer matrix swollen with liquid solvents, lithium salts, and functional additives [1]. Ionic conduction primarily occurs through the liquid phase, while the polymer network provides mechanical integrity, maintaining a quasi-solid state and mitigating leakage hazards. Among the most studied polymer matrices are those based on polyethylene oxide (PEO), valued for their chain flexibility, electrochemical stability, low glass transition temperature (*Tg*), and high salt solubility. Nonetheless, their low room-temperature conductivity and limited mechanical and thermal robustness restrict their practical deployment [2].

In this study, polymers were synthesized via Atom Transfer Radical Polymerization (ATRP), using monomers with varying ethylene oxide side chains. GPEs were formulated with lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) as the salt, propylene carbonate (PC) as the solvent, and crosslinking agents. The materials were characterized using Differential Scanning Calorimetry (DSC), and Thermogravimetric Analysis (TGA). Ionic conductivity of the electrolytes was evaluated via Electrochemical Impedance Spectroscopy (EIS), using coin cells. Conductivity values on the order of 10^{-3} S/cm were obtained, surpassing those reported for PEO/LiTFSI systems at room temperature (10^{-5} S/cm). The most promising formulation, exhibiting superior thermal stability and conductivity, was further evaluated in lithium-metal cells, yielding a lithium transference number (tLi^{+}) of 0.8. Linear Sweep Voltammetry (LSV) confirmed electrochemical stability up to 4.4 V.

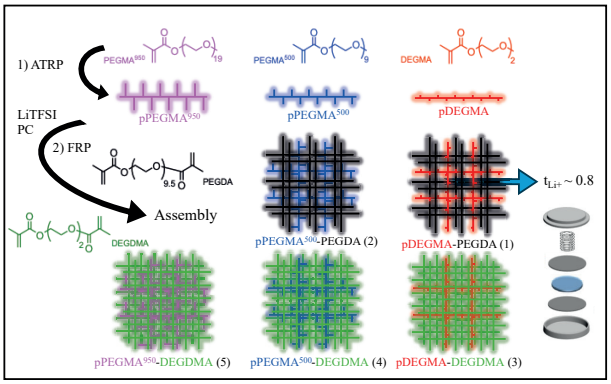

Figure 1. Schematic representation of the GPE prepared.

References

[1] D. Mecerreyes, *et al.*, *Macromolecular Chemistry and Physics*, **221** (2020) 1900490.

[2] D. Zhou, *et al.*, *Chem*, **5** (2019) 2326–2352.

Tuesday
(June 2nd)

Tuesday, June 2nd

Session 1, 9:00−10:30, Chairperson: Maria Forsyth

PL2 9:00-9:40	**Sandrine Lyonnard, CEA, France** Coupled ion-solvent-polymer dynamics revealed by Quasi Elastic Neutron Scattering
K3 9:40-10:10	**Stefano Passerini, Karlsruhe Institute of Technology/Nanchang University, Germany/China** Conformal Polymer Electrolyte Enabled by Nitrile Coordination for Long-Cycle Solid-State Lithium Metal Batteries
I5 10:10-10:30	**Lauréline Marchal, Arkema, France** New polymer materials for next-generation of batteries (SSB and ASSB)

Coffee Break (10:30−11:00)

Session 2, 11:00−12:55, Chairperson: Renaud Bouchet

K4 11:00-11:30	**Daniel Brandell, Uppsala University, Sweden** Ion transport in solid polymer electrolytes with active and passive fillers
I6 11:30-11:50	**Maryam Nojabaee, German Aerospace Center, Germany** Electrolytes for Calcium Metal Batteries: Opportunities and Challenges from Liquid to Polymer Systems
I7 11:50-12:10	**Maider Zarrabeitia, Institute of Materials Science of Seville-CSIC, Spain** Anion-Cation Chemical Engineering of Polymer Electrolytes for High Performance Potassium-Ion Batteries
O10 12:10-12:25	**Timofey Kolesnikov, Karlsruhe Institute of Technlogy, Germany** Influence of Salt Anions on Ionic Transport in Solid Polymer Electrolytes for Potassium Metal Batteries
O11 12:25-12:40	**Matteo Milanesi, Politecnico di Torino, Italy** Biopolymer-Based Gel Electrolytes for Stabilizing Zn Anodes in Aqueous Zn-Ion Batteries
O12 12:40-12:55	**Xuesong Ge, Chinese Academy of Science, France** Development of Polymer Electrolytes for High-Temperature Magnesium Metal Batteries

Lunch (13:00−14:30)

Session 3, 14:30−16:05, Chairperson: Guiomar Hernández

K5 14:30-15:00	**Masayoshi Watanabe, Yokohama National University, Japan** Double Conductivity Maxima in Polymer Electrolytes Complexed with Supercooled Salts: Distinct Transport and Electrochemical Properties
I8 15:00-15:20	**Heng Zhang, Huazhong University of Science and Technology, China** Tailoring Anion Chemistry for Better Rechargeable Batteries
O13 15:20-15:35	**Kazuhide Ueno, Yokohama National University, Japan** Molecular Design and Ion Transport in Lithium Ionic Liquid Electrolytes for Lithium Batteries
O14 15:35-15:50	**Manuela M. Silva, University of Minho, Portugal** Solid Bio Polymer Electrolytes for Advanced Electrochemical Applications
O15 15:50-16:05	**Jennifer Pringle, Deakin University, Australia** Zwitterionic additives for polymer electrolytes with high target ion transport

Coffee Break (16:05−16:30)

Session 4, 16:30−17:35, Chairperson: María Martinez

I9 16:30-16:50	**Gunther Brunklaus, Helmholtz Institute Münster, Germany** Hybrid-Polymer Electrolytes for Lithium Metal Batteries

O16	**Arnaud Prébé, University of Montreal, Canada**
16:50-17:05	Impact of processing parameters on the properties of thermoplastic vulcanizate (TPV) solid polymer electrolytes
O17	**Chengyin Fu, Battery Innovation Hub, Switzerland**
17:05-17:20	Initiator-driven in situ formation of gradient polymer electrolyte for high performance lithium metal batteries
O18	**Yuta Maeyoshi, Research Institute of Electrochemical Energy-AIST, Japan**
17:20-17:35	Effects of Cationic Polymer Coatings on Lithium Plating/Stripping Reactions

Poster Session 2, 17:35−19:30 (Miramar Palace-Ground Floor)

Coupled ion-solvent-polymer dynamics revealed by Quasi Elastic Neutron Scattering

Sandrine Lyonnard[1,2,3]

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Ionic transport in polymer-based electrolyte materials underpins the performance of electrochemical energy conversion and storage devices as fuel cells and batteries. However, the fundamental mechanisms governing ion mobility remain incompletely understood, particularly due to the interplay between ionic motion, polymer dynamics, and solvent organization across multiple scales. In this lecture, I will show how quasi-elastic neutron scattering (QENS) provides a unique window into these coupled processes [1], capturing molecular dynamics on picosecond-to-nanosecond timescales and complementing techniques as NMR, electrochemical impedance spectroscopy, total scattering and molecular dynamics simulations [2-3].

In fuel cell membranes, the structure-hydration-transport coupling dictates ion mobility [4-6]. Using a radiation-grafted anion exchange membrane platform in both Cl^- and OH^- forms, we disentangled how ion identity reorganises confined water and, in turn, how this reorganisation controls transport pathways, dictating membrane performance beyond simple electrostatic effects [5]. Using graphitic additives in Nafion, we also induced transformations in nanoscale phase separation, mesoscale connectivity and molecular-scale diffusion mechanisms, revealing the role of connected hydrogen-bond networks [6].

In lithium-based polymer electrolytes, ion transport is often coupled to polymer segmental dynamics (as in PEO) [7], but a more complex picture emerges due to coordination exchange mechanisms dominating over simple ion hopping. Moreover, the balance between polymer-mediated and ion-mediated transport can evolve with salt concentration, polymer structure and chemistry [8-9]. For instance, we will show how the strength of ion–polymer coordination critically influences local mobility and macroscopic conductivity, challenging simplified transport models [10-11].

By bringing together these considerations, this lecture will identify the fundamental mechanisms that control ion transport in polymer systems and outline strategies for designing high-performance electrolytes.

References

[1] F. Foglia et al., J. Phys.: Condens. Matter 2021, 33 264005.
[2] Q. Berrod et al., Macromolecules 2015 48 (17), 6166-6176.
[3] Q. Berrod et al., Scientific Report, 2017, 7, 8326.
[4] F. Foglia et al. Nature Materials, 2022, 21(5):555-563.
[5] K. Smith et al., The critical role of water structure in anion transport, in review.
[6] K. Smith et al., Multiscale Design Rules for Additive-Engineered PFSA Membranes: Linking Hydration Structure to Proton Transport and Durability, in review.
[7] D. Bresser et al., Mol. Syst. Des. Eng. 2019,4, 779-792.
[8] N. J Shah et al. Nature Materials 2024 May;23(5):664-669.
[9] R. Andersson et al., ChemElectroChem 2024, 11, e202400415
[10] H. Gudla, et al. ACS Appl. Polym. Mater. 2025, 7, 4716−4724.
[11] S. Lyonnard, Q. Berrod, M. Andersson, D. Brandell, J. Mindemark et al, Molecular-scale dynamics in PEO and PCL, in preparation.

Conformal Polymer Electrolyte Enabled by Nitrile Coordination for Long-Cycle Solid-State Lithium Metal Batteries

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Abstract

Lithium metal is a highly promising anode for next-generation high-energy-density batteries due to its high theoretical capacity, yet its practical application remains hindered by poor interfacial compatibility with polymer solid-state electrolytes (PSEs). Herein, an in-situ solidification PSE that utilizes poly(ethyleneglycol)methyletheracrylate (PEGMEA) and methylated pivalonitrile (PN) was developed (PNF), which forms a conformal and mechanically robust solid electrolyte interphase (SEI) on lithium metal surface. The coordination between the nitrile group (–C≡N) and Li^+ regulates interfacial ion transport, while the formed organic-inorganic (hybrid) SEI effectively combines mechanical flexibility and interfacial rigidity to buffer lithium volume fluctuations and inhibits dendrite growth. Benefiting from the enhanced Li^+ hopping sites and improved ionic mobility, the PNF electrolyte exhibits high ionic conductivity, i.e., 3.47×10^{-4} S cm^{-1} at 30°C. Li|PNF|Li symmetric cells show exceptional cycling stability, surpassing 1000 hours at 0.5 mA cm^{-2}. Notably, Li|PNF|$LiFePO_4$ cells achieve a capacity retention of 92.8% after 1000 cycles at 0.5C and 78.9% after 2000 cycles at 1C rate, both at 30 °C, highlighting the exceptional conformal properties of the electrolyte resulting in the superior cycling performance.

Acknowledgments

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References

L. Huang, L. Lan, Y. Wu, X. Ao, N. Zhou, Y. Zhou, C. Peng, Y. Liu, S. Fang, S. Passerini *Conformal Polymer Electrolyte Enabled by Nitrile Coordination for Long-Cycle Solid-*

State Lithium Metal Batteries Adv. Funct. Materials 36, (2026): e29705. https://doi.org/10.1002/adfm.202529705

New polymer materials for next-generation of batteries (SSB and ASSB)

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The rapid electrification of mobility and industrial applications, combined with the emergence of next generation batteries (semisolid, all-solid-state with the target to use lithium metal), is driving a growing demand for advanced polymer materials that can simultaneously enhance cell performance, safety, and sustainability. Arkema is developing a comprehensive portfolio of solutions—including polymer binders, gel polymer electrolytes (GPEs), and hybrid polymer/ceramic systems—to support the transition toward low impact manufacturing processes and high energy battery architectures.

In this presentation, we present (i) the development of **ex situ and in situ GPEs** formulated with polymers, ionic liquids, monomers, and thermal or photochemical initiators and (ii) **hybrid polymer/ceramic composites** compatible with sulfide- and oxide-based solid electrolytes.

In situ polymerization technologies for GPEs offer several key advantages: solvent-free (100% solids), shorter SEI formation time, excellent compatibility with standard cell-assembly processes, and ionic conductivities approaching those of liquid electrolytes. The first ex-situ GPE membranes developed show conductivities above 1 $mS.cm^{-1}$ at 25 °C and strong thermal stability without exudation. Hybrid approaches demonstrate the ability of polymers to improve mechanical flexibility, reduce the pressure required during cell assembly and operation, and maintain lithium-ion transport in solid electrolytes.

These results highlight the crucial role of polymers in enabling high-energy, safer, and more sustainable next-generation batteries. The work presented illustrates Arkema's advanced material solutions designed to meet the requirements of future manufacturing processes and emerging battery chemistries.

Ion transport in solid polymer electrolytes with active and passive fillers

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It is well-known that micron- and nano-sized ceramic fillers can boost the electrolyte in solid polymer electrolytes (SPEs), with notable early examples being Al_2O_3, SiO_2 and TiO_2. Traditionally, this has argued to be due to the reduced crystallinity in polymer matrices such as poly(ethylene oxide) (PEO). However, more recent discoveries have challenged this picture, where fully amorphous polymer hosts such as poly(trimethylene carbonate) (PTMC) has also experienced a similar phenomenon of increased conductivity – but which seems to be inherently dependent on the type of filler, despite them being intrinsically non-conductive and thereby *passive* [1]. The inclusion of passive fillers also appears to have a profound impact on important ionic transport properties such as transference. Parallel to this development has several studies during the recent decade used ion-conductive – *active* – fillers in SPE materials, without being able to truly utilize the higher ionic conductivity of the ceramic phase [2]. To interpret these often highly contradictory results, there is a need to better understand the ionic transport in these complex systems. Here, Molecular Dynamics (MD) modelling studies have recently shown helpful [3]. In this work, we present recent progress in systems comprising various forms of polymer hosts and both active (e.g. $Li_{6.7}Al_{0.3}La_3Zr_2O_{12}$; LLZO) and ceramic fillers (e.g. γ-$LiAlO_2$), studied through modelling and experimental characterization. We also discuss their applicability in Li-metal batteries.

References

[1] K. Elbouazzaoui, A. Mahun, V. Shabikova, L. Rubatat, K. Edström, J. Mindemark, D. Brandell, *Adv. Energy Mater.,* **15** (2025) 2405249.

[2] K. Elbouazzaoui, F. Nkosi, D. Brandell, J. Mindemark, K. Edström, *Electrochim. Acta,* **462** (2023) 142785

[3] M. Kozdra, L. Hölzer, D. Brandell, A. Heuer, *Phys. Chem. Chem. Phys.,* **27** *(*2025) 24918.

Electrolytes for Calcium Metal Batteries: Opportunities and Challenges from Liquid to Polymer Systems

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Calcium metal batteries are increasingly considered as a promising alternative to lithium-based systems due to their high volumetric capacity, improved safety, and the natural abundance of calcium. However, their cycle life remains limited by irreversible reactions and the sluggish kinetics of divalent ions. The strong coordination of Ca^{2+} reduces ion mobility and leads to sluggish diffusion kinetics, hindering charge transport and transfer [1]. In addition, interfacial effects, particularly the formation of the solid electrolyte interphase (SEI) due to electrolyte decomposition, contribute to capacity loss and can lead to rapid electrochemical inactivity.

In this contribution, we discuss the behavior of different calcium electrolyte systems, ranging from conventional liquid electrolytes to gel/polymer-based approaches, with the aim of identifying both opportunities and limitations across these concepts. Particular attention is given to how electrolyte composition and ion coordination influence ion mobility, polarization behavior, and interfacial stability. To this end, transport parameters such as ionic conductivity, diffusion coefficients, and cation transference numbers are investigated and discussed along with interfacial processes governing calcium deposition and dissolution. Starting from established systems such as $Ca[B(hfip)_4]_2$ in glyme-based solvents, different salts combined with a range of solvents and polymer backbones are explored to understand the role of anion chemistry and solvation structure [2]. Here symmetrical Ca‖Ca cells are used to probe plating and stripping behavior through polarization experiments and electrochemical impedance spectroscopy with adapted protocols [2,3].

We further discuss the challenges associated with this different electrolyte classes, including trade-offs between stability and transport in liquid systems, as well as limited Ca^{2+} mobility in polymer-based materials. Overall, this work provides a comparative perspective on electrolyte concepts for calcium metal batteries and highlights the key factors that must be controlled to enable more stable and reversible cycling.

References

[1] Z.Li, S.Cui, J.Häcker, M.Nojabaee, M.Fichtner, G.Cui, Z.Zhao-Karger, Angew. Chem. Int. Ed.2025, 64, e202415942. https://doi.org/10.1002/anie.202415942
[2] J.Häcker, T.Rommel, L.Rademacher, S.Riedel, Zh. Zhao-Karger, J. A. Blázquez, K.A. Friedrich, M. Nojabaee Adv. Sci.12, no. 33 (2025): 12, e06498. https://doi.org/10.1002/advs.202506498
[3] Häcker. J; Rommel, T; Lange, P; Kampmann., F; Remmlinger, J; Zhao-Karger, Zh; Friedrich. A; Nojabaee. Journal of Magnesium and Alloys, 2025, 13,6, https://doi.org/10.1016/j.jma.2025.04.001.

Anion-Cation Chemical Engineering of Polymer Electrolytes for High-Performance Potassium-Ion Batteries

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The European Union has established a policy to achieve climate neutrality by 2050 [1]. Renewable energy sources should be drastically increased, as well as energy storage devices, particularly batteries, which are the key technology to achieve the target. Sodium-ion batteries (SIBs) are postulated as a complementary technology to lithium-ion batteries (LIBs) for light electromobility and stationary applications, owing to their lower cost and greater sustainability, as they are made of non-critical raw materials [2]. However, alternative batteries based on other chemistries, while maintaining sustainability and cost, should be investigated and developed to cover diverse applications. Potassium-ion batteries (PIBs) might be one of the next possible chemistries due to K, as Na is abundant and widely distributed on the Earth's crust, exhibits lower reduction potential than Na (i.e., -2.71 V and −2.93 V vs. SHE) and faster K ion diffusion in liquid than Li and Na, with the possibility of providing higher power and energy density [3]. In fact, the Group 1 start-up announced plans to produce PIBs at large scale by 2027 [4]. Unfortunately, PIBs' current performance is inferior to that of both LIBs and SIBs. Therefore, further advances are necessary to enhance the viability of PIBs.

Liquid electrolytes are the conventional electrolytes for PIBs due to their excellent performance in LIBs and SIBs. Although they exhibit high ionic conductivity at RT, they suffer from (usually) low thermal stability, high volatility, and high flammability. In addition, as occurs in SIBs, the electrolyte selection for K-technology is more critical, mainly due to the poor properties of the formed electrode-electrolyte interphases. Therefore, developing advanced electrolytes, *i.e*., solid-state electrolytes (SEs), that are more electrochemically stable and safer, is crucial to enhance the performance of PIBs. Polymer electrolytes have attracted considerable interest for next-generation batteries due to their good mechanical properties, flexibility, and safety [5]. In general, the polymer electrolytes exhibit low ionic conductivity at RT due to their inherent poor chain flexibility. Incorporating ionic liquids (ILs) as plasticizers significantly enhances ionic conductivity while ensuring thermal stability and safety.

This work focuses on the design and electrochemical characterization of K-based polymer electrolytes by optimizing the anion & cation chemistry of the K salt and the IL and by thoroughly investigating their influence on the structural, thermal and electrochemical properties [6,7]. The best-performing polymer electrolyte was further optimized by adding an additive to enhance oxidation stability, and its oxidation stability was characterized using *operando* X-ray photoelectron spectroscopy at the HIKE endstation of BESSY II facilities. Finally, the optimized electrolyte was validated by fabricating pouch cells, which delivered a capacity retention of 93.6% and an average Coulombic efficiency of 99.6% over 200 cycles at 0.1C.

References

[1] https://setis.ec.europa.eu/implementing-set-plan-2020-report-2020-11-23_en
[2] I. Hasa, S. Mariyappan, D. Saurel, P. Adelhelm, A.Y. Koposov, C. Masquelier, L. Croguennec, M. Casas-Cabanas, *J. Power Sources*, **482** (2021) 228872.
[3] M. Zarrabeitia, J. Carretero-Gonzalez, M. Leskes, H. Adenusi, B. Iliev, T. J. S. Schubert, S. Passerini, E. Castillo-Martinez, *Energy Mater.* **3** (2023) 300046.
[4] www.energy-storage.news/potassium-ion-batterystartup-group1-lfp-is-our-benchmark/
[5] I. Osada, H. de Vries, B. Scrosaty, S. Passerini, *Angew. Chem. Int. Ed*. **55** (2016) 500.
[6] J. Chen, S. Ebrahimi-Koodehi, B. Iliev, Y. Steinberg, M. Leskes, T. J. S. Schubert, E. Castillo-Martínez, D. Bresser, E. Kataev, M. Zarrabeitia, *J. Mater. Chem. A* **13** (2025) 25476.
[7] J. Chen, Y. Zhou, N. Martin Hernández, B. Iliev, T. J. S. Schubert, E. Castillo-Martínez, M. A. Muñoz-Márquez, D. Bresser, E. Kataev, M. Zarrabeitia (submitted)

Influence of Salt Anions on Ionic Transport in Solid Polymer Electrolytes for Potassium Metal Batteries

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The transition beyond lithium-based energy storage systems require alternative chemistries that combine resource abundance, low cost, and high energy density. Potassium metal batteries (PMB) are a promising candidate, but their practical application is limited by the instability and safety issues of conventional liquid electrolytes, primarily due to uncontrolled interfacial reactions and unstable solid–electrolyte interphases (SEI). In this context, solid polymer electrolytes can offer important benefits over liquid electrolytes, including non-flammability, the absence of leakage, and improved mechanical robustness, which together can enhance the overall safety and stability of PMB. In this work, we systematically investigate the influence of different potassium salt anions on the properties and performance of poly(ethylene oxide) (PEO) based solid polymer electrolytes (SPEs) for PMB. Three fluorinated salts – potassium bis(fluorosulfonyl)imide (KFSI), potassium bis(nonafluorobutanesulfonyl)imide (KNFBSI), and potassium 1,1,2,2,3,3-hexafluoropropane-1,3-disulfonimide (KHFPSI) – were combined with high-molecular-weight PEO over a broad composition range (EO:K = 20:1–12:1). The chemical structures of the salts are shown in Figure 1.

The SPEs were characterized by differential scanning calorimetry and X-ray diffraction to investigate how different anions influence the polymer structure at various temperatures. All systems exhibited a gradual reduction in crystallinity with increasing salt content, while KHFPSI enabled nearly complete amorphization of PEO at high salt loadings. The optimized compositions demonstrated ionic conductivities on the order of 10^{-3} S·cm^{-1} at 60–70 °C, comparable to state-of-the-art KTFSI-based systems. In contrast, SPEs containing KNFBSI exhibited significantly reduced conductivity at room temperature, which is attributed to the formation of additional crystalline salt–polymer phases.

Pulsed-field gradient nuclear magnetic resonance (PFG-NMR) measurements further confirmed the presence of such crystalline phases in KNFBSI-based SPEs, which hinder ion transport. Linear sweep voltammetry demonstrated that KNFBSI provides the highest oxidative stability, while KFSI shows the lowest stability but forms beneficial decomposition products that contribute to SEI formation.

The possibility of cycling was evaluated in K|SPE|$K_2Fe[Fe(CN)_6]$ half-cells at 55 °C. Despite its lower intrinsic stability, KFSI enabled long-term cycling with discharge capacities of ~90 mAh·g^{-1} over 200 cycles, indicating the formation of a stabilizing SEI on potassium metal. SPEs based on KNFBSI and KHFPSI showed improved electrochemical stability and competitive rate performance, highlighting the crucial role of anion structure in balancing polymer morphology, ionic transport, and interfacial processes.

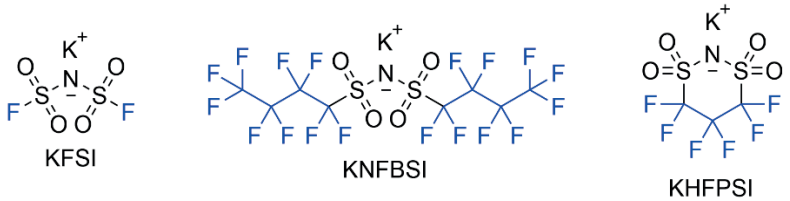

Figure 1. Chemical structures of KFSI, KNFBSI and KHFPSI.

Biopolymer-Based Gel Electrolytes for Stabilizing Zn Anodes in Aqueous Zn-Ion Batteries

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The Green Deal drives the need for safe, sustainable, and cost-effective energy storage systems. Aqueous Zn-ion batteries are promising due to their safety, low cost, and high ionic conductivity, however their performance is limited by Zn anode issues such as poor plating/stripping reversibility, non-uniform deposition, and parasitic reactions. In particular, the hydrogen evolution reaction (HER) reduces coulombic efficiency and accelerates degradation. Gel polymer electrolytes (GPEs) have emerged as a promising strategy to mitigate these limitations [1]. By combining the high ionic conductivity of liquid electrolytes with the mechanical stability of polymer networks, GPEs can promote more homogeneous Zn deposition, mitigate interfacial instability, and suppress parasitic reactions such as HER [2]. Agarose-based GPEs containing simple $ZnSO_4$ electrolytes, prepared in this work, demonstrate higher efficiency than the corresponding liquid electrolyte, a wider electrochemical stability window (2.2 V), and markedly improved plating/stripping stability, reaching thousands of hours. Differential electrochemical mass spectrometry (DEMS) was used to directly monitor gas evolution during operation. It enabled the evaluation and quantification of HER, highlighting the ability of agarose-based GPEs to suppress hydrogen evolution and improve Zn anode stability [3], an utmost important result for the future development of rechargeable Zn-based batteries.

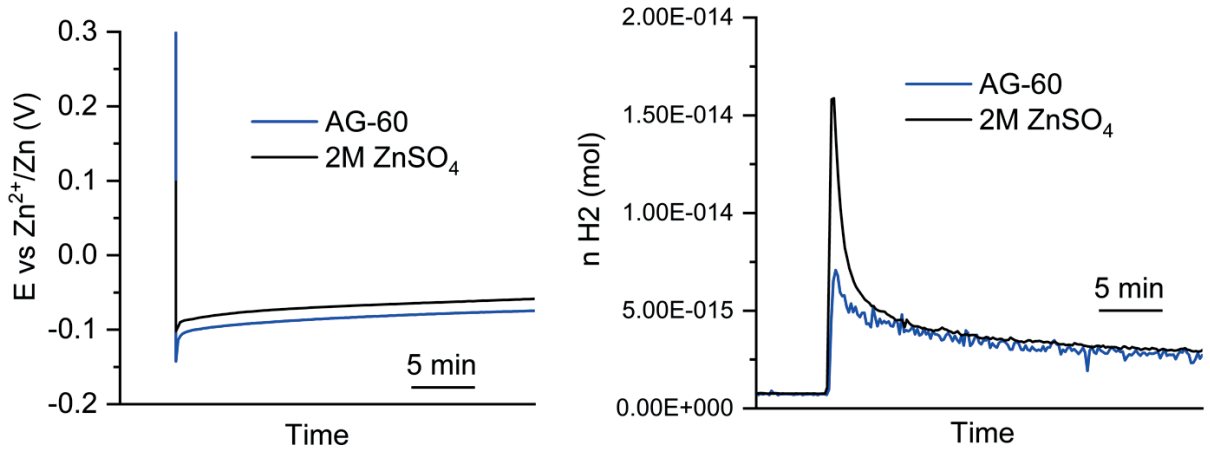

Figure 1 a) Potential vs time behavior during zinc deposition. b) H_2 evolution monitored by DEMS during zinc deposition.

Acknowledgements

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References

[1] Peng et al., ACS Nano **18** (2024) 21779–21803
[2] Milanesi et al., Battery Energy **5** (2026) e70060
[3] Rana et al., Adv. Energy Mater. **15** (2025) e03155

Development of Polymer Electrolytes for High-Temperature Magnesium Metal Batteries

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Magnesium metal batteries (MMBs), with their pronounced superiority in high safety, high volumetric capacity, and low cost, are emerging as promising next-generation energy storage systems for operation under elevated temperatures. The electrolyte and its interfacial chemistry are crucial for the development of high-temperature MMBs. We developed a series of high-temperature polymer electrolytes for MMBs through electrolyte structure design and interfacial chemistry regulation (Figure 1). First, a poly(alkoxide magnesium) single-ion conducting electrolyte was designed to introduce an anion-rectifying effect, which effectively suppresses anion-induced side reactions at the Mg metal anode and significantly improves the coulombic efficiency of Mg plating/stripping. To further enhance the mechanical robustness and thermal tolerance of the electrolyte, a self-standing polymer electrolyte was subsequently developed, enabling stable battery operation at temperatures up to 150 °C. Building upon interphase stabilization, we further proposed and developed a "polymer-dominated interfacial chemistry regulation" strategy. By introducing electron-withdrawing groups such as fluorinated moieties and quaternary ammonium units into the polymer framework, the LUMO energy level of the polymer is lowered, allowing it to preferentially decompose on the Mg metal surface. This process leads to the formation of polymer-derived organic–inorganic composite interphases enriched with MgF_2 and Mg_3N_2 species, which effectively stabilize the electrode–electrolyte interface under elevated temperatures. As a result, RMBs based on these polymer electrolytes can demonstrate stable cycling over 200 cycles at 150 °C with 80% capacity retention. The above studies provide a new materials platform and mechanistic basis for energy storage systems designed for extreme environments.

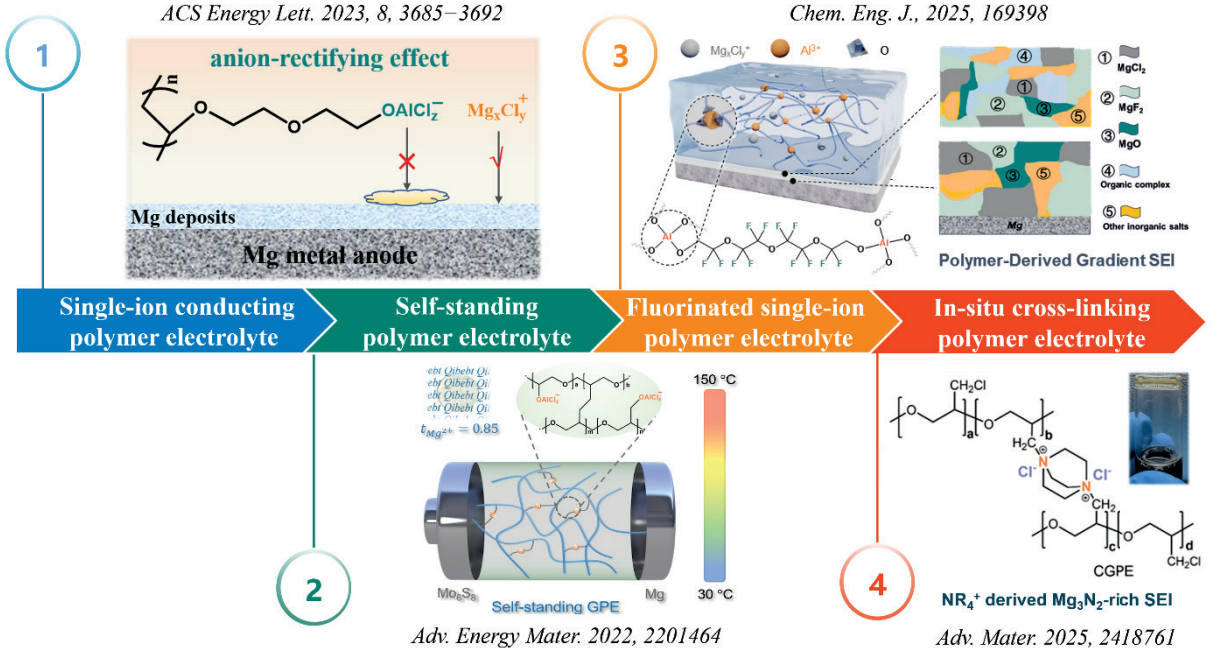

Figure 1 Polymer electrolytes for high-temperature magnesium metal batteries.

Double Conductivity Maxima in Polymer Electrolytes Complexed with Supercooled Salts: Distinct Transport and Electrochemical Properties

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Ionic conductivity in conventional polymer electrolytes is generally assumed to exhibit a single maximum as a function of salt concentration, reflecting an optimal balance between ion dissociation and segmental mobility. Here, we report a qualitatively different behavior: the emergence of two distinct conductivity maxima arising from fundamentally different ion coordination environments. We show that an equimolar mixture of $Li[N(SO_2F)_2]$ (LiFSA) and $Li[N(SO_2CF_3)_2]$ (LiTFSA) forms a stable supercooled liquid ($Li[FSA_{0.5}TFSA_{0.5}]$) at room temperature [1], characterized by a low glass transition temperature (T_g = 245 K) and single-cation transport (t_{Li^+} = 1 under anion-blocking conditions). Incorporation of this supercooled salt into poly(ethylene oxide-*co*-propylene oxide) ($P(EO_{0.8}PO_{0.2})$)–based network polymers [2] over a wide composition range (weight fraction of the salt: W_{Li} = 0.1–0.8) reveals how ion coordination governs both bulk transport and interfacial electrochemistry.

The T_g of the polymer electrolytes exhibited a maximum at W_{Li} = 0.5 and increased again at higher salt contents (W_{Li} = 0.8–1.0) (**Figure 1(a)**). This behavior reflects a competition between Li^+ coordination by the polyether chains and plasticization by the low-T_g supercooled salt. At W_{Li} = 0.1–0.5, the coordination effect dominates (salt-in-polymer regime), whereas at W_{Li} = 0.5–0.8, the plasticization effect becomes predominant (polymer-in-salt regime) [3]. The resulting polymer electrolytes exhibit two conductivity maxima at W_{Li} = 0.2 and 0.8 (**Figure 1(b)**). Raman spectroscopy and MD simulations indicate full coordination of Li^+ by the polyether at low salt content, whereas ion aggregation and partial coordination by the polyether dominate at high salt content. The maximum at W_{Li} = 0.2 is typical of salt-in-polymer electrolytes, whereas the second maximum at W_{Li} = 0.8 arises from plasticization of the supercooled salt by the polyether matrix [4]. Strikingly, despite comparable conductivities at the two maxima, their transport and electrochemical characteristics differ fundamentally. The salt-in-polymer regime exhibits low Li^+ transference numbers (t_{Li^+} < 0.1) and sluggish interfacial kinetics at graphite and $LiFePO_4$ electrodes due to significant Li^+ desolvation barrier. In contrast, the polymer-in-salt regime shows increasing t_{Li^+} with W_{Li}, approaching unity in the supercooled salt (W_{Li} = 1), and enables reversible electrode reactions at both graphite and $LiFePO_4$ interfaces.

These findings highlight supercooled salts as a platform to decouple ionic conductivity from segmental dynamics and interfacial electrochemistry from Li^+ desolvation constraints in polymer electrolytes.

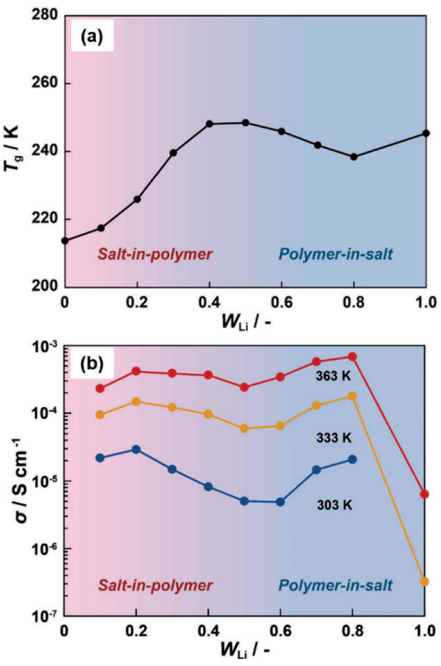

Figure 1 (a) T_g and (b) ionic conductivity (σ) as a function of W_{Li}.

References

[1] R. Masui, Y. Yokoyama, K. Matsumoto, S. Tsuzuki, M. Watanabe, S. Seki, *Chem. Mater.*, **37** (2025) 5636–5646.
[2] M. Kono, E. Hayashi, M. Watanabe, *J. Electrochem. Soc.*, **145** (1998) 1521–1527.
[3] C. A. Angell, C. Liu, E. Sanchez, *Nature*, **362** (1993) 137–139.
[4] T. Sudoh, Y. Ugata, K. Fujii, W. Shinoda, K. Ueno et al., *Adv. Energy Mater.*, **16** (2026) e05229.

Tailoring Anion Chemistry for Better Rechargeable Batteries

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Rechargeable batteries are increasingly important for integrating intermittent and unstable energy power (e.g., solar, wind, hydropower, etc.) into today's energy landscapes, thus gradually reducing the consumption of fossil fuels, coal, and other carbon-intensive fuels. To improve the cycling and storage performance of rechargeable batteries, the design of robust salt anions is of great importance, not only for the prevailing liquid electrolyte-based lithium-ion batteries, but also for next-generation solid-state batteries.[1]

During the past decades, the sulfonimide anions, with four oxygen atoms and one nitrogen in conjugation, stand out for both liquid and solid electrolytes developed for battery use. For example, the bis(trifluoromethanesulfonyl)imide ($[N(SO_2CF_3)_2]^-$, $TFSI^-$) shows relatively weak interactions with a variety of inorganic and organic cations, enabling facile dissociation and rapid migration of the cationic species in liquid and solid polymer electrolytes.[1] To cope the stringent demands in electrode-electrolyte interphases/interfaces, new sulfonimide anions beyond classic $TFSI^-$ anion and its derivatives are highly desired.[2]

In this talk, we will introduce a family of sulfonimide anions with certain functionalities optimized for improving the electrochemical compatibility between liquid/solid polymer electrolytes and high-energy electrodes.[3-5] Explicit correlations between the physical and electrochemical properties of liquid and polymer electrolytes with anionic structure will be discussed. Possible directions for chemically modifying sulfonimide anions for specific electrochemical scenarios will be elaborated.

References

[1]Z. Song, X. Wang, W. Feng, M. Armand, Z. Zhou and H. Zhang, *Advanced Materials* 2024, **36**, 2310245.

[2]H. Zhang, M. Armand. *Science China Chemistry* **2026**. 10.1007/s11426-026-3302-9.

[3]X. Wang, W. Feng, Z.-B. Zhou, H. Zhang, *Chemical Communications* **2024**, *60*, 11434.

[4]Z. Song, M. Tian, J. Zhu, J. Chen, W. Feng, L. Ben, H. Yu, X. Huang, M. Armand, Z. Zhou, H. Zhang, *Advanced Materials* **2024**, *36*, 2410954.

[5]J. Yang, X. Zhang, G. Cen, P. Li, Q. Wang, W. Feng, H. Yu, M. Armand, X. Huang, Z. Zhou, H. Zhang, *Journal of Power Sources* **2026**, *668*, 239339.

Molecular Design and Ion Transport in Lithium Ionic Liquid Electrolytes for Lithium Batteries

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Electrolytes with a high lithium-ion transference number ($t_{Li} \approx 1$) are essential for mitigating concentration polarization and improving rate capability in lithium secondary batteries. [1] While oxide- and sulfide-based solid electrolytes have been intensively studied to achieve such single-ion conduction, their practical implementation is often limited by interfacial instability, processing complexity, and scalability.[2] As an alternative approach, we explore lithium molten salts and lithium ionic liquid electrolytes, composed solely of Li salts without molecular solvents. In these systems, the absence of neutral solvent species suppresses the formation of salt concentration gradients, enabling intrinsically high lithium-ion transference under electrochemical conditions.[3] However, most Li salts exhibit high crystallinity and melting points, which limits their applicability as liquid electrolytes. In this study, we demonstrate two molecular design strategies to overcome these limitations. First, we show that lithium (fluorosulfonyl)(trifluoromethanesulfonyl)amide (Li[FTA]) can be stabilized in a deeply supercooled liquid state at room temperature by incorporating a small fraction of polymer additives.[4] Second, we develop ether-functionalized asymmetric Li imide salts with reduced melting points and glass transition temperatures, where conformational flexibility of the asymmetric imide anions contributes to suppressed crystallization and enhanced ion transport.[5] These lithium ionic liquid systems exhibit lithium-ion transference numbers approaching unity and support efficient lithium plating/stripping behavior. As a result, improved rate performance is achieved in Li/$LiCoO_2$ cells (Fig. 1). In addition, the intrinsic adhesive nature of the supercooled Li salt system with polymer additives enables the fabrication of composite electrodes without conventional binders, allowing for the construction of thick electrodes, and their electrochemical performance was evaluated in lithium cells. The present results highlight the potential of solvent-free lithium salt systems as a platform for developing safe and high-performance electrolytes for next-generation lithium batteries.

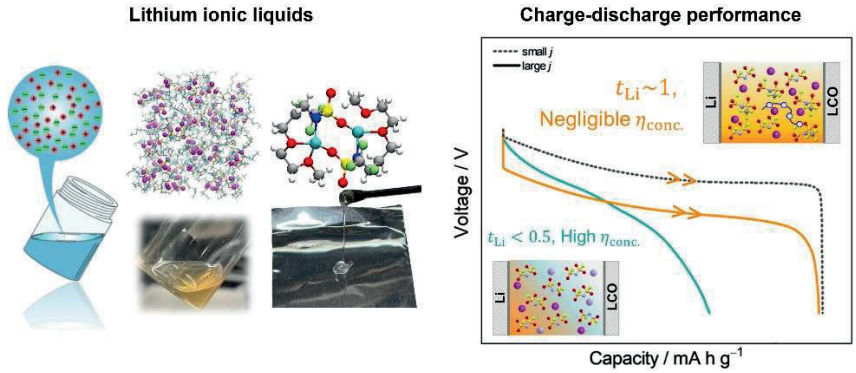

Fig. 1. Molecular design of lithium ionic liquids and discharge curves of lithium batteries using these electrolytes.

References

[1] K. M. Diederichsen *et al.*, *ACS Energy Lett.* **2** (2017) 2563–2575.
[2] J. Janek et al., *Nat. Energy* **1** (2016) 16141.
[3] K. Shigenobu *et al.*, *Phys. Chem. Chem. Phys.* **25** (2023), 6970–6978.
[4] T. Sudoh et al., *Adv. Energy Mater.* **16** (2026) e05229.
[5] Y. Matsuyama et al., *Energy Adv.* **4** (2025) 1438–1442.

Solid Bio Polymer Electrolytes for Advanced Electrochemical Applications

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Nowadays, it is the pursuit of the materials science community to implement sustainable materials in all fields of applications. It is expected that materials encompass properties like high abundance in nature; low cost; eco-friendliness; recyclability and suitable properties for the envisaged application.

The transition to renewable energy systems and high-energy-density applications requires safer and more efficient storage technologies [1–3]. In this context, solid polymer electrolytes (SPEs) have emerged as promising alternatives to conventional liquid electrolytes. Over the last decades, having cheap, efficient, and environmentally friendly energy storage supplies has become paramount. Therefore, SPEs have been studied since there is a need for effective, safer, and environmentally friendly devices. SPEs are promising materials for practical electrochemical applications, such as rechargeable batteries, supercapacitors, fuel cells, and electrochromic devices (ECDs).
Nevertheless, the need for the increasing use of biopolymers to make materials more environmentally friendly has become increasingly constant.

This work reports the development of novel electrolytes for electrochemical devices, based on a natural polymer, (for example the cellulose derivatives, doped with green ionic liquids (ILs) and/or salts). Membranes were characterized using techniques such as ultraviolet-visible spectroscopy, attenuated total reflectance Fourier transform infrared spectroscopy, electrochemical impedance spectroscopy, X-ray diffraction, scanning electron microscopy, thermogravimetric analysis, tensile testing, and contact angle measurements.

The results emphasize the huge potential of the developed green electrolytes in technological applications, as diverse as batteries and electrochromic windows.
These advances contribute to the development of functional materials based on natural polymers and highlight the role of solid polymer electrolytes in enabling safer, sustainable, and intelligent energy storage solutions.

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References

[1] J.M. Tarascon, M. Armand, Nature, 414 (2001) 359.
[2] X.B. Cheng, R. Zhang, C.Z. Zhao, Q. Zhang, Chemical Reviews, 117 (2017) 10403.
[3] D. Larcher, J.M. Tarascon, Nature Chemistry, 7 (2015) 19.

Zwitterionic additives for polymer electrolytes with high target ion transport.

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The advancement of polymer electrolytes is important to enable safer, more stable Li and Na batteries, and one novel approach is the use of zwitterionic additives to develop electrolytes with high target ion transport.[1] Here we discuss the use of novel zwitterions (ZIs) combined with single ion conducting polymers to make advanced solid-state electrolytes, and how the use of different zwitterionic and polymer structures impacts the important physical and electrochemical properties.

Three small zwitterionic structures were employed: two featuring pyrrolidinium-based cationic groups and one with an ammonium-based cation, tethered to either trifluoroborate or sulfonylimide-based anionic groups. This approach enabled us to investigate the impact of ionic moiety on the thermal and transport behavior and the dissociation of target ions. The electrolyte mixtures comprising the ZI and homopolymer (PMA(LiTFSI)) or copolymer PMA(LiTFSI-co-EG) were subjected to different physico-chemical and electrochemical characterization. For some of the electrolytes, the addition of Li TFSI or PC was also investigated to further improve the properties. The optimum system produced free-standing polymer gel electrolyte membranes, which were then subjected to linear sweep voltammetry and cycling studies to demonstrate their applicability in lithium battery applications. The membranes exhibited a better balance of ionic conductivity, cycling ability and transference number compared to the liquid 50 mol% LiTFSI-ZI mixture or the neat lithium homopolymer.

Finally, we will discuss the analogous approach used to develop gel polymer electrolytes for sodium batteries.

References

[1] F. Makhlooghiazad, L. A. O'Dell *et al*. *Nat. Mater.* **21**, (2022), 228–236.

Hybrid-Polymer Electrolytes for Lithium Metal Batteries

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Polymer–ceramic hybrid solid electrolytes are currently explored as promising materials class to leverage advantages of each constituent, that is, sufficiently high ionic conductivity of ceramics and superior mechanical flexibility of the polymer. Such systems may provide unexpectedly high ionic conductivities, though actual origins of this observation are still under discussion.[1] In practice, hybrid electrolytes may be produced invoking active or non-active ceramic fillers[2] or upon grafting of polymers to oxidic particles, often with up to 20 wt-% of ceramic components.[3]

Considering the increasing demand for affordable but high energy density batteries for various applications, significant efforts are devoted to enable thin lithium metal electrodes despite their challenges of inhomogeneous lithium deposition or loss of lithium inventory.[4,5] Grafted hybrid-polymer electrolytes demonstrated tolerance against Li dendrites while providing reasonable cell performance,[3] provided that the often porous cathodes may be polymer-infiltrated[6] or operated with catholytes.[7] The present work introduces aspects of extrusion processing of hybrid polymer electrolytes, highlighting not only viability of dry processing,[8] including electrolyte membranes and artificial CEIs for polyester based polymers, but also analyzing the fate of ceramic fillers and particle size distribution within the hybrid-polymer membranes upon cycling.[9]

In addition, to bestow sufficient compatibility of Li metal electrodes at demanding cell operation conditions, host structures from electro-spun scaffolds that accommodate lithium deposition are employed to boost the reversibility of lithium inventory,[10] whereas imaging data from 3D-SEM reconstructions reveal variations of particle size distributions and are evaluated as input structures for numerical simulation. The latter reflect localized ionic conductivities of grafted domains in the hybrid materials while providing insights that pave ways for better design strategies of hybrid polymer materials, thereby resolving governing mechanisms of enhanced ionic conductivities and hence fast charge capabilities of Li metal cells.

References:

[1] Sand, S. C.; Yildiz, B. et al, *Chemical Society Reviews* **2025**, *54*, 178
[2] Overhoff, G. M.; Brunklaus, G. et al., *ACS Applied Materials & Interfaces* **2022**, *14*, 53636
[3] Scharf, F.; Brunklaus, G. et al., *Small* **2024**, *20*, 2404537
[4] Brunklaus, G.; Winter, M. et al, *Nature Reviews Electrical Engineering* **2024**, *1*, 79
[5] Wichmann, L.; Brunklaus. G. et al., *Nature Communications* **2025**, *16*, 7216
[6] Buchheit, A.; Brunklaus, G. et al., *Energy Storage Materials* **2025**, 83, 104670
[7] Glomb, P. J.; Brunklaus, G. et al., *Batteries & Supercaps* **2026**, *9*, e202500679
[8] Scharf, F.; Brunklaus, G. et al., **2026**, *manuscript submitted*
[9] Tänzer, N., Brunklaus, G. et al., **2026**, *manuscript to be submitted*
[10] Wichmann, L., Brunklaus, G. et al., *Advanced Functional Materials* **2026**, *36*, e11672

Impact of processing parameters on the properties of thermoplastic vulcanizate (TPV) solid polymer electrolytes

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A new type of polymer blend electrolyte was developed based on the principle of thermoplastic vulcanizates (TPV). TPV materials have been extensively used in the automotive and manufacturing sectors since the 1970's[1]. These vulcanized polymer alloys have several desirable properties including strength, flexibility, amenability to reprocessing and customizability. However, these polymeric systems have yet to be introduced as electrolytes for use in battery systems. These electrolytes, which are obtained via melt-processing, combine the high ionic conductivity and processibility of a thermoplastic phase with the improved mechanical strength of a crosslinked elastomeric phase. They deliver promising results in terms of ionic conductivity, electrochemical stability and mechanical strength[2]. The presence of a cross-linked phase increases the elastic domain of the solid polymer electrolyte resulting in better creep resistance even when doped with electrolyte solvents.

This work presents the impact of the processing parameters on the end properties of these SPE. A TPV electrolyte composed of Ethylene-Propylene-Diene Monomer (EPDM) and poly(caprolactone) (PCL) was prepared using four different component addition orders. The effect of addition order and the resultant component interactions on the final properties of the SPE will be discussed (Fig1)[3]. A second TPV example composed of hydrogenated nitrile butadiene rubber (HNBR), PCL and poly(ethylene carbonate) (PEC) where the PEC is decomposed in-situ to yield EC (ethylene carbonate) will also be introduced. Electrolyte processing parameters and component addition order were shown to influence the structure of the resultant SPE as well as solvent and salt distribution in the final materials. These studies demonstrate that TPV electrolyte preparation methods strongly influence the end properties of these materials, providing a practical means to control electrolyte properties.

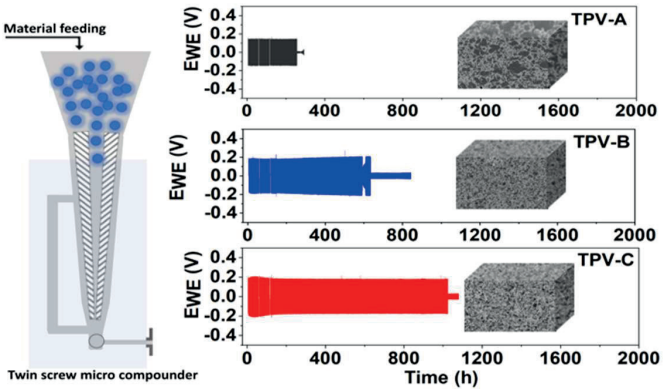

Fig1: a) schematic of extrusion processing, b) Li-Li cell cycling at 0.1mA/cm^2 of EPDM/PCL TPV electrolytes prepared using different processing sequences (same formulation).

References

[1] M.C. Boyce, K. Kear, S. Socrate, K. Shaw, *J Mech Phys Solids* **49** (2001) (5) 1073.
[2] L. Caradant, G. Foran, D. Lepage, P. Nicolle, A. Prébé, D. Aymé-Perrot, M. Dollé, *Journal of Power Sources Advances* **28** (2024).
[3] M. Azimi, G. Foran, C. Barcha, C. St-Antoine, A. Prébé, M. Dollé, *Materials Advances* **6** (2025) (12) 3945.

Initiator-driven in situ formation of gradient polymer electrolyte for high-performance lithium metal batteries

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Solid polymer electrolytes (SPEs) prepared by in situ polymerization approach within batteries enable not only substantially simplified manufacturing processes but also significantly improved contact between the SPEs and the active materials. The initiator is an essential component to trigger an in situ polymerization reaction. Benzoyl peroxide (BPO) and azobisisobutyronitrile (AIBN) are the most commonly used initiators for thermal-induced free radical polymerization. However, the utilization of these initiators is not consistent in the literature, and their impacts on the material properties and electrochemical performance of the resulting SPEs remain unclear. Here, a poly (vinylene carbonate) (PVC)-based SPE has been utilized to systematically investigate the impacts of the initiators in different cell formats. We observed that the type of initiator employed significantly influences not only the properties of the SPEs but also the electrochemical performance of the cells. Especially in full cells, employing BPO has been shown to result in a substantially lower monomer conversion within the cathode compared to cells employing AIBN, leading to a gradient SPE within the cell. This gradient SPE, synthesized via a single-step in situ polymerization, allows fast Li^+ transfer within the cathode and offers remarkable mechanical properties to suppress dendrite formation on the Li metal anode. Consequently, the gradient SPE enabled by BPO exhibits significantly improved cycle life and rate capability when compared to the SPE employing AIBN in full cells. This has led to a remarkable cycling performance of the full cells consisting of a NMC622 cathode, a Li metal anode, and an ultrathin SPE.

Effects of Cationic Polymer Coatings on Lithium Plating/Stripping Reactions

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For the development of next-generation rechargeable batteries with high energy density, lithium metal is an essential anode material due to its high theoretical capacity (3860 mAh g^{-1}) and low electrochemical potential (−3.04 V vs. standard hydrogen electrode). However, Li metal anodes suffer from low Coulombic efficiency and short cycle life due to the growth of whisker-like Li (so-called Li dendrites) and the reductive decomposition of electrolytes. To solve these issues, various interfacial engineering technologies, including electrolytes and polymer coatings, have been developed [1–3]. Cationic polymer coatings have been reported to control Li deposition morphology and improve the electrochemical performances of Li metal anodes [4,5]. However, the underlying mechanism remains controversial. In this study, we selected poly(diallyldimethylammonium) (PDADMA) as the cationic polymer backbone and investigated the effects of PDADMA-based polymer coatings on Li plating/stripping reactions. We found that sulfonylamide-type anions, such as bis(fluorosulfonyl)amide and bis(trifluoromethanesulfonyl)amide anions, are effective in improving the Coulombic efficiency of Li plating/stripping reaction. We studied the mechanism of Li plating/stripping behaviors via PDADMA-based polymer coatings by electrochemical measurements, microscopic observations, and spectroscopic characterization. The findings obtained are useful for the development of interfacial engineering technologies toward the practical application of lithium metal anodes.

Acknowledgements

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References

[1] J.-G. Zhang, W. Xu, J. Xiao, X. Cao, J. Liu, *Chem. Rev.*, **120** (2020) 13312−13348.
[2] Y. Maeyoshi, K. Yoshii, H. Sano, H. Sakaebe, R. Tamate, T. Kaneko, K. Sodeyama, *ACS Appl. Polym. Mater.*, **7** (2025) 1629–1638.
[3] Z. Huang, H. Lyu, L. C. Greenburg, Y. Cui, Z. Bao, *Nat. Energy*, **10** (2025) 811−823.
[4] S.-Y. Lee, J. Shangguan, J. Alvarado, Sophia Betzler, S. J. Harris, M. M. Doeff, H. Zheng, *Energy Environ. Sci.*, **13** (2020) 1832–1842.
[5] J. Wu, Z. Rao, X. Liu, Y. Shen, C. Fang, L. Yuan, Z. Li, W. Zhang, X. Xie, Y. Huang, *Adv. Mater.*, **33** (2021) 2007428.

Poster Session 2

(Tuesday, June 2nd)

Poster Session 2, Tuesday, June 2nd, 17:35-19:30

No	Presenter	Poster Title
P1-1	**Carlos Villacis**	PFAS-free Poly(ionic liquid) Reinforced with Cellulose Nanofibrils as Sustainable Electrolyte for High-Voltage Solid-State Lithium Batteries
P1-2	**Elene Sasieta Barrutia**	Lithium Fluoroborate Ester Salts for Enhanced Stability in PEO Electrolytes
P1-3	**David Fraile**	Towards Safer and Sustainable Lithium Metal Batteries: Fluorine-Free Solid Polymer Electrolyte
P1-4	**Daniel Weindl**	Polymer based hybrid solid electrolytes for all solid-state batteries
P1-5	**Nicole Llewellyn**	Structure-function relationships of mixed Li/K highly concentrated electrolytes
P1-6	**Lucas Marquart**	Gel polymer electrolyte implemented (Poly)phosphazene cathodes for possible IoT applications
P1-7	**Iliass Jalit**	Polysiloxane-based polymer electrolyte for lithium metal batteries
P1-8	**Rafael Del Olmo Martinez**	Enabling High-Voltage Solid-State Lithium Metal Batteries through Polymer Electrolyte Engineering
P1-9	**Walid Alkarmo**	Solid Composite Polymer Electrolyte as Enabler for High Energy Density Solid-State Lithium Batteries Operated at Ambient Conditions
P1-10	**Mattia Longo**	In-situ UV cured deep eutectic solvent-based gel polymer electrolyte for Li metal batteries
P1-11	**Leo Gräber**	Fluorine-Free Single-Ion Conducting Polymer Electrolytes for High-Energy Lithium-Metal Batteries
P1-12	**Soukaina Darmal**	Molecular Plasticization and Li3PO4 Reinforcement for Enhanced Ion Transport and Li+ Selectivity in PEO Solid Polymer Electrolytes
P1-13	**Théo Deschamps**	Single-Ion Electrolyte via DES Polymerization: Transport Mechanisms and Performance
P1-14	**Alberto Laggioni**	Comparative Study of Backbone Influence on the Internal Morphology of Radiation Grafted Anion-Exchange Membranes
P1-15	**Ilias Maniatis**	In-situ polymerization of comb-like PEO electrolytes: structure-property relationships
P1-16	**Milo Jouan**	Engineering Lithium-Ion Transport Through Multiscale Hybrid Polymer Electrolyte Architectures
P1-17	**Jichen Li**	Transport and mechanical properties of pseudo-zwitterionic polymer electrolytes from non-equilibrium molecular dynamics simulations
P1-18	**Yuliana Pairetti**	New Family of Protic Poly(diallyl ammonium)s and its Application as Fluorine-free Water-processable Binders for Batteries

P1-19	**Arunjunai Raja Shankar Santha Kumar**	Poly(Ether-Ester) based Solid Polymer Electrolytes derived from Cyclic Ketene Acetal and its Copolymers
P1-20	**Kevin Vattappara**	Single-ion polymer electrolytes enabling 5 V Li-LNMO solid-state batteries
P1-21	**Mónica Cobos**	Waterborne high-loading Ni-rich cathodes tailored for semi-solid-state lithium metal batteries
P1-22	**Lorena García**	Beyond LiTFSI: New Generation PFAS-free Salts for Enhanced Solid Polymer Electrolytes in Lithium Metal Batteries
P1-23	**Paul Imaz Elizalde**	Desing & optimization of dry processing for ex-situ gelled polymer electrolytes
P1-24	**Pierre Joncourt**	Ionic Transport properties of concentrated liquid electrolytes, beyond Debye Hückel Onsager (DHO) model
P1-25	**Elisabeth Springl**	Electrospun Sodium-Ion Conducting PAN/PEO-Based Electrolytes for ASSBs
P1-26	**Fy Maminoronirina**	Preparation of anion conducting plasticized polymer electrolytes by thiol-ene click chemistry for anionic organic batteries
P1-27	**Elvira Lind**	Single-step electrografting of copolymer electrolytes with enhanced mechanical integrity for separator-less structural sodium batteries
P1-28	**Laurence Beauregard**	Polymer Electrolytes Towards Structural Lithium-Sulfur Batteries
P1-29	**Kento Kimura**	Understanding the Effects of Structure and Composition on Mechanical Properties of Solid Polymer Electrolytes by Dynamic Viscoelasticity
P1-30	**Dunja Asceric**	Bio-Inspired Ion Conducting Membranes
P1-31	**Mohammad Baghban Shemirani**	Probing SEI Evolution via a Model System in Solid Polymer Electrolytes with Pseudo-Operando X-ray Photoelectron Spectroscopy
P1-32	**Deborah Feld**	Improved Determination of Li^+ Transference Numbers in PEO-based Electrolytes
P1-33	**Taku Sudoh**	Anomalous Plasticization in Polymer-assisted Deep Supercooling of Li-salt
P1-34	**Kewei Cai**	Design principles of polycations, polyanions and natural polymers in salt systems for next-generation solid polymer electrolytes

PFAS-free Poly(ionic liquid) Reinforced with Cellulose Nanofibrils as Sustainable Electrolyte for High-Voltage Solid-State Lithium Batteries

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Gel-like poly(ionic liquid) electrolytes are promising candidates to develop solid-state lithium metal batteries (SSLMBs). However, they hardly prevent lithium dendrite growth due to poor mechanical stability, which remains a critical challenge for the safety and long-term cycling in SSLMBs. Simultaneously, the increasing demand for environmentally friendly electrolytes has driven the search for sustainable, PFAS-free, and biodegradable alternatives. To address these challenges, in this work, PFAS-free composite polymer electrolytes are developed using poly(diallyldimethylammonium) bis(fluorosulfonyl)imide (PDADMA-FSI), N-methyl-N-propylpyrrolidinium bis(fluorosulfonyl)imide ($PYR_{13}FSI$), and lithium bis(fluorosulfonyl)imide (LiFSI). In addition, cellulose nanofibrils (CNFs) (from 0 to 8.5 vol.%, corresponding from 0 to 10 wt.%) are used to mechanically reinforce the electrolyte. The most promising properties are observed with a change in the volume ratio to 2.5% of CNFs, which led to enhanced lithium ionic conductivity, a higher storage modulus (G´), and reduced Li metal interface resistance. These improvements enable prolonged room temperature cycling of symmetric Li/Li and high-voltage Li/NMC622 coin cells compared to reference samples equipped with an electrolyte without CNFs. This study demonstrates that incorporating nanoscale cellulose harvested from renewable resources results in a promising strategy to enhance the electrochemical performance of gel poly(ionic liquid) electrolytes.

Lithium Fluoroborate Ester Salts for Enhanced Stability in PEO Electrolytes

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The development of solid polymer electrolytes (SPEs) with enhanced electrochemical stability is critical for next-generation high-voltage lithium metal batteries. Conventional lithium salts such as lithium bis(fluorosulfonyl)imide (LiFSI), while offering high ionic conductivity, often suffer from limited oxidative stability and interfacial degradation at elevated potentials, which ultimately restricts their applicability in high-energy systems. In this context, fluoroborate ester-based lithium salts have emerged as promising alternatives due to their weakly coordinating nature, high oxidative stability, and ability to delocalize negative charge, which can reduce ion pairing and improve interfacial compatibility.[1,2] In this work, a fluoroborate-based lithium salt, lithium tetrakis(hexafluoroisopropoxy)borate (LiBHFip),[3] is incorporated into poly(ethylene oxide) (PEO) matrices, and its electrochemical performance is systematically compared with the widely used PEO–LiFSI system. The incorporation of LiBHFip into PEO leads to a clear expansion of the electrochemical stability window toward higher potentials, with oxidative stability extending beyond 5.2 V vs Li^+/Li, demonstrating improved resistance against oxidative decomposition. This enhanced stability is particularly relevant for compatibility with high-voltage cathode materials. In addition, the PEO–LiBHFip electrolyte demonstrates superior stability during symmetric cell cycling and maintains improved performance at higher current densities compared to its LiFSI-based counterpart. This behavior indicates the formation of more robust and stable electrode–electrolyte interfaces, which are essential for long–term battery operation, positioning fluoroborate-based PEO electrolytes as promising candidates for solid-state LMBs.

References

[1] Z. Liu, J. Chai, G. Xu, Q. Wang, G. Cui, Functional lithium borate salts and their potential application in high performance lithium batteries, *Coord. Chem. Rev*. 292 (2015) 56–73.
[2] C. Han, L. Qiao, G. Xu, G. Chen, K. Chen, S. Zhang, J. Ma, S. Dong, X. Zhou, Y. Han, Z. Cui, G. Cui, Self-Standing Single-Ion Borate Salt-Based Polymer Electrolyte for Lithium Metal Batteries, ACS *Appl. Mater. Interfaces* 16 (2024) 48792–48802.
[3] B. Roy, P. Cherepanov, C. Nguyen, C. Forsyth, U. Pal, T.C. Mendes, P. Howlett, M. Forsyth, D. MacFarlane, M. Kar, Lithium Borate Ester Salts for Electrolyte Application in Next-Generation High Voltage Lithium Batteries, *Adv. Energy Mater.* 11 (2021).

Towards Safer and Sustainable Lithium Metal Batteries: Fluorine-Free Solid Polymer Electrolyte

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Lithium metal is widely regarded as the most promising anode material in the field of energy chemistry, owing to its notably high specific capacity (3860 $mAh \cdot g^{-1}$). Nevertheless, it is important to emphasize that the use of lithium metal as an anode material is conditional upon the transition to solid state electrolytes, such as polymer electrolytes, due to their incompatibility with highly flammable commercial liquid electrolytes.

Moreover, commonly used electrolytes are dual lithium-ion conductors, both cations and anions are mobile, which leads to concentration polarization and consequently, to formation of lithium dendrites that could cause cyclability and safety issues. Single lithium-ion conducting solid polymer electrolytes (SLIC-SPE), which have the anions covalently bound to the polymer backbone, have only mobile cations, thus limiting electrolyte polarization and the formation of lithium dendrites.

Besides that, the use of fluorine is a subject that is currently under debate. The primary reasons for this are its potentially harmful characteristics, including its cost, the safety of its electrolytes and its environmental impact.

This work presents that the blend of fluorine free salts with polyethylene oxide (PEO) for lithium metal batteries can be a viable alternative to fluorine-based electrolytes. Three salts (Figure 1.) were characterized for this purpose: lithium 4,5-dicyano-2-(pentafluoroethyl)imidazolate (LiPDI), lithium 4,5-dicyano-2-(trifluoromethyl)imidazolate (LiTDI) and lithium 2,4,5-tricyanoimidazol-1-ide (LiTIM). The first two, fluorinated, were analyzed in order to compare them with the LiTIM salt, fluorine-free, to see the impact of fluorine on the prepared electrolytes. In addition, a new fluorine free SLIC-SPE will be presented based on a cyano-substituted imidazole.

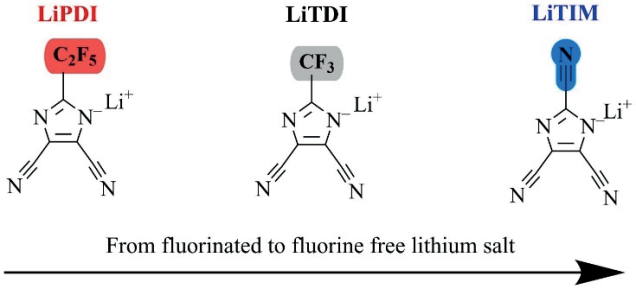

Figure 1. Chemical structures of the employed lithium salts from fluorinated to fluorine free anions: LiPDI, LiTDI and LiTIM.

References

[1] Martinez-Ibañez, M. et al., Adv. Funct. Mater. 2020, 30, 2000455.

[2] Doyle, M. et al., Electrochim. Acta, 1994, 39, 2073–2081.

Polymer based hybrid solid electrolytes for all solid state batteries

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Solid polymer electrolytes (SPE) like Polyethylene oxide (PEO): Lithium bis-triflylsulfonylimide (LiTFSI) have been widely researched for their use in all-solid-state batteries in pursuit of greater operational safety. However, several shortfalls have hindered their wider application, including limited thermal stability, low conductivity and poor mechanical stability.[1] To enhance polymer electrolytes towards general usability the field of hybrid polymer solid-state electrolytes has emerged. They consist of an active filler of a solid-state electrolyte (SSE) class like oxides or sulfides blended into a polymer electrolyte matrix, synergistically combining benefits of both material classes.[2] Hybrids with this design show high ionic conductivity and mechanical stability, low interfacial resistance and upscaling potential towards an industrial level.[3]

In this work we examine the interactions of sample polymer:salt electrolyte systems like PEO:LiTFSI with different solid electrolytes based on ceramics or sulfides like Li_6PS_5Cl. Using different preparation methods like hot pressing, solution casting and other slurry-based methods samples with varying SSE to SPE ratios are prepared. Furthermore, different potential solvents are tested for application in a slurry process with more sensitive hybrid sulfide electrolytes and their impact on ionic conductivity, polymer solubility and phase changes associated with degradation of solid electrolytes are screened. For all samples, morphology and crystallinity of hybrid electrolytes are characterized using scanning electron microscopy and X-ray diffraction. Potentiostatic electrochemical impedance spectroscopy and cyclic voltammetry is employed to analyze the compatibility of combinations of different electrolytes like PEO and Li_6PS_5Cl and potential degradation over time. Determining the time-resolved contact resistance of a solid-solid electrolyte interface between SPE and SSE is also used to observe structural changes between the different electrolytes. Shuttling experiments in symmetric cells evaluate the stability against alkaline metal like Lithium over time.

References

[1] Hwang, J.-Y., *et. al*., *Chem. Soc. Rev.*, **46**, (2017) 3529–3614.
[2] Chauhan, A. K., *et. al., J. Electron. Mater*., **50**, (2021) 5122–5133.
[3] Zhang, X., *et. al*., *Energy Storage Mater*., **79**, (2025) 104339–104406.

Structure-function relationships of mixed Li/K highly concentrated electrolytes

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Lithium-ion batteries (LIBs) have high energy density, long cycle life, and fast charging capability. Hybrid batteries consisting of mixed cation electrolytes incorporating multiple charge carriers such as Li^+ and K^+ have recently emerged as an alternative to single cation systems to improve ion transport, interfacial stability, and overall electrochemical performance.

Highly concentrated electrolytes (HCEs) have been shown to enhance ionic conductivity and thermal stability [1] by correlated ion transport through contact ion pairs and larger ionic aggregates that also render less or no "free" solvent. Moreover, the solid electrolyte interphase (SEI) is mainly formed by anionic species, which promote long-term cycling stability and reduces electrolyte degradation [2].

Combining the two concepts of mixed cation electrolytes and HCEs has some promises or at least prospects of synergies. The presence of both Li^+ and K^+ can promote an altered, yet anion-based, dense and inorganic SEI, and within graphite as anode active material the storage of Li^+ offers high capacity, while K^+ should offer faster dynamics to deliver improved rate capabilities [4].

As proof-of-concept, we have studied a HCE of 2.5 M KFSI + 2.5 M LiFSI in PC in Li || graphite half-cells. Through physico-chemical and electrochemical characterization, we correlate HCE composition vs. battery performance. Initial results show improved performance both in terms rate capability and cycling stability vs. conventional single cation HCEs. By using electrochemical impedance spectroscopy (EIS) we can reveal a lower charge transfer resistance, and by Raman spectroscopy a higher degree of ion pairing and aggregation. In addition, both increased glass transition temperature (T_g) and higher onset decomposition temperature were demonstrated, consistent with strong ion–solvent coordination and aggregate formation. Finally, XPS analysis of cycled electrodes shows an SEI enriched in both LiF and KF.

In future work, we will optimize the Li/K HCE formulations, salt, solvent, and concentrations to advance the design of hybrid batteries.

References

[1] Y. Yamada and A. Yamada, *J. Electrochem. Soc.*, **162** (2015) A2406-A2423
[2] S.-K. Jeong et al., *Electrochem. Commun.*, **10** (2008) 635–638
[3] M.R. Lukatskaya et al., *Energy Environ. Sci.*, **11** (2018) 2876–2883
[4] W. Deng et al., *Energy Storage Mater.*, **20** (2019) 373–379

Gel polymer electrolyte implemented (Poly)phosphazene cathodes for possible IoT applications

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The demand for efficient and reliable batteries is rapidly increasing, driven by various factors such as the growing market of health monitoring, wearable technology and other applications of the internet-of-things.[1,2] Raw material security concerns, trade conflicts, and supply chain issues contribute to market volatility and uncertainty and highlight the necessity for alternative solutions.[3] In response to these challenges, organic batteries currently emerge as promising and sustainable alternative to conventional batteries.[4] Organic batteries enable fast charging due to the underlying electron transfer mechanism, allow for high operational safety and are mechanically rather flexible, rendering them ideally suited for wearable applications.[4,5] However, several challenges remain to be solved for organic batteries to be realistic alternatives to other commercial batteries, particularly overcoming the comparably low mass loading and lower specific capacities of organic materials than inorganic materials. Also, organic materials tend to be electrical insulator, which makes excessive use of conductive additive necessary.[6]

In this work, TEMPO grafted (poly)-phosphazenes(TG-PPZ) are presented as promising organic active material that may counter challenges of resource scarcity and flammability due to their flexible backbone made from abundant materials and their inherently flame extinguishing properties.[7,8] TEMPO grafted phosphazenes represent a derivative to poly(2,2,6,6-tetramethyl-1-piperidinyloxy-4-yl methacrylate) (PTMA) which is considered the gold standard in organic electrode research due to its high chemical and mechanical stability, straightforward synthesis and comparably high theoretical specific capacity (C_{theo} = 111 mAh g^{-1}).[4] TG-PPZ should surpass the performance of PTMA because of a higher theoretical specific capacity and more flexible polymer backbone (C_{theo} = 132 mAh g^{-1}).[8] The material demonstrates initial specific discharge capacities of 62.9 mAh g^{-1} at rates of up to 1C (0.1 mAh), though a key focus remains to limit the active material dissolution, a common challenge in organic batteries. This is addressed through the implementation of gel polymer electrolytes designed to inhibit detachment of the active material from the carbon black, thereby enhancing battery cycle life and overall performance. Notably, preliminary data indicate that TG-PPZ cells utilizing a gel polymer electrolyte achieve initial discharge capacities comparable to those observed with liquid electrolytes (61.2 mAh g^{-1}). Future research will focus on optimizing the performance of acrylate-based gel polymer electrolytes reinforced with PVDF-HFP separators through systematic variation of acrylate content, membrane thickness, and lithium salt composition.

References

[1] A. K. M, Ramani. R, R. Krishnamoorthy, S. Gogula, Baskar. S, S. Muthu, G. Chellamuthu, K. Subramaniam, *Sci. Rep.* **2024**, *14*, 6151.
[2] J. Panidi, D. G. Georgiadou, T. Schoetz, T. Prodromakis, *Adv. Funct. Mater.* **2022**, *32*, 2200694.
[3] M. A. Rajaeifar, P. Ghadimi, M. Raugei, Y. Wu, O. Heidrich, *Resour. Conserv. Recycl.* **2022**, *180*, 106144.
[4] D. T. Daniel, S. Oevermann, S. Mitra, K. Rudolf, A. Heuer, R.-A. Eichel, M. Winter, D. Diddens, G. Brunklaus, J. Granwehr, *Sci. Rep.* **2023**, *13*, 10934.
[5] L. Zuo, D. Lu, T. Yang, D. Yue, W. Li, Q. Ma, Y. Chen, C. Zheng, X. Wu, *Carbon Neutralization* **2022**, *1*, 316–345.
[6] H. Guo, C. Wang, *ChemSusChem* **2024**, *17*, e202301586.
[7] A. W. Knights, M. A. Nascimento, I. Manners, *Polymer* **2022**, *247*, 124795.
[8] H. R. Allcock, C. Chen, *ACS Polym. Au* **2025**, *5*, 811–826.

Polysiloxane-based polymer electrolyte for lithium metal batteries

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The development of high-energy lithium metal batteries requires advanced electrolytes that not only serve as media for lithium-ion transport but also largely determine the stability of electrode/electrolyte interfaces. Solid electrolytes are a promising alternative, since they could offer improved thermal stability and potentially suppress dendritic lithium deposition [1]. Among them, due to their flexibility and modularity, solid polymer electrolytes (SPEs) are excellent candidates. For their commercial implementation, however, one must simultaneously ensure acceptable ionic conductivity, high mechanical strength, and an electrochemical stability window. Conventional polymer electrolytes based on poly(ethylene oxide) (PEO) consist of a lithium salt dissolved in the polymer. The need for a conducting salt result in transference numbers far below 0.5. This leads to concentration gradients and cell polarization reversal issues, which worsen the long-term cycling stability [2]. SPEs with transference number close to 1 known as "single-ion polymer electrolytes" (SIPEs), might overcome these issues. However, in the polymeric materials, high mechanical properties go often hand-in-hand with low mobility, which is antagonistic to high ionic conductivity (i.e. high chain mobility). Among the polymers, the polydimethylsiloxane (PDMS)-based materials are attractive due to their low transition temperature [3], flexible backbone, and favorable interfacial compatibility with lithium metal [4,5]. In this work, we report on the synthesis and characterization of SIPE based on PDMS functionalized with prepared trifluoromethylsulfonyl amide-based lithium salt. The SIPE were incorporated poly(butyl malonate) (PBM) that was then crosslinked by using diacrylate with different length chains to form a self-supporting membrane. The relationship between polymer structure and transport properties is examined through physicochemical and electrochemical characterization (^{1}H ^{19}F ^{7}Li NMR, chronoamperometry, DSC, EIS). The developed blend highlighted an ionic conductivity of 1.52 x 10^{-5} $S.cm^{-1}$ at 80°C at O/Li = 20, with a transference number of 0.9 estimated using Watanabe equation, indicating strongly restricted anion mobility. Furthermore, anodic stability evaluation shows value around 4.3 V *vs*. Li^{+}/Li at 25°C. Moreover, PBM-PSX membrane blend provided low glass temperature at around -30°C and good thermal stability.

References

[1] Arico, A et al. *Nature materials*, (2005) *4*(5), 366-377.
[2] Tikekar, M. D., Choudhury, S., Tu, Z., & Archer, L. A. *Nature Energy*, (2016) *1*(9), 1-7.
[3] Ovornlc, P. R. Chapter 7: thermal properties of polysiloxanes. (2000).
[4] Yue, L et al. *Energy Storage Materials*, (2016) *5*, 139-164.
[5] Liang, H et al. *Advanced energy materials*, (2022) *12*(16), 2200013.

Enabling High-Voltage Solid-State Lithium Metal Batteries through Polymer Electrolyte Engineering

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Achieving safe and high-energy lithium metal batteries requires rethinking cathode design beyond conventional liquid-based architecture. In solid-state systems, the cathode evolves into a multifunctional domain where ionic transport, electrochemical stability, and mechanical integrity must be simultaneously ensured, particularly under high-voltage operation.

Within the scope of the HyLiST project,[1] this work explores the development of polymer-based electrolytes derived from poly(methacrylate) bis(trifluoromethanesulfonyl)imide (PMTFSI) as enabling materials for next-generation high-voltage batteries. Emphasis is placed on single-ion conducting polymer systems and their compositional tuning to enhance ionic transport while preserving structural robustness and oxidative stability up to 5 V. The materials are processed using dry-processing techniques, combining performance with compatibility for scalable and sustainable manufacturing.

A progressive evaluation methodology is adopted to assess electrolyte performance across different levels of complexity, enabling the decoupling of key phenomena related to ion transport and electrochemical stability. This approach supports the identification of critical parameters governing stability and efficiency, while guiding the transition toward fully solid-state configurations.

Overall, this work contributes to the establishment of design guidelines for polymer catholytes capable of supporting high-voltage operation, paving the way for the development of safe and high-performance solid-state lithium metal batteries.

[1] https://hylist.eu/

Solid Composite Polymer Electrolyte as Enabler for High Energy Density Solid-State Lithium Batteries Operated at Ambient Conditions

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The growing demand for safe and sustainable energy storage solutions in electromobility has driven advancements in solid-state battery technology. We present a novel solid-state battery featuring a functional three-dimensional crosslinked polymer network with a lithium-selective, transport-regulated composite polymer electrolyte architecture.

This solid electrolyte architecture provides mechanical stability, reduced anion mobility, and suppressed concentration polarization. The electrolyte enables stable room-temperature operation in large cell scale of 0.86 Ah using thin lithium metal and NMC811 pouch cells at high C-rate, demonstrating sustained cycling performance, with low interfacial resistance and improved rate capability compared to conventional solid polymer electrolytes.

In-situ UV cured deep eutectic solvent-based gel polymer electrolyte for Li metal batteries

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The global rise in energy consumption has driven research towards safer, more efficient energy storage systems. The commonly employed Li-ion batteries are prone to safety risks due to flammable liquid organic solvents, which can evaporate or leak and pose hazards in case of a short circuit. Deep eutectic solvents (DES) are a relatively new family of solvents with room temperature molten salt behaviour and several advantageous characteristics in the context of Li-ion batteries electrolytes. Their low vapour pressure and non-flammability, as well as good electrochemical performances, render them interesting candidates for application in contact with Li metal [1]. In this work, a DES composed of trifluoroacetamide (TFA) and lithium bis(trifluoromethanesulfonyl) imide (LiTFSI) is prepared and thoroughly characterized. A gel polymer electrolyte (GPE) is subsequently developed with the focus on the up-scalability of the GPE production. The polymeric matrix, composed of polyethylene glycol diacrylate (PEGDA) and pentaerythritol tetrakis(3-mercaptopropionate) (T4), is crosslinked via thiol-ene polymerization directly in the DES solution, demonstrating a fast, cheap and easy reaction. The thiol ene mechanism, moreover, avoids the sensitivity towards O2 of the otherwise commonly employed UV-mediated radical polymerization and enables the crosslinking reaction to be carried out in a dryroom [2]. The precursor solution is deposited directly on the surface of the investigated cathodes and irradiated with 10 W UV light to obtain a GPE with excellent interfacial contact. Moreover, a 10 %*wt* addition of ethylene carbonate and diethyl carbonate (1:1 *wt*, namely EDL) in the DES is thoroughly investigated via NMR, FTIR and Raman spectroscopy to elucidate the effect of the carbonate addition on the Li solvation shell. The resulting DES10 solution displays superior electrochemical performances of the GPEs, increasing the ionic conductivity of the GPEs from $6.36 \cdot 10^{-4}$ S cm^{-1} to $1.25 \cdot 10^{-3}$ S cm^{-1} at room temperature and enabling a long lifetime of 220 cycles with a high specific capacity of 164 mAh g^{-1} when cycling in a Li||LFP cell at 1C and at room temperature.

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References

[1] K. Zhou, X. Dai, P. Li, L. Zhang, X. Zhang, C. Wang, J. Wen, G. Huang, S. Xu, Prog. Mater. Sci. 146 (2024) 101338.
[2] C.E. Hoyle, C.N. Bowman, Angew. Chem. Int. Ed. 49 (2010) 1540 – 1573.

Fluorine-Free Single-Ion Conducting Polymer Electrolytes for High-Energy Lithium-Metal Batteries

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Fluorine-based lithium salts such as lithium hexafluorophosphate ($LiPF_6$) are currently exploited in presumably all conventional battery electrolytes [1]. However, these materials are facing increasing criticism regarding safety considerations, including the related corrosive and hazardous decomposition products, and owing to the environmental persistence of fluorinated compounds [2]. Meanwhile, the growing global demand for energy storage systems urges the development of chemically stable electrolytes to enable the use of high-voltage electrode materials and boost the energy density of lithium batteries, ensuring at the same time safe long-term cycling [3]. In this regard, single-ion conducting polymer electrolytes (SIPEs) are considered a promising approach due to their intrinsically high lithium transference number of essentially unity, which suppresses concentration polarization and, thus, lithium dendrite formation [4]. Over the past decades, SIPEs have attracted significant interest, yet there have been only a few reports on fluorine-free alternatives due to the undeniable contribute of fluorinated compounds in stabilizing the electrode|electrolyte interface [4,5].

Following this thread, we have recently reported a completely fluorine-free lithium-metal battery relying on a novel SIPE and a $LiFePO_4$ cathode, benefiting of a water-based binder [5]. The SIPE consisted of lithium(4-styrenesulfonyl)(dicyanomethide) grafted onto a poly(methylmercaptopropyl)siloxane-backbone via thiol-ene click-reaction. Despite showing remarkable electrochemical properties comparable to fluorinated analogues, the relatively low electrochemical stability towards oxidation of 4.2 V vs. Li^+/Li limited its application with high-voltage cathodes, such as $LiNi_xMn_yCo_{1-x-y}O_2$ (NMC).

Herein, we report the incorporation of a fluorine-free functional additive into our fluorine-free SIPE, which leads to a significantly enhanced stability towards oxidation, finally allowing for the very stable cycling of Li||NMC_{622} cells. As a result, this new polymer electrolyte system marks an important step forward towards the realization of safer, more sustainable and easy-to-recycle high-energy lithium batteries.

References

[1] G. Hernández, R. Mogensen, R. Younesi, J. Mindemark, Fluorine-Free Electrolytes for Lithium and Sodium Batteries, Batter. Supercaps 5 (2022) e202100373. https://doi.org/10.1002/batt.202100373.
[2] J.-P. Hoffknecht, J. Atik, C. Krause, J. Thienenkamp, G. Brunklaus, M. Winter, E. Paillard, Beyond fluorine: sustainable ternary polymer electrolytes for lithium batteries, Green Chem. 23 (2021) 9935–9944. https://doi.org/10.1039/D1GC02451E.
[3] X. Lu, Y. Wang, X. Xu, B. Yan, T. Wu, L. Lu, Polymer-Based Solid-State Electrolytes for High-Energy-Density Lithium-Ion Batteries – Review, Adv. Energy Mater. 13 (2023) 2301746. https://doi.org/10.1002/aenm.202301746.
[4] J. Zhu, Z. Zhang, S. Zhao, A.S. Westover, I. Belharouak, P. Cao, Single-Ion Conducting Polymer Electrolytes for Solid-State Lithium–Metal Batteries: Design, Performance, and Challenges, Adv. Energy Mater. 11 (2021) 2003836. https://doi.org/10.1002/aenm.202003836.
[5] L. Gräber, V. Marangon, M. Kumar, M. Weil, D. Bresser, Fluorine-free polysiloxane-based single-ion-conducting polymer electrolyte for lithium–metal batteries, Green Chem. 27 (2025) 8226–8236. https://doi.org/10.1039/D5GC00252D.

Molecular Plasticization and Li_3PO_4 Reinforcement for Enhanced Ion Transport and Li^+ Selectivity in PEO Solid Polymer Electrolytes

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Solid polymer electrolytes (SPEs) based on poly(ethylene oxide) (PEO) are widely investigated for solid-state lithium batteries due to their safety and processability; however, their practical implementation remains limited by high crystallinity, moderate ionic conductivity, and insufficient Li^+ selectivity [1]. In this work, we explore a combined strategy of inorganic filler reinforcement and molecular plasticization to engineer ion transport and electrochemical stability in composite PEO electrolytes. Building on a Li_3PO_4-reinforced PEO–LiTFSI framework [2], succinonitrile (SN) was introduced as a molecular plasticizer to tune the structural and transport properties of the polymer matrix [3].

Structural analysis indicates that SN primarily plasticizes the polymer matrix, reducing PEO crystallinity and increasing chain mobility while indirectly promoting LiTFSI dissociation. Consequently, the plasticized composites exhibit a significant increase in ionic conductivity, reaching 2.8×10^{-4} S cm^{-1} at 60 °C, compared with 6.59×10^{-5} S cm^{-1} at 60 °C for the non-plasticized composite electrolyte. Electrochemical measurements also reveal enhanced oxidative stability. While the filler-free PEO electrolyte decomposes at approximately 4.4 V vs. Li/Li^+, Li_3PO_4-reinforced systems extend the stability window to about 5.2 V, and the introduction of SN further increases decomposition voltages up to 5.5 V.

Lithium-ion transference measurements indicate that SN modifies ion transport depending on the host matrix structure, maintaining relatively stable Li^+ selectivity in the crystalline 20 wt% Li_3PO_4 system while promoting greater anion mobility in highly filled formulations. Interfacial stability studies confirm stable polarization behavior and reversible lithium plating/stripping. Galvanostatic cycling with high-voltage cathodes validates the electrochemical compatibility of the series under practical operating conditions.

Overall, these results demonstrate that SN plasticization fundamentally reshapes ion transport in Li_3PO_4-reinforced PEO electrolytes, introducing a conductivity–selectivity interplay governed by polymer crystallinity and filler content. This study highlights the importance of balancing polymer mobility with controlled ion transport and provides design guidelines for high-conductivity SPEs compatible with high-voltage solid-state lithium batteries.

References

[1] M.R. Bonilla, M. Armand, et al., PEO(LiTFSI) polymer–ceramic composites for solid electrolyte applications, J. Colloid Interface Sci. 623 (2022) 594–605.

[2] S. Darmal, S. Abouricha, K. Elbouazzaoui, D. Brandell, R. Hakkou, H. Ben Youcef, I. Saadoune, Lithium phosphate as a novel filler in PEO-based solid polymer electrolytes: New pathways to improved high-voltage cathode performance, J. Power Sources 662 (2026) 238684.

[3] L.-Z. Fan, Y.-S. Hu, A.J. Bhattacharyya, J. Maier, Succinonitrile as a versatile additive for polymer electrolytes, Adv. Funct. Mater. 17 (2007) 2800–2807.

Single-Ion Electrolyte via DES Polymerization: Transport Mechanisms and Performance

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Solid polymer electrolytes (SPEs) represent a promising route toward safer lithium-metal batteries, owing to their processability, the absence of volatile solvents, and their potential mechanical barrier effect. However, two major bottlenecks still hinder their development: (i) ion transport coupled to segmental mobility, resulting in limited ionic conductivity at room temperature, and (ii) the co-migration of cations and anions in SPEs, which leads to concentration polarization and detrimental interfacial gradients.

To address these limitations, two complementary strategies have emerged. Gel polymer electrolytes (GPEs) incorporate a liquid ion-conducting phase within a polymer matrix, thereby improving ionic conductivity, but potentially compromising interfacial stability and thermal robustness. Among such conductive liquid phases, deep eutectic solvents (DESs)—liquids formed through association of a hydrogen-bond donor and acceptor (HBD/HBA), or, in the case of Li-DESs, of a Li^+ salt and an HBD—strongly depress the melting point and combine low volatility with high solvation ability. For example, a lithium bis(trifluoromethanesulfonyl)imide (LiTFSI)/N-methylacetamide (NMAc) mixture at a 1:5 molar ratio reaches $\sigma(25\ °C) = 1.75 \times 10^{-3}\ S \cdot cm^{-1}$ [1]. In parallel, single-ion conducting polymer electrolytes mitigate cell polarization by enforcing a Li^+-dominated flux through tethering the anionic moieties to the polymer backbone [2].

In this work, we combine these approaches by developing new single-ion polymer electrolytes through the polymerization of a DES formed from the single-ion monomer STFSILi, which bears a polymerizable styrenic moiety and a covalently tethered TFSI-type anion, and the hydrogen-bond donor NMAc. By varying the molar ratio between the single-ion monomer and the HBD, we identify three regimes of structure and ion transport, with the electrolyte transitioning from a "dry" single-ion regime to a "diluted" single-ion regime, and exhibiting an optimal intermediate regime. In this regime, the electrolyte reaches $\sigma(30\ °C) \approx 5 \times 10^{-4}\ S \cdot cm^{-1}$, shows a near-unity lithium transference number ($tLi^+ \approx 1$), and features an electrochemical stability window compatible with NMC- and LFP-type positive electrodes.

Beyond this specific system, polymerization of DESs provides a versatile route to electrolytes in which the eutectic fraction promotes ion transport while the polymer network ensures mechanical cohesion and predominantly Li^+ conduction. This strategy offers a promising pathway toward solid-state electrolytes that better balance transport, stability, and processability.

References

[1] An-Sifue Kelchtermans, Bkorn Joos, Dries de Sloovere, Jonas Mercken, Elien Derveaux, Peter Adriaensens, Alexander Tesfaye, Marlies K. Van Bael, and An Hardy, « How Polymer Influences the Electrochemical Behavior of Eutectogels for Lithium Batteries », ACS Appl. Polym. Mater. 2024, 6, 90-101

[2] Luca Porcarelli, Alexander S. Shaplov, Federico Bella, Jijeesh R. Nair, David Mecerreyes et Claudio Gerbaldi, "Single-Ion Conducting Polymer Electrolytes for Lithium Metal Polymer Batteries that Operate at Ambient Temperature", ACS Energy Lett. 2016, 1, 678-682

Comparative Study of Backbone Influence on the Internal Morphology of Radiation Grafted Anion-Exchange Membranes

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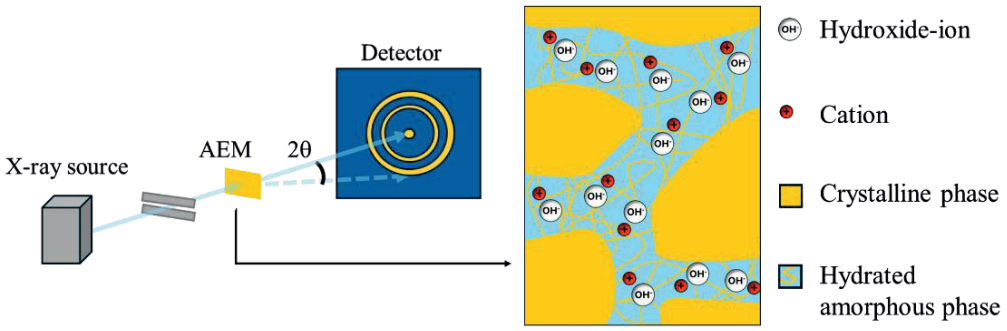
Ion-exchange membrane (IEM)-based devices are central to the green energy transition, enabling electrochemical energy conversion and storage across a wide range of applications. In these devices, membrane nanoscale morphology governs ion and water transport, directly determining overall performance.[1] Among IEMs, radiation-grafted anion exchange membranes (AEMs) are gaining traction, yet the influence of polymer backbone chemistry on their properties remains insufficiently explored.

Here, we investigate how backbone chemistry differences between ethylene tetrafluoroethylene (ETFE) and low-density polyethylene (LDPE) affect AEMs bearing identical functional groups, combining bulk hydration characterization with small-angle X-ray scattering (SAXS) to capture how each backbone drives distinct nanoscale reorganization upon hydration. LDPE-based membranes show ~30% higher water uptake and swelling compared to ETFE films, attributed to the greater chain mobility of the more amorphous polyethylene matrix, which facilitates structural reorganization to accommodate water molecules. In contrast, ETFE-based membranes exhibit better dimensional stability, owing to the high hydrophobicity of the fluorinated backbone. The d-spacing values extracted from SAXS profiles range between 22 and 35 nm and do not always mirror bulk hydration behavior: higher degrees of functionalization of LDPE membranes show greater swelling yet reduced spacing, demonstrating that bulk and nanoscale structural responses are decoupled and that swelling measurements alone may not fully capture structural reorganization. Additionally, to lower the barrier for SAXS adoption in the membrane community, we developed TRAMA, a Jupyter-notebook-based platform that guides researchers through the data analysis workflow without prior expertise.[2] This work highlights the critical role of backbone chemistry in determining AEM morphology and establishes a structural benchmark for future radiation-grafted membrane design. These findings lay the groundwork for correlating nanoscale organization with ionic transport and device performance.



SAXS probing of AEM nanoscale morphology: incident X-rays produce a scattering pattern encoding information on the membrane inner structure.

References

[1] G. Shu, Z. Song, W. Wang, H. Tian, Review of emerging multiple ion-exchange membrane electrochemical systems for effective energy conversion and storage, Sustainable Energy Technologies and Assessments 70 (2024) 103926. https://doi.org/10.1016/j.seta.2024.103926.

[2] A. Laggioni, L. Besley, T. Saatkamp, J. Fonsaca, E. Santiago A. Konovalova, Analysis Tool for Rapid SAXS Data Set Interpretations: Comparative Study of Backbone Influence on Internal Morphology of Anion-Exchange Membranes, *submitted*.

In-situ polymerization of comb-like PEO electrolytes: structure-property relationships

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The growing demand for high-energy-density and safer lithium-ion batteries (LIBs) necessitates the replacement of traditional liquid electrolytes with inorganic or polymer alternatives. In this context, gel polymer electrolytes (GPEs) have emerged as promising candidates due to their high ionic conductivity (~10^{-3} S cm^{-1} at RT), enhanced interfacial contact, improved safety, and processability [1]. Among various polymer matrices, polyethylene oxide (PEO) stands out due to its structural flexibility, strong solvating power toward Li^+, and sufficient electrochemical stability. However, its semicrystalline nature, which leads to a drastic decrease in ionic conductivity, limits its use to high-temperature systems (above its melting point, 65 °C). To circumvent this obstacle, comb-like polymers featuring oligo(ethylene oxide) side chains have attracted significant attention. Their highly amorphous nature and tunable molecular architecture provide enhanced ionic conductivity, while they can be readily synthesized in a single step via free-radical polymerization of (meth)acrylate macromonomers, thereby supporting in situ polymerization strategies suitable for scalable manufacturing processes such as extrusion [3]. Thus, a careful investigation of the behavior of these polymerizable systems is imperative.

In our work, three-component polymer electrolytes were developed. Specifically, a poly(ethylene glycol) methyl ether acrylate (PEGMEA) macromonomer with a pendant of approximately nine ethylene oxide units, was polymerized via free-radical polymerization in the presence of varying concentrations of a glycol-based plasticizer and a Hückel-anion lithium salt. The influence of each constituent on the polymerization kinetics was systematically examined and the physicochemical characteristics and microstructural features of the resulting polymer electrolytes were directly correlated with their ion transport properties. These insights enable the rational design of room-temperature GPEs based on comb-like PEO architectures for scalable solid-state lithium batteries.

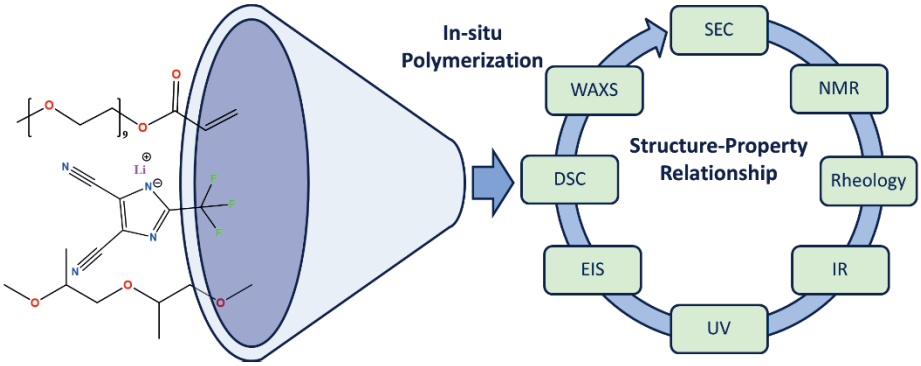

Fig. 1. Investigation of structure-property relationships in in-situ polymerization of PEGMEA.

References

[1] D. Zhou, D. Shanmukaraj, A. Tkacheva, M. Armand, G. Wang, *Chem*, **5** (2019) 2326-2352.
[2] X. Wang, Z. Song, H. Wu, J. Nie, W. Feng, H. Yu, X. Huang, M. Armand, Z. Zhou, H. Zhang, *ChemElectroChem*, **9** (2022) e202101590 (1-6).
[3] Z. Jia, Y. Liu, H. Li, Y. Xiong, Y. Miao, Z. Liu, F. Ren, *Journal of Energy Chemistry*, **92** (2024) 548-571.

Engineering Lithium-Ion Transport Through Multiscale Hybrid Polymer Electrolyte Architectures

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Hybrid polymer-based separators are emerging as key components to overcome ion transport limitations in next-generation lithium-ion batteries.[1] Here, we investigate lithium-ion transport in hybrid electrospun separators (HES) composed of interpenetrated PVDF-HFP polymer domains and inorganic SiO_2 networks, forming highly porous and tuneable ion-conducting pathways. Their unique fractal-like architecture,[2] characterised by multiscale X-ray scatterings, enables a direct correlation between morphology and ion transport in confined liquid electrolytes within polymer-based porous architectures (Figure 1).

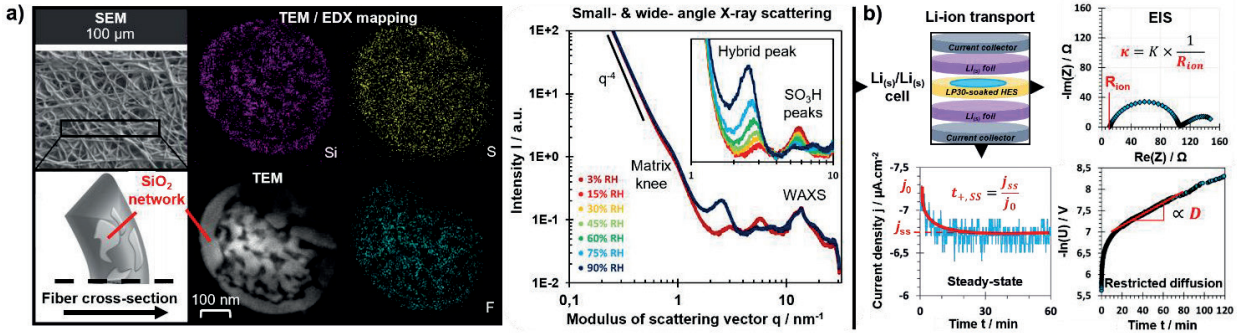

Figure 1. Structure–transport relationship in hybrid electrospun separators (HES): (a) multiscale architecture; (b) experimental workflow for quantifying lithium-ion transport properties (κ, D, t_{+}, $t_{+,\,Ne}$).

Within the concentrated solution theory framework[3] developed by Newman and Balsara, key transport parameters—ionic conductivity (κ), diffusion coefficient (D), and lithium transference number (t_{+})—were determined using electrochemical methods in symmetric $Li_{(s)}/Li_{(s)}$ cells. Concentration cell measurements further enabled access to the Newman transference number ($t_{+,\,Ne}$), providing insight into ion transport in confined polymer electrolytes. Compared to commercial polyolefin separators (Celgard 2325), HES exhibit significantly enhanced transport properties, including higher ionic conductivity, faster lithium diffusion, and increased lithium transference numbers, as confirmed by both electrochemical measurements and PFG-NMR (Table 1). These results demonstrate that tailored polymer/inorganic architectures with controlled electrolyte confinement strongly promote lithium-ion transport. This work highlights the critical role of multiscale architecture in governing ion transport, providing a framework for the rational design of advanced hybrid ion-conducting systems.

Table 1. Enhanced lithium-ion transport in hybrid electrospun separators: transport properties at 25 °C for bulk LP30 and LP30 confined in conventional and hybrid polymer-based separators.

Sample	κ (mS·cm^{-1})	D (10^{-10} m^2·s^{-1})	$t_{+,\,PFG\text{-}NMR}$
Bulk LP30	12.3	3.0	0.39
LP30 in Celgard 2325	0.240	0.72	0.30
LP30 in pristine HES	2.22	4.80	0.86
LP30 in sulfonated-HES	-	-	0.85

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References

[1] M. Winter et al., *Chem. Rev.*, **118** (2018) 11433–11456. [2] L. Dos Santos et al., *Adv. Funct. Mater.*, **26** (2016) 594–604. [3] N.P. Balsara, J. Newman, *J. Electrochem. Soc.*, **162** (2015) A2720–A2722.

Transport and mechanical properties of pseudo-zwitterionic polymer electrolytes from non-equilibrium molecular dynamics simulations

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Pseudo-zwitterionic polymer electrolytes (PZPEs) have emerged as promising candidates for multifunctional components in solid-state batteries, where efficient ion transport must be reconciled with mechanical robustness and long-term stability[1-2]. However, quantitatively linking polymer composition to coupled transport and mechanical properties remains challenging due to the high computational cost of conventional equilibrium simulation approaches[3-4]. In this work, we employ the non-equilibrium molecular dynamics (NEMD) simulations[5-6] to systematically investigate how compositional variations in PZPEs govern both ionic transport and mechanical response. The calibrated NEMD approach enables very efficient and robust evaluation of key material properties, including ionic conductivity, Young's modulus, and bulk modulus. Our results reveal a clear composition–property map that highlights the dual role of zwitterionic motifs in modulating ion mobility and mechanical strength. This work establishes a computational workflow for the fast screening of transport and mechanical properties of polymer electrolytes and provides a molecular-level understanding of PZPEs for their potential self-healing capability.

References

[1] https://www.salamander-eu.com/
[2] K. Edström, R. Dominko; M. Fichtner; S. Perraud, *Battery 2030+ Roadmap*, (2020)
[3] H. Gudla, C. Zhang, D. Brandell, *J. Phys. Chem. B*, **124** (2023) 8124–8131
[4] Y. Shao, H. Gudla, J. Mindemark, D. Brandell, C. Zhang, *Acc. Chem. Res*., **57** (2024) 1123–1134
[5] H. Gudla, K. Edström, C. Zhang, *ACS Materials Au,* **4** (2024) 300–307
[6] S. Lee, C. J. Rasaiah, *J. Chem. Phys.*, **101** (1994) 6964–6974

New Family of Protic Poly(diallyl ammonium)s and its Application as Fluorine-free Water-processable Binders for Batteries

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Nowadays, fluorinated electrochemically stable polymer PVDF is the common binder which requires toxic N-methyl-2-pyrrolidone (NMP) as a processing solvent. This toxicity has motivated a growing interest in water-processable, fluorine-free alternatives. Aqueous binders such as carboxymethyl cellulose (CMC), polyacrylic acid (PAA), alginates, and other polysaccharide-based systems have been explored extensively[1]. However, they often suffer from intrinsic limitations: low electrochemical stability at high voltages, incompatibility with certain active materials, poor processability, and insufficient ionic conductivity. These limitations have driven the exploration of functional binders based on poly(ionic liquid)s, which combine aqueous processability, electrochemical stability, and ionic conductivity[2,3].

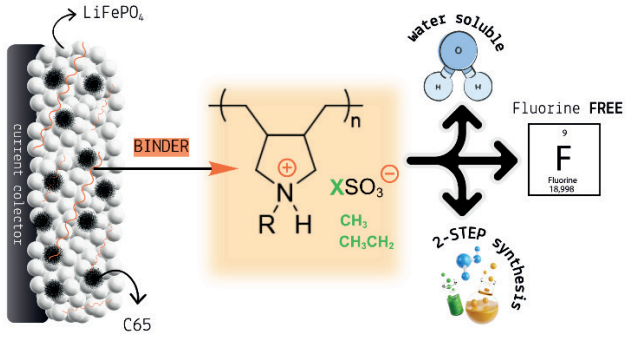

Figure 1. Sustainable fluorine-free protic poly(ionic liquid) binders for $LiFePO_4$/C composite electrodes

In this work, we present a new family of fluorine-free protic poly(ionic liquid)s, based on a PDADMA-like backbone, incorporating two cations ($DAMA^+$, $DAAH_2^{2+}$) and three sulfonate anions (MsO^-, EsO^-, $DBsO^-$), enabling a systematic investigation of the relationship between ionic structure and functional properties. Four of these polymers are completely water-soluble and were evaluated as sustainable binders for $LiFePO_4$ (LFP) cathodes. Electrochemical characterization by cyclic voltammetry confirmed stability up to 4.5 V vs. Li/Li^+, enabling application as binders in lithium-ion batteries. When used as aqueous-processable binders for $LiFePO_4$ cathodes, these materials provide competitive rate capability and enhanced performance at high current densities compared to PVDF, highlighting their potential as fluorine-free sustainable alternatives for aqueous electrode manufacturing.

References

[1] A. C. Rolandi, I. de Meatza, N. Casado, M. Forsyth, D. Mecerreyes, and C. Pozo-Gonzalo, Jun. 28, 2024, *Royal Society of Chemistry*.
[2] A. C. Rolandi, C. Pozo-Gonzalo, I. de Meatza, N. Casado, D. Mecerreyes, and M. Forsyth, *Advanced Energy and Sustainability Research*, vol. 4, no. 12, Dec. 2023.
[3] J. S. Lee, K. Sakaushi, M. Antonietti, and J. Yuan, *RSC Adv.*, vol. 5, no. 104, pp. 85517–85522, 2015.

Poly(Ether-Ester) based Solid Polymer Electrolytes derived from Cyclic Ketene Acetal and its Copolymers

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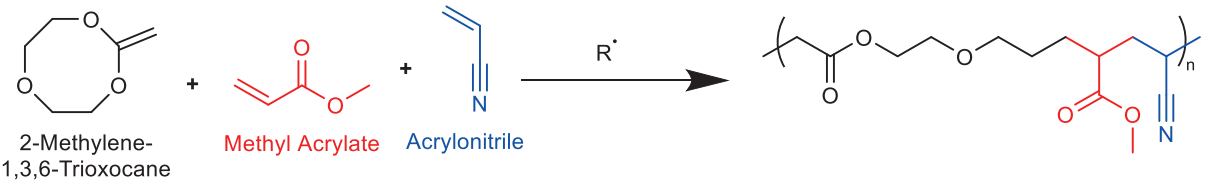
Solid polymer electrolytes (SPEs) are safer alternatives to liquid electrolytes for lithium metal batteries, offering advantages in processability, mechanical integrity, and cell safety. However, the development of polymer backbones that enable both high ionic conductivity and sufficient electrochemical stability remains a major challenge[1]. In this work, we report the synthesis of a novel class of poly(ether-ester)-based SPEs obtained by radical ring-opening copolymerization of 2-methylene-1,3,6-trioxocane and its copolymers with methyl acrylate and acrylonitrile. Radical ring-opening polymerization (rROP) is a versatile technique that combines the benefits of both ring-opening and radical polymerization systems. It enables the incorporation of heteroatoms, such as oxygen, nitrogen, or sulfur, directly into the polymer backbone, which is challenging in the traditional radical polymerization of vinyl monomers[2]. This molecular design combines the flexible ether segments that facilitate lithium-ion transport with polar nitrile and ester groups that enhance lithium salt dissociation and offer oxidation stability. Comprehensive thermal analysis using DSC and TGA confirms the amorphous nature of the copolymers and their excellent thermal stability, with degradation temperatures well above typical battery operation limits. Electrochemical measurements further demonstrate a broad electrochemical stability window up to 4.7 V versus Li/Li$^+$, making these materials compatible with high-voltage cathodes. The ionic conductivity reaches 4×10^{-7} S cm^{-1} at 60 °C, indicating sufficient segmental motion and ion transport through the polymer matrix. Symmetric Li|SPE|Li cells exhibit remarkable cycling stability for over 500 h without short-circuiting, confirming stable interfacial behavior. When paired with $LiFePO_4$ cathodes, the polymer electrolyte delivers an initial discharge capacity of 151 mAh g^{-1} at 60 °C, with good cycling retention. These results demonstrate that poly(ether-ester) matrices derived from 2-methylene-1,3,6-trioxocane offer a versatile platform for designing thermally robust, electrochemically stable polymer electrolytes suitable for next-generation all-solid-state lithium batteries.



Scheme 1: Copolymerization scheme of 2-methylene-1,3,6-trioxocane with methyl acrylate and acrylonitrile

References

[1] J. Mindemark, M.J. Lacey, T. Bowden, D. Brandell, *Prog. Polym. Sci*. **81** (2018) 114–143.
[2] Y. Deng, F. Mehner, J. Gaitzsch, *Macromol. Rapid Commun*. **44** (2023) 2200941.

Single-ion polymer electrolytes enabling 5 V Li-LNMO solid-state batteries

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Electrochemical energy storage is pivotal for transport electrification and renewable integration, motivating solid-state batteries. Solid polymer electrolytes (SPEs) improve safety and manufacturability versus liquid electrolytes and can enable lithium-metal anodes and high-voltage cathodes. In single-ion conducting polymers (SiCP), the anion is covalently tethered to the backbone, making Li^+ the dominant mobile species and suppressing Li dendrites formation, but practical use is often limited by high glass-transition temperatures, insufficient conductivity, and processing complexities [1].

Here, we report the development of a hybrid single-ion solid polymer electrolyte (HSiPE) designed to couple mechanically robust membranes (compatible with pouch-cell assembly) and stable electrochemical performance at elevated temperatures. Our work integrates electrolyte formulation design and processing into a single scalable workflow. We used a one-pot synthesis incorporating a high fraction of plasticizers directly into the single-ion polymer network, eliminating post-fabrication swelling/soaking steps while maintaining solid electrolyte membrane integrity and improving scalability. In this work, effects of molecular weight of SiCP, additives, various plasticizers and ceramic fillers were investigated to improve the ionic conductivity and high-voltage stability of the HSiPE electrolytes. In *Figure 1a*, ionic conductivity of investigated HSiPE electrolytes acted as initial filter on electrolyte selection process for further characterization in solid-state cells. HSiPE_19 (without any fillers), HSiPE_20 (with Al_2O_3) and HSiPE_28 (lithium-ion conducting glass-ceramics LICGC™, Ohara Corp.) electrolytes were selected for testing in 5 V "Li/HSiPE/LNMO" based cells. In solid-state cells cycled between 3.0 to 5.0 V, the cells with electrolytes with ceramic fillers (HSiPE_20 and HSiPE_28) demonstrated higher stable electrochemical performance, achieving a discharge capacity of ~140 mAh/g (relevant to LNMO theoretical capacity) at 60 ºC, as seen in *Figure 1b*.

In addition to the results summarized here, our contribution includes additional formulation-screening and cell data, including ionic conductivity values, Li|Li symmetric-cell cycling, and full-cell testing with 5 V-class LNMO composite cathodes at 60 °C.

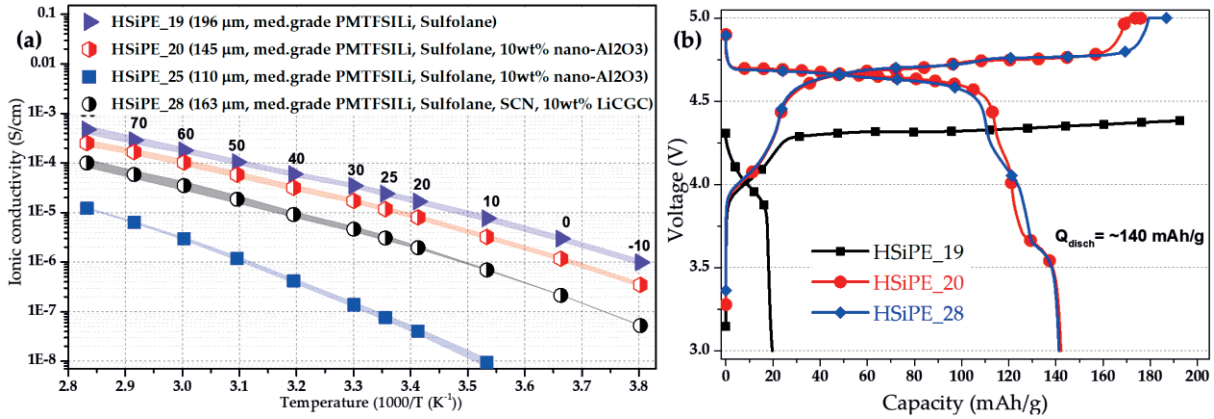

Figure 1. a) Ionic conductivity of the investigated HSiPEs; b) Charge-discharge profiles for 1st cycle of "Li/ HSiPE /LNMO" solid state coin cells. Cell configuration: composite LNMO cathode (LL 0.7-1.5 mAh/cm^2), Li foil (50 µm) as anode. Cycling conditions: 60 ºC, C/30-C/30, 3.0–5.0 V.

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References

[1] Y. Shao, F. Alloin, D. Bresser, C. Iojoiu, *J. Mater. Chem. A,* 2024,12, 30032-30040

Waterborne high-loading Ni-rich cathodes tailored for semi-solid-state lithium metal batteries

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Semi-solid-state lithium metal batteries (sSS-LMBs) based on *in situ* polymerized gel polymer electrolytes (ISP-GPEs) are a promising battery technology which combines high energy density, low internal resistance, and easy scalability. However, the development of sSS-LMBs requires careful tailoring of cathode active layer architectures to ensure effective GPE infiltration and sufficient ionic transport. Design rules established for conventional liquid-electrolyte LIBs, where electrode densification in the 3.0–3.6 g·cm^{-3} range is typically pursued to maximize volumetric energy density, are not directly transferable to ISP-GPE-based systems, particularly at high areal capacities.

Herein, an environmentally friendly water-based process [1] has been optimized and employed to dense Ni-rich cathodes with high areal capacity of 3 mAh·cm^{-2} used for sSS-LMBs application. An aqueous binder based on a single-ion conducting copolymer electrode formulation has been incorporated into the formulation to promote ionic transport within the structure. The effect of positive electrode density on the electrochemical performance of semi-solid-state "Ni-rich NMC/GPE/Li" cells has been systematically investigated. As shown in Figure 1, increasing cathode density from 2.5 to 3.4 g·cm^{-3} induces discharge C-rate limitations. While cells with higher-density electrodes retain comparable capacities at low discharge C-rates, a marked decrease is observed as the discharge C-rate increases. The cells with cathodes having the lowest density 2.5 g·cm^{-3} (~27% porosity) delivers significantly higher discharge capacities at moderate-to-high C-rates compared to cells with more compacted cathodes. These results highlight the need to rebalance cathode design strategies for ISP-GPE-based sSS-LMBs, prioritizing adequate ionic accessibility over maximum electrode compaction.

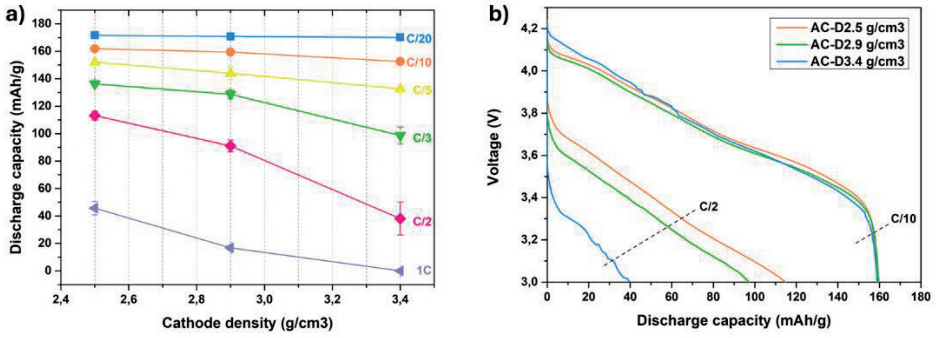

Figure 1. Electrochemical performance of "Ni-rich NMC/GPE/Li" semi-solid state coin cells: (a) discharge capacity vs. cathode density at selected discharge C-rates, (b) discharge voltage profiles at C/10 and C/2 for cells with different cathode densities. Cycling conditions: 25 °C, cathode loading 3 mAh·cm^{-2}, Li metal - 50 µm, charge rate - C/10, discharge - 0.1–1C, cycling range - 3.0–4.3 V.

This work is supported by Horizon Europe under the SOLIDBAT project (Grant Agreement No. 101147533).

References

[1] S. Sananes-Israel, I. Urdampilleta, G. Kvasha, I. Landa-Medrano, I. de Meatza, Batter. Supercaps, 7 (2024) e202400358.

Beyond LiTFSI: New Generation PFAS-free Salts for Enhanced Solid Polymer Electrolytes in Lithium Metal Batteries

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LiTFSI/PEO system, although it is the most employed system in SPEs, still presents some limitations such as low lithium-ion conductivity, poor SEI layer and poor electrochemical stability towards Al current collector.[1] Additionally, utilization of fluorinated compounds in lithium secondary batteries, including per- and polyfluoroalkyl substances (PFAS), raises environmental concerns, prompting a shift toward PFAS-free alternatives.[2] Hence, the development of new electrolyte materials is needed to fulfill with regulatory scrutiny and to improve battery performance addressing current limitations, particularly in enhancing Li-ion conductivity and stabilizing the SEI layer at the lithium anode. In addition to the widely exploited sulfonimide-based anions[3], Hückel anions bearing cyanide substituents emerge as a promising alternative due to their strong negative charge delocalization, high ionic conductivity, excellent thermal stability, and wide electrochemical stability window.[4]

In this work, we present the synthesis and characterization of two novel PFAS-free lithium salts and their application in lithium metal batteries. When incorporated in a PEO-based SPE, both salts demonstrate excellent thermal stability for use in LMBs and show enhanced compatibility and cycling stability with the lithium anode. These findings highlight the potential of PFAS-free materials to be competitive options for next-generation, high-performance solid-state batteries.

References

[1] M. B. Armand, *Rev. Mater. Sci,* **16** (1986), 245-261.
[2] (a) E. M. Sunderland, X. C. Hu, C. Dassuncao, A. K. Tokranov, C. C. Wagner, J. G. Allen, *J Expo Sci Environ Epidemiol*, **29** (2019), 131-147; (b) S. E. Fenton, A. Ducatman, A. Boobis, J. C. DeWitt, C. Lau, C. Ng, J. S. Smith, S. M. Roberts, *Environmental Toxicology and Chemistry*, **40** (2020), 606-630.
[3] (a) H. Zhang, F. Chen, O. Lakuntza, U. Oteo, L. Qiao, M. Martinez-Ibañez, H. Zhu, J. Carrasco, M. Forsyth, M. Armand, *Angewandte Chemie International Edition*, **58** (2019) 12070-12075. (b) M. Martinez-Ibañez, E. Sanchez-Diez, U. Oteo, I. Gracia, I. Aldalur, H. B. Eitouni, M. Joost, M. Armand, H. Zhang, *Chem Matter*, **34** (2022), 3451-3460.
[4] (a) Bitner-Michalska, G. M. Nolis, G. Żukowska, A. Zalewska, M. Poterała, T. Trzeciak, M. Dranka, M. Kalita, P. Jankowski, L. Niedzicki, J. Zachara, M. Marcinek, W. Wieczorek, *Sci Rep*, **7** (2017) 40036. (b) G. Z. Żukowska, M. Piszcz, K. Gańko, M. Więckowski, M. Królikowski, M. Poterała, M. Dranka, *Materials Chemistry and Physics*, **315** (2024), 128999.

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Desing & optimization of dry processing for *ex-situ* gelled polymer electrolytes

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Nowadays, one of the main goals of the energy storage industry is to transition from conventional lithium-ion technologies toward next-generation systems beyond Li-ion, based on more abundant, sustainable, and less environmentally impactful materials. In that context, Solid-State-Batteries have emerged as one of the most interesting alternatives, as they only employ solid components throughout the entire cell. This configuration enhances safety compared to conventional batteries and provides improved mechanical stability; however, their main limitation lies in the relatively low ionic conductivity of solid electrolytes.[1] Gel-Polymer-Batteries are a variant that can possibly solve those problems, as they combine both liquid and solid-state features.[2] However, gel-polymer electrolytes and jellified cathodes are still commonly processed by wet methods,[3] which rely on organic solvents and drying steps, increasing process complexity, energy demand, and environmental impact. For this reason, dry-processing approaches are attracting growing interest, as they eliminate solvent use while still requiring a careful balance between mechanical integrity and electrochemical performance.

This study aims to establish a solvent-free process to manufacture gel-polymer electrolytes and jellified composite cathodes. The materials will be processed using a torque-controlled HAAKE mixer, which applies shear through rotors while enabling control of temperature and rotor speed, without the use of solvents. The objective is to obtain a self-standing, processable gel with good mechanical properties and ionic conductivity. To optimize the process, the effect of the mixing parameters, addition order, PEGDME content and EO:Li ratio on the electrochemical and mechanical properties will be investigated. The resulting insights will be used as a basis for the development of jellified composite cathodes.

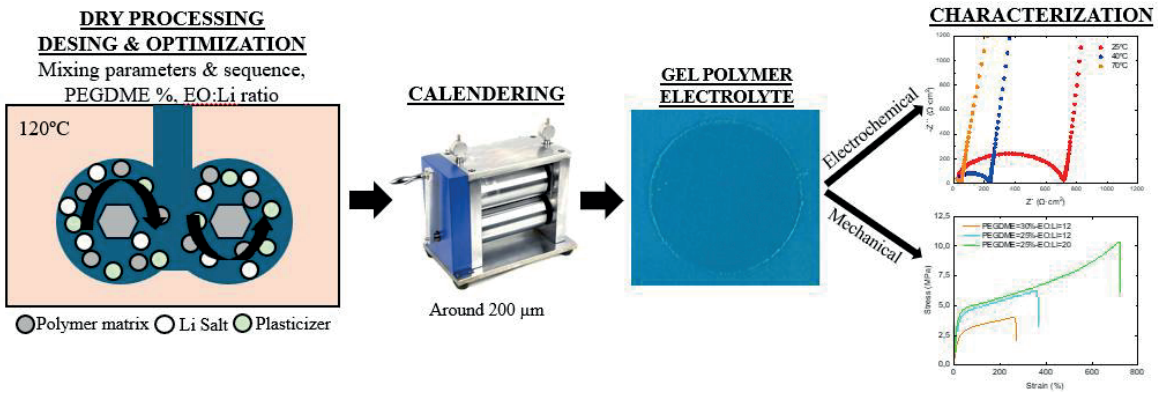

Figure 1. Scheme of the Gel-Polymer Electrolyte manufacturing process

[1] A. Joshi, D. Kumar Mishra, R. Singh, J. Zhang, Y. Ding, *Applied Energy*, **386** (2025) 125546-125566.

[2] S. Guo, X. Li, Z. Zhang, X. Xu, H. Wang, C. Zhao, Y. Gu, *Energy Materials*, **5** (2025) 500041-500061.

[3] J. Castillo, A. Santiago, X. Judez, J. A. Coca Clemente, M.C. Morant-Miñana, A. Villaverde, J.A. Gonzalez-Marcos, H. Zhang, M. Armand, C. Li, *Chemistry of Materials*, **33** (2021) 8812-8821.

Ionic Transport properties of concentrated liquid electrolytes, beyond Debye Hückel Onsager (DHO) model

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The scramble to find an electrolyte suitable for Lithium metal battery gave way to a lot of innovations for gel and polymer electrolytes[1]. At the same time, for about fifteen years, there is a resurgence of interest in highly concentrated electrolytes[2]. The absence of free solvent and the modifying of the activity of the solvent greatly improved the electrochemical stability of those electrolyte[3] and thus make them more suitable versus lithium metal negative electrode.

This improvement of stability come with drawbacks. As the number of charge carriers increases in the medium their mobilities start to decrease, around 1M[2], causing the well-known bell-shape curve for conductivity curve versus concentration. As more salt is put in the solvent the viscosity rises, causing a slowing of the ionic mobilities due to the viscous hindering. Also, it has been commonly proven that at higher concentration even in strong electrolyte such at cyclic carbonate (eg propylene carbonate PC) complex charges carriers appears[1] combining anions, cations and solvent molecules (ion pairing and aggregation). It's not yet clear if those charge carriers contribute to the ionic conduction and to what extent[4].

In this presentation, the nature of charge carrier will be analysed on a wide range of concentrations for 2 types of carbonates- cyclic PC ε_r=64.9 at 25°C (strong electrolyte) and linear DMC with ε_r=3.1 at 25°C[5] (weak electrolyte), laden with LiTFSI (0,01 to 3,2 mol/L). The link between the transport properties, i.e., ionic conductivity, viscosity and diffusion coefficients, and the nature and quantity of charge carriers characterized by Raman spectroscopy, will be investigated. Then, thanks to a complete set of measured parameters for these 2 carbonates solvents, the results are confronted to the historical and current models for transport in concentrated liquid electrolytes as function of temperature. For very low concentration verify the adequation of experimental data to the DHO model, extend the model to moderate and high concentration for carbonates electrolytes.

Sources:

(1) Long, L et al. *J. Mater. Chem. A* **2016**, *4* (26), 10038–10069. https://doi.org/10.1039/C6TA02621D.
(2) Borodin, O. et al. *Joule* **2020**, *4* (1), 69–100. https://doi.org/10.1016/j.joule.2019.12.007.
(3) Zheng, J et al. *Advanced Science* **2017**, *4* (8), 1700032. https://doi.org/10.1002/advs.201700032.
(4) Yamada, Y et al. *Nat Energy* **2019**, *4* (4), 269–280. https://doi.org/10.1038/s41560-019-0336-z.
(5) Xu, K. *Chem. Rev.* **2004**, *104* (10), 4303–4418. https://doi.org/10.1021/cr030203g.

Electrospun Sodium-Ion Conducting PAN/PEO-Based Electrolytes for ASSBs

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Transitioning away from long-term dependence on fossil fuels and nuclear energy is essential to meeting the Paris Agreement's 1.5 °C target and reducing energy import demands.[1] Cutting CO_2 emissions while maintaining a reliable energy supply requires effective storage of renewable energy from wind, water, and solar sources, with all-solid-state batteries (ASSBs) being a promising candidate.
We present a solid polymer electrolyte manufactured via electrospinning, suitable for use as a separator or catholyte in ASSBs. In our previous work, we incorporated polyacrylonitrile (PAN), a structural support polymer, into ion-conductive polyethylene oxide (PEO), producing a phase-separated fiber structure.[2] The resulting free-standing membrane offers mechanical flexibility, low crystallinity, and significantly improved thermal stability, maintaining integrity up to 100 °C. Lithium transport through the electrolyte was confirmed using galvanostatic lithium plating/stripping at 0.1 mA cm^{-2} and solid-state NMR spectroscopy.[3]

However, the growing demand for lithium-based battery technologies has driven up costs and reduced availability. In addition, most global lithium production is concentrated in Australia, Chile, and China, raising concerns about geopolitical independence and supply security. To address these challenges, sodium emerges as a substitute for lithium, as it is low-cost, widely available, more evenly distributed geographically, and can be extracted in a more environmentally sustainable way.[4]
Here, we apply the sodium salts $NaBF_4$ and NaTSFI to the electrospun PAN/PEO system. While using $NaBF_4$ in polymer electrolytes, which shows ionic conductivity in the order of 10^{-7} S cm^{-1} at 55 °C, doesn't match the performance of lithium-based polymer electrolytes, NaTFSI is a promising candidate. We screened different compositions of sodium salt and plasticizers, succinonitrile and propylene carbonate, to identify the optimal system. Using potentiostatic electrochemical impedance spectroscopy and X-ray diffraction, we demonstrate the plasticizing effect of NaTFSI. No additional plasticizer and only a small amount of NaTFSI (1/36 molar ratio referenced to the polymers' repetition unit) are sufficient to achieve a conductivity of 0.01 mS cm^{-1} at 55 °C. Furthermore, we confirmed the mechanical, thermal, and electrochemical stability of the lithium-based system by thermogravimetric analysis, differential scanning calorimetry, scanning electron microscopy, and cyclic voltammetry, making it a promising solution for safer, more durable battery systems. Our elastic, dense, and free-standing membranes demonstrate the vast potential of electrospun solid polymer electrolytes in beyond lithium all-solid-state battery systems.

References

[1] *The Paris Agreement,* **COP21**, 2018.
[2] A. M. Kirchberger, P. Walke, J. Venturini, L. van Wüllen, T. Nilges, *Membranes,* **15** *(*2025) 196.
[3] E. B. Springl, D. Sarkar, M. Mühlau, V. K. Michaelis, T. Nilges, *Inorg. Chem.*, **64** (2025) 19752.
[4] M. M. Mashfy, T. A. Alvy, N. Hossain, M. A. Haque, F. T. Mohsin, T. Sharmin, M. Nasim, *Next Energy,* **10** (2026) 100478.

PREPARATION OF ANION CONDUCTING PLASTICIZED POLYMER ELECTROLYTES BY THIOL-ENE CLICK CHEMISTRY, FOR ANIONIC ORGANIC BATTERIES

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Li-ion batteries are currently the most common energy storage devices. They have a high gravimetric energy density of 260 $Wh.kg^{-1}$ [1]. However, they contain critical materials such as cobalt or nickel which are not naturally present in Europe and are excavated through destructive mining processes[2]. It is therefore necessary to develop more sustainable energy storage solutions, such as organic batteries for example. Organic batteries have the advantage of being composed of abundant materials, such as C, H, O, S, which lower their environmental impact [3], but their performances are not on par with the current commercial Li-ion batteries. Among the multiple challenges of such systems is the solubility of the organic active materials in liquid organic electrolytes. This hinders the performance and the cycling stability of the battery.

In the context of more sustainable energy storage, France 2030 finances via the PEPR batterie, projects such as SONIC (Solid-state Organic anioNIC battery). The SONIC project aims to prepare an all-solid-state all-organic anionic battery, that contains no metal and no critical raw material, operating with organic electrode materials and a solid polymer electrolyte (SPE).

Herin, we present the preparation of an anion-conducting SPE, with commercially available monomers, via thiol-ene click chemistry, plasticized with carbonate solvents, propylene carbonate (PC) or ethylene carbonate (EC). The composition of the plasticized SPE was optimized, and a conductivity of 10^{-4} $S.cm^{-1}$ at room temperature was obtained. The preparation of the dry and plasticized SPEs and the conductivity measurement are presented on Figure 1.

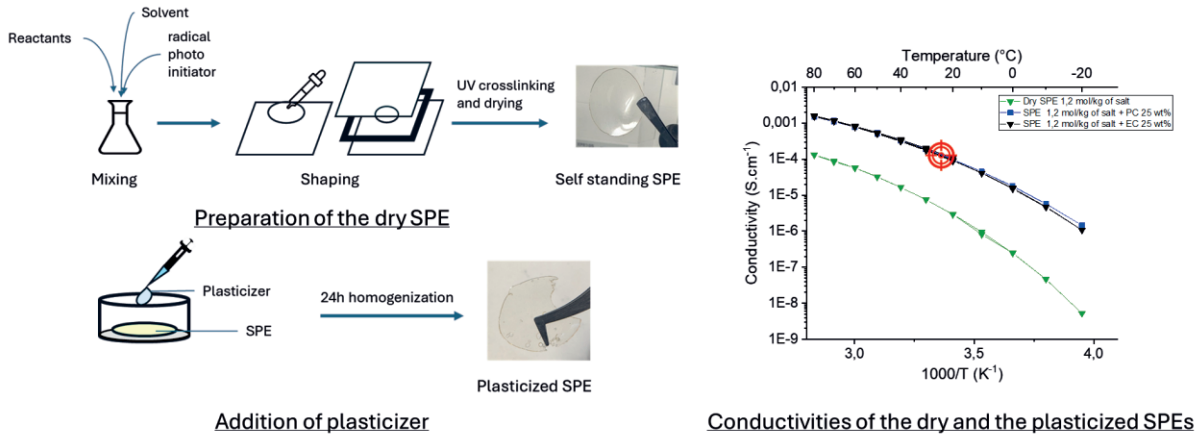

Figure 1: Preparation of the dry and plasticized SPEs and measured conductivity of the prepared membranes

References

[1] J. Światowska, D. Giaume, *Conversion de l'énergie électrique* **2024**.
[2] M. Salado, E. Lizundia, I. Oyarzabal, D. Salazar, *Physica Status Solidi (a)* **2022**, *219*.
[3] P. Poizot, J. Gaubicher, S. Renault, L. Dubois, Y. Liang, Y. Yao, *Chemical reviews* **2020**, *120*, 6490.

Single-step electrografting of copolymer electrolytes with enhanced mechanical integrity for separator-less structural sodium batteries

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Electrografting sodium-based solid polymer electrolytes (SPEs) presents a promising route toward electrochemically enhanced structural and solid-state sodium batteries. This approach enables the replacement of conventional monofunctional, thick separators with conformal ultrathin SPE coatings, which can improve both energy and power density. Previous work has shown electrografting to be a scalable, single-step method for forming such homopolymer electrolyte layers on carbon fibres that can be used as negative electrodes in separator-less structural sodium batteries.[1] However, the ability of these coatings to function as both electrolyte and separator depends critically on their coupled electrochemical and mechanical properties, which are intrinsically coupled and often competing. While their minimal thickness underpins the electrochemical advantages of reduced ionic transport distances and low internal resistance, it also requires them to be mechanically robust to maintain electrode separation and prevent dielectric breakdown or mechanically induced internal short circuits.

Here, we investigate electrografted copolymer SPEs formed from monofunctional and bifunctional PEG-acrylates coated onto carbon fibres. Three copolymer compositions are examined and compared with homopolymer-coated and pristine fibres. Introducing the bifunctional PEG-acrylate improves the mechanical properties of the resulting copolymer coatings compared to the homopolymer coating while they remain micrometre thin, flexible, and ionically conductive. The mechanically improved copolymer SPEs ensure electrode separation in half-cell configurations against sodium metal, while their maintained flexibility enabling fibre handling. Their low thickness combined with adequate ionic conductivity introduces negligible system resistance and no difference in electrochemical performance is observed for the copolymer coated fibres compared to the homopolymer coated and pristine fibres. However, an electrochemical threshold is reached at the highest evaluated content of bifunctional monomer content in the monomer mixture (18 mol%), where the added resistance of the resulting copolymer SPE significantly suppresses electrochemical performance.

Post-cycling analysis shows that all SPE coatings remain intact within the 0.01-2.00 V vs Na/Na^+ potential window used to cycle the half-cells. Further, the C-C bond between the fibre surface and the SPE appears stable within a voltage window of 0.01-4.00 V vs Na/Na^+ which indicates suitability to function as SPE and separator in full cells.

These results demonstrate that electrografted PEG-acrylate copolymers provide ultrathin SPE coatings with improved mechanical robustness without compromising electrochemical performance, offering a scalable strategy for separator-less structural and solid-state battery architectures.

References

[1] E. Lind, V. Nieboer, M. Cattaruzza, M. Johansson, K. Odelius, D. Zenkert, G. Lindbergh, EES Batteries, (2026) 1–11. DOI: 10.1039/d5eb00212e.

Polymer Electrolytes Towards Structural Lithium-Sulfur Batteries

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Global demand for clean energy has driven the need for advanced storage systems beyond Li-ion batteries (LiBs), which are rapidly approaching their theoretical limits. Research into 'beyond LiB' systems is therefore necessary for the electrification of energy intensive industries such as transport and aerospace. Structural LiBs offer a promising approach by increasing system-level energy density. By simultaneously storing energy and bearing mechanical load, they eliminate the heavy packaging materials required in conventional battery packs. This enables direct structural integration into cars, aircraft, drones, and other weight-sensitive systems. Yet the practical cell-level energy density of structural LiB remains low.

Another way to increase energy density further is to use LiS chemistry which has a far superior theoretical energy density from the multi-step redox of S in the cathode. LiS batteries (LiSBs) also offer a more sustainable alternative by replacing critical cathode minerals with abundant S. However, their commercialisation is severely limited by short cycle life. The incorporation of LiS chemistry into a structural battery remains unexplored, and its success would enable a battery with the commercially viable energy density for next-generation transport and aerospace technologies.

The electrolyte is key to the practical realisation of such a system. It must mitigate issues associated with LiS chemistry while also contributing to battery structural integrity. Polymers are particularly well suited for such applications, as their versatile chemistry enables a high degree of tunability to target these challenges. Li metal dendrite growth and the polysulfide shuttle effect (PSSE), where S discharge products migrate between electrodes, significantly limit LiSB performance. Replacing conventional liquid electrolytes (LE) with polymer electrolytes can address many of these issues thanks to their polymer network structure and functional groups. However, polymers used for structural composites are typically poor electrolyte candidates as the crystallinity that confers mechanical properties results in low ionic conductivity. To address this, structural biphasic electrolytes (SBEs) have been designed using a mix of typical LiB electrolytes with methacrylate resins containing the same bisphenol A backbone used in epoxy resins for carbon fiber (CF) composites. With a high-modulus polymer backbone phase and an ion-conducting liquid phase, they can simultaneously provide structural integrity and facilitate ion transfer, mitigating the typical tradeoff between the two.

This study compares a conventional gel polymer electrolyte (GPE) to a newly developed LiS SBE. A GPE with PEO-like characteristics and functional groups, known to help mitigate the PSSE, was chosen to allow comprehensive comparison of electrochemical behaviour. The GPE was prepared in situ during cell assembly followed by thermally initiated polymerisation. Combined with commercial cathodes, which can later be substituted by the CF/S cathodes required for structural batteries, the cycle life was increased compared to a LE and a range of benchmark metrics were achieved. The SBE, adapted from a LiB system, consists of bisphenol A ethoxylate dimethacrylate and a standard LiS LE (1:1 mixture of 1,3-dioxolane (DOL) and 1,2-dimethoxyethane (DME) with lithium bis(trifluoromethanesulfonyl)imide (LiTFSI)), where polymerisation is initiated by AIBN. The crosslinking reaction was investigated to understand and avoid unwanted side reactions with S species and their incorporation into the polymer chains. The effect of crosslinking degree and temperature on conductivity and mechanical properties was investigated using electrochemical impedance spectroscopy and dynamic mechanical analysis, while its subsequent cycling behaviour was assessed with galvanostatic charge-discharge tests. Full characterisation using scanning electron microscopy, X-ray photoelectron spectroscopy, and Raman spectroscopy enables further optimisation of both mechanical and electrochemical properties, working towards a truly multifunctional SBE for structural LiSBs.

Understanding the Effects of Structure and Composition on Mechanical Properties of Solid Polymer Electrolytes by Dynamic Viscoelasticity

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In widely studied polyether-based solid polymer electrolytes (SPEs), the flexibility of the ether chains results in a low glass transition temperature (T_g) and relatively high ionic conductivity at low salt concentrations. However, increasing the salt concentration raises the T_g and reduces the conductivity. This is generally attributed to a physical crosslinking between ether groups and cations. On the contrary, some SPEs, such as those based on poly(ethylene carbonate) (PEC), exhibit the opposite behavior: the conductivity increases, while the T_g decreases with increasing salt concentration. Although numerous materials have been tested, achieving a high ionic conductivity and favorable mechanical properties "simultaneously" is challenging because reasonable conductivity often coincides with relatively low T_gs. Polymeric materials are typical viscoelastic materials. Viscoelasticity is expected to significantly influence the mechanical properties of SPEs. However, few studies have thoroughly examined how the structure and composition of SPE affect the mechanical relaxation dynamics across broad timescales.

The present study examines the stress relaxation of polymers in SPEs via dynamic viscoelasticity measurements. Poly(ethylene oxide/propylene oxide) copolymer (P(EO/PO)) was selected as a polyether model, while polymers such as poly(ethylene carbonate) (PEC), poly(propylene carbonate) (PPC), and poly(trimethylene carbonate) (PTMC) were chosen as polycarbonate models. Stress relaxation of polymers is a sum of various modes across broad timescales, such as local or segmental motion, entanglement, and diffusion of the entire chains. Based on the time-temperature superposition principle, the relaxation modes that induce transitions at each temperature correspond to modes with different rates when seen at a certain temperature. Fig. 1 shows the results of temperature sweeps for PTMC and LiTFSI electrolytes as examples. From the low-temperature side, T_g-related large relaxations due to local or segmental motions are observed, followed by rubbery plateau regions, and then terminal relaxations. Notably, while the T_g shifts to higher temperatures, the G' at the plateau which corresponds to the degree of chain entanglement and the terminal relaxation temperature decrease. The salt addition suppresses the segmental motions, while exhibiting plasticization effect on entanglement and the overall chain diffusion. We obtained results indicating that the effect of salt addition varies depending on the materials used. In the presentation, we will discuss implications for the potential to achieve both favorable conductivity and mechanical properties, incorporating master curve analysis covering a broad frequency range.

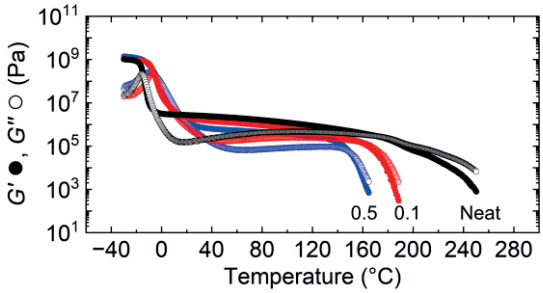

Fig.1. Results of temperature sweep dynamic viscoelasticity measurements for PTMC and its LiTFSI electrolytes ([Li]:[C=O] = 0.1, 0.5).

Bio-Inspired Ion Conducting Membranes

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The development of next-generation polymer electrolytes is critical for electrochemical energy technologies, particularly in hydrogen-based applications such as fuel cells and water electrolysis, where proton exchange membrane (PEM) and anion exchange membrane (AEM) systems play a central role. However, current commercial membranes are often made from per- and polyfluoroalkyl substances (PFAS), which raise environmental concerns and require a balance between ionic conductivity, chemical stability, and dimensional integrity.

Here, we present a bio-inspired approach based on molecular building blocks with a strong propensity for self-assembly, enabling the development of ion-conducting polymers through simple and scalable synthetic routes. Like biological membranes, these systems self-assemble into nanostructures with interconnected ionic domains, facilitating ion transport. A key advantage of this platform lies in its modularity, enabling the generation of diverse molecular structures via high-throughput and tunable synthetic approaches. Solvent casting and further chemical processing results in robust polymeric materials with enhanced mechanical integrity and stability. Importantly, the same design principles enable adaptation of the materials for both proton-conducting (PEM) and anion-conducting (AEM) systems. The developed membranes address key limitations of state-of-the-art systems across their lifecycle, from fabrication to operation. Their simplified processability could improve manufacturability and material quality, while minimal swelling and shrinkage reduces mechanical failure. The materials already show enhanced dimensional integrity and superior nanostructural stability at high temperatures compared to selected commercial membranes. Beyond membranes, the molecular precursors show strong potential as functional ionomers, highlighting their versatility across electrochemical applications.

Overall, this research establishes a simple and scalable platform for advanced ion-conducting polymers, offering a promising pathway toward PFAS-free and more sustainable membrane materials for hydrogen technologies.

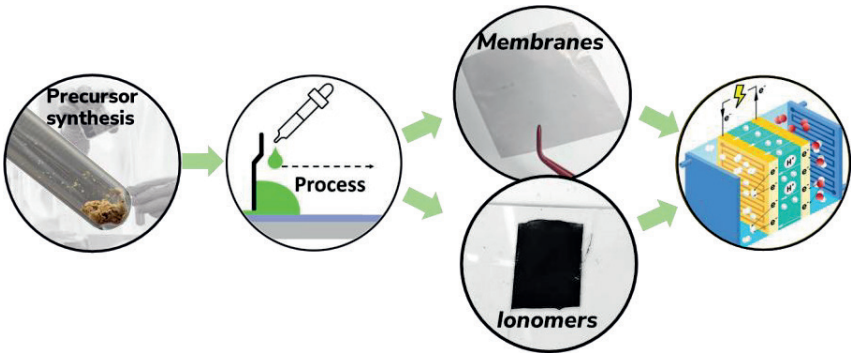

Figure 1: Schematic showing the preparation of ionomer and membrane fabrication

Probing SEI Evolution via a Model System in Solid Polymer Electrolytes with Pseudo-Operando X-ray Photoelectron Spectroscopy

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Solid polymer electrolytes (SPEs) show great promise when it comes to energy density and safety for Li ion batteries. The importance of solid electrolyte interphase (SEI) formation is well known and is understood in alkaline earth metal battery systems but their dynamic formation is still poorly understood. Surface characterisation of the SEI in SPEs can be specially challenging as the glue-like nature of polymers prevents a clean SPE/electrode separation.

In-operando studies are a viable strategy to probe the SEI formation dynamically. In one study, an open cell configuration was used with a 6 nm thin layer of nickel deposited on Li_6PS_5Cl, allowing the SEI evolution to be tracked using high energy x-rays[3].

A model system was developed to mimic the electrode/electrolyte interface and to track the SEI evolution in-operando. NMC 111, conductive carbon and PEO/LiTFSI (as binder) were coated on an Al mesh, which were then assemble into a half cell vs Li into a small coin cell, with an opening allowing the mesh to be probed via x-rays (Al source). The core levels of interest were investigated.

The C 1s spectra Show that As the Working electrode potential increases towards 4.5 V, the C-O (PEO peak) and C-F(salt peak) peaks start to shift to lower BE. This is while the peak intensity of the former decreases, while for the latter there is an increase. This agrees with the literature that PEO starts to degrade after 3.8 V and that the degradation products include volatile species such as methanol[1], 2-methoxyethanol[1], 1,4-dioxane[2], 2-methoxy-1-propanol[2], 1,2-ethylene glycol diformate[2], and 1,3,6-trioxocane[2].

The cell was then discharged to its initial potential to see if any of the peaks returned back to their initial state. The intensity of the peak was lowered alightly as expected, but the state of the peak spectra was similar to the spectrum collected at 3.8 V, with the difference that the salt peak was still more prominent.

The survey spectrum reveals that the metal peaks are visible, proving that the polymer binder is in contact with the NMC.

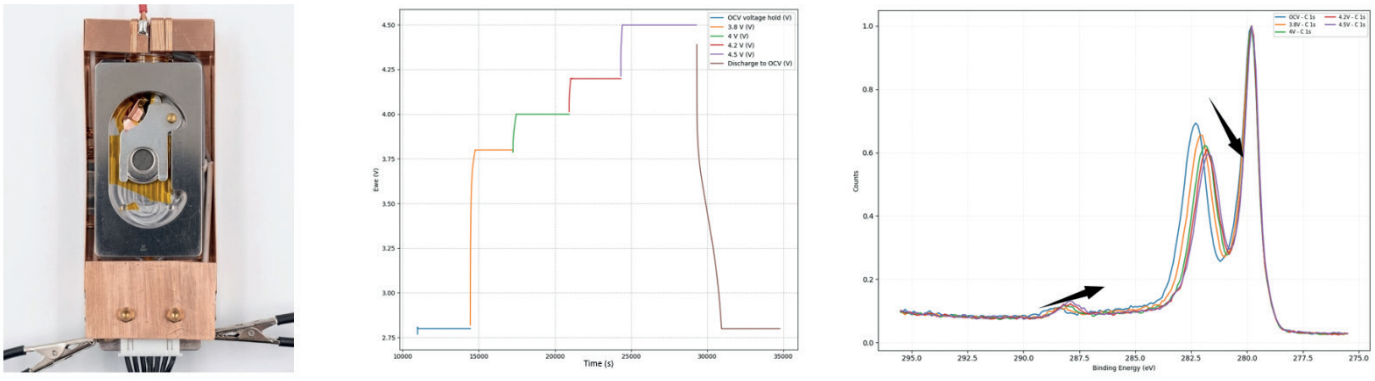

Figure. Left: Operando cell with coin-cell setup. Middle: Voltage profile, with measurements taking place towards the end of voltage plateaus. Right: C 1s core level spectra of working electrode during the voltage plataus.

References

[1] L. Seidl, R. Grissa, L. Zhang, S. Trabesinger, and C. Battaglia, *Advanced Materials Interfaces*, **9** (2022), 2100704.
[2] L. Herbers *et al.*, *Advanced Energy and Sustainability Research*, **4** (2023), 2300153.
[3] B. Aktekin, E. Kataev, L. M. Riegger, R. Garcia-Diez, Z. Chalkley, J. Becker, R. G. Wilks, A. Henss, Marcus Bär, and J. Janek, *ACS Energy Letters* (2024) 9 (7), 3492-3500

Improved Determination of Li^+ Transference Numbers in PEO-based Electrolytes

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Despite extensive research on PEO-based Li^+-conducting polymer electrolytes, reported transference numbers T_+ still vary considerably across the literature. This variability can largely be attributed to the application of different experimental methods, as some of them are relying on idealized assumptions, which are not fully satisfied in real electrolyte systems. The applied approaches range from electrochemical to thermodynamic and theoretical methods. For instance, electrochemical methods can be challenged by the accurate determination of interfacial resistances of electrodes, theoretical methods can be limited by the comparison with experimental data, particularly with respect to the chosen frame of reference. [1, 2]

To address these discrepancies, this study employs electrophoretic NMR (eNMR) to determine transference numbers of poly(ethyleneoxide) (PEO) in combination with lithium bis(trifluoro-methanesulfonyl)imide (LiTFSI), based on the electrophoretic mobilities μ_i of the investigated species i within the electrolyte. The application of electric field pulses during a pulsed-field gradient NMR (PFG-NMR) experiment induces a field-induced phase shift in the NMR signal, which can be used to calculate μ_i for the investigated species. Notably, eNMR enables a species-selective determination including the direction of drift of each species, which is often not possible for other commonly used methods. [1]

Following the initial publications on the use of eNMR for the investigation of charge transport in polymer electrolyte systems, the methodology has been further refined, yielding more precise results compared to earlier studies. [1] In particular the integration of reference electrodes into the experimental setup improves measurement precision and allows to compensate for unwanted flux contributions that may occur during eNMR experiments.[3] Additional methodological refinements include optimized samples and probe handling, as well as improved data processing and evaluation procedures.

We demonstrate that these recent advancements in eNMR methodology enable a more precise and reliable T_+ determination, yielding slightly larger mobilities for all electrolyte components, as well as higher T_+. In particular, the positive mobility of PEO chains is consistent with the expected migration of the polymer to maintain local volume conservation during ion migration. [4] Comparison to transference numbers obtained by other methods in different reference frames is made and allows to reconcile conflicting results.

References

[1] M. P. Rosenwinkel, M. Schönhoff, *J. Electrochem. Soc.*, **166** (2019), A1977-A1983.
[2] Y. Shao, H. Gudla, D. Brandell, C. Zhang, *J. Am. Chem. Soc.*, **144** (2022), 7583-7587.
[3] S. Buyting, M. Schönhoff, *Manuscript in preparation.*
[4] M. Lorenz, F. Kilchert, P. Nürnberg, M. Schammer, A. Latz, B. Horstmann, M. Schönhoff, *J. Phys. Chem. Lett*. **13** (2022), 8761-8767.

Anomalous Plasticization in Polymer-assisted Deep Supercooling of Li-salt

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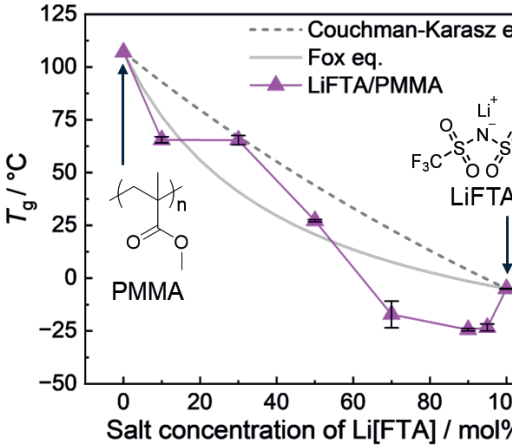
Lithium secondary batteries capable of fast charging require electrolytes with both high ionic conductivity and high Li^+ transference number [1]. Inorganic solid-state electrolytes used in all-solid-state batteries meet both of these requirements, but forming good electrolyte-electrode interfacial contact remains a significant challenge. In contrast, in liquid electrolytes, the interface chemistry can be well controlled, but the lithium transference number (t_{Li}) is usually low. The presence of solvent that can diffuse freely causes a concentration gradient of Li salt under anion-blocking conditions. Recent studies suggested lithium molten salts as a potential solution to achieve $t_{Li} \sim 1$ in a battery cell. However, the high melting point and crystallinity make most of these salts unsuitable for battery applications at ambient temperature. In response to this issue, stabilizing the supercooled liquid state is an effective way to extend the liquid temperature range of lithium molten salts. While in a different field, polymeric additives have been utilized in pharmaceutical formulations to suppress crystallization and stabilize amorphous states. Inspired by this concept, we previously reported that adding a small amount of polymer enables Li salts to remain liquid at room temperature [2]. The addition of 10 mol% of poly(methyl methacrylate) (PMMA), based on repeat units, to lithium (fluorosulfonyl)(trifluoromethanesulfonyl)amide (LiFTA or LiFTFSA) resulted in the formation of a viscous liquid at room temperature. In the resulting 90 mol% LiFTA/PMMA mixture, differential scanning calorimetry showed no melting peak attributable to LiFTA, and the liquid state was maintained for several months at ambient temperature. We refer to this system as a deeply supercooled Li salt (Li-DSS). Interestingly, this Li-DSS: 90mol% LiFTA/PMMA shows not only stable supercooled state and anomalous plasticization, which is a kind of deviation from empirical mixing relations in glass transition point: T_g. As shown in Figure, we compare experimental T_g value obtained from DSC measurement and empirical mixing relations, here using the Couchman-Karasz and Fox models [3, 4]. Generally reported in literature, ionic liquids/PMMA mixture follows empirical relations [5]. However, LiFTA/PMMA shows a pronounced T_g depression near ~90 mol% LiFTA composition.

In this presentation, we discuss this deviation of T_g from empirical law in Li-DSS, with particular attention to how polymer additives affect its structure and dynamics through applying the basis of Adam-Gibbs theory.



compared with empirical mixing relations

References
[1] K. M. Diederichsen, E. J. McShane and B. D. McCloskey, *ACS Energy Lett.*, 2017, **2**, 2563-2575
[2] T. Sudoh *et al*., *Adv. Energy Mater*., 2026, **16**, e05229
[3] P. R. Couchman and F.E. Karasz, *Macromol.*, 1978, **11**, 117-119
[4] T. G. Fox, *Bull. Am. Phys. Soc.*, 1956, **1**, 123
[5] M. A. B. H. Susan *et al*., *J. Am. Chem. Soc.*, 2005, **127**, 4976-4983

Design principles of polycations, polyanions and natural polymers in salt systems for next-generation solid polymer electrolytes

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Poly(ionic liquids) (PolyILs), also known as polymerized ionic liquids, have garnered increasing research interest as novel polymer electrolytes (PEs) for future lithium and sodium batteries. Cationic PolyILs-based polymer-in-salt systems have shown great promise in addressing the challenges of low ionic conductivity and low metal cation transference numbers related to conventional polymer electrolytes. We recently reported several ammonium cation-based PolyILs, poly(diallyldimethylammonium) bis(fluorosulfonyl)imide (PDADMA FSI). With the addition of a high concentration of LiFSI or NaFSI salt, the alkali metal ion transference number can reach between 0.5 and 0.65, with ionic conductivities up to 6.4×10^{-4} S cm^{-1} for the Li system at a 1:1.5 polyIL-to-salt molar ratio and nearly 10^{-3} S cm^{-1} for the Na system at 80°C at a 1:2 ratio.[1,2] Furthermore, when the salt concentration was extended to an extreme range at a 1:8 ratio, a further enhancement Li^+ transference number (~ 0.8) and improved electrochemical performance were reported,[3] indicating the impact of a salt-dominated environment. However, the role of polymer chemistry in extreme salt systems are not fully understand yet. In this research, we further extend our investigation to two additional types of polymer electrolytes: a neutral polymer (polyacrylonitrile, PAN) and an anionic polysalt (Poly(lithium sulfonyl(trifluoromethane sulfonyl)imide methacrylate, PMTFSILi), to understand their behaviors across a wide range of salt concentrations from 2:1 to 1:8 ratios. By combining molecular dynamics (MD) simulations and density functional theory (DFT) calculations, we systematically elucidate the role of polymer chemistry in developing polymer-in-salt electrolytes, thereby guiding the design of next-generation polymer electrolytes for lithium metal barriers.

References

1 Chen, F., Wang, X., Armand, M. & Forsyth, M. Cationic polymer-in-salt electrolytes for fast metal ion conduction and solid-state battery applications. *Nature Materials* **21**, 1175-1182 (2022).
2 Wang, X. *et al.* Poly(ionic liquid)s/Electrospun Nanofiber Composite Polymer Electrolytes for High Energy Density and Safe Li Metal Batteries. *ACS Applied Energy Materials* **2**, 6237-6245, doi:10.1021/acsaem.9b00765 (2019).
3 Kondou, S. *et al.* Poly(Ionic Liquid) Electrolytes at an Extreme Salt Concentration for Solid-State Batteries. *Journal of the American Chemical Society* **146**, 33169-33178, doi:10.1021/jacs.4c12616 (2024).

Wednesday
(June 3rd)

Wednesday, June 3rd

Session 1, 9:00−10:30, Chairperson: Cristina Iojoiu

PL3 9:00-9:40	**Louis Madsen, Virginia Tech, USA** New Understanding, Fabrication Methods, and Applications of Molecular Ionic Composite Electrolytes
K3 9:40-10:10	**Claudio Gerbaldi, Politecnico di Torino, Italy** Balancing ion transport, high-voltage stability and sustainable design in solvent-free UV-crosslinked solid polymer electrolytes
I10 10:10-10:30	**María Martinez, CIC Energigune, Spain** Single-ion conducting polymers for lithium–metal batteries: design, performance, and future strategies

Coffee Break (10:30−11:00)

Session 2, 11:00−12:55, Chairperson: Steven Greenbaum

K7 11:00-11:30	**Renaud Bouchet, University of Grenoble Alpes, France** Decoupling Bulk and Interfacial Contributions in Polymer–Ceramic Electrolytes
I11 11:30-11:50	**Guiomar Hernández, Uppsala University, Sweeden** Towards Polymers with High Voltage Stability
I12 11:50-12:10	**Laurent Rubatat, University of Pau and the Adour Region, France** In Situ Fabrication of Quasi-Solid Polymer Electrolytes via Photopolymerization-Induced Microphase Separation
O19 12:10-12:25	**Sajal Arwish, University of Münster, Germany** The Role of Li^+ ions in Polyzwitterionic Ionogels: Gelator or Mobile Charge Carrier?
O20 12:25-12:40	**Alejandro Herranz Berzosa, Polymat-EHU, Spain** Phosphonium Poly(Ionic liquid) Electrolytes for Fast Lithium-Ion Conduction
O21 12:40-12:55	**Samuel D.T. Power, Newcastle University, UK** Stability-Driven Design of Anion Exchange Polyelectrolytes: A New Versatile Protocol for Functionalisation

Lunch (13:00−14:30)

Social Activity # Guided visit in Donostia and Hondalea (15:30−)

New Understanding, Fabrication Methods, and Applications of Molecular Ionic Composite Electrolytes

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Molecular ionic composites (MICs) are high modulus solid electrolytes based on a rigid-rod charged polymer that can incorporate a variety of ionic liquids, salts, and/or other polar molecules. MICs represent a vast and flexible design space for tailoring solid materials to applications in energy storage and sustainability. At the same time, much of the fundamental phenomena in MICs require new observations, and I will present new insights and new questions. Our progress on MIC-based lithium-metal batteries includes designs that withstand hundreds of cycles with 90% capacity retention at up to 4.4 V. I will present new MIC compositions tailored to high energy cathode materials like NMC811, higher charge/discharge rates, and cycling with no dendrite formation as observed by SEM and X-ray tomography. I will also discuss details of electrolyte breakdown mechanisms via synchrotron X-ray methods in post-cycled battery cells. A new electrolyte fabrication method gives films with a factor of 2-10 higher modulus compared to materials from previous methods that have equivalent polymer content (e.g., > 2 GPa at only 13 wt% polymer). Finally, I will present MICs tailored for solid-state gas separations applications involving, for example, CO_2, N_2, H_2O, and CH_4.

Balancing ion transport, high-voltage stability and sustainable design in solvent-free UV-crosslinked solid polymer electrolytes

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Commercial alkali metal-ion secondary batteries currently rely on liquid electrolytes containing toxic and volatile organic carbonate solvents, which are prone to side reactions, oxidative decomposition, gas production, and combustion, particularly at high temperatures. Adoption of all-solid-state designs by polymer materials, ceramics, low-volatility additives, and hybrids thereof represent a promising solution towards next-gen, advanced alkali metal-ion secondary batteries; in addition to safety advantages, these offer higher energy density and longer runtime than traditional Li-ion cells [1,2]. However, low ionic conductivity, low cation transport properties and issues in sustainable manufacturing processes must be overcome for market deployment.

Herein, recent advancements in polymer-based electrolytes with high ionic mobility, designed for safe and efficient solid-state batteries, are highlighted. Polymer matrices include poly(ethylene oxide), poly(carbonates), poly(vinylidene fluoride), and various blends thereof. Sustainable techniques, including solvent-free extrusion and UV-induced photo-polymerization, are used for solid electrolyte production. Incorporating room-temperature ionic liquids, low-volatility additives, ceramics and even biosourced materials further improves electrochemical performance, achieving near-theoretical capacities at high C-rates with stable cycling at ambient conditions, also in combination with high-energy 4V class cathodes [3,4]. Single-ion conducting polyelectrolytes are also particularly promising as they feature immobilized anions and free-to-move alkali metal counterions, resulting in a lithium transference number close to unity and resistance towards dendrite growth [5]. In terms of sustainable development, the aim is at exploring alternative recycling pathways for materials holding potential for a second life within the energy storage domain as components in advanced solid-state batteries, particularly Na-based batteries [6].

Acknowledgements

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References

[1] S. Ferrari et al. *Adv. Energy Mater.,* **11** (2021) 2100785.
[2] H. Darjazi et al. *Adv. Mater.*, **36** (2023) 2313572.
[3] M. Gastaldi et al. *Mater. Today Energy*, **52** (2025) 101947.
[4] G. Lingua et al. *ACS Materials Lett.*, **6** (2024) 5429.
[5] H. Darjazi et al. *Adv. Funct. Mater.*, (2025) 2426075.
[6] R. Bajaj et al. *J. Power Sources*, **674** (2026) 239777.

Single-ion conducting polymers for lithium–metal batteries: design, performance, and future strategies

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Lithium metal batteries (LMBs) offer significant advantages over traditional lithium-ion batteries (LIBs), primarily due to the high specific capacity of LiM (3860 mAh·g−1), which is about ten times that of graphite. This allows LiM batteries to store more energy in a smaller, lighter package, making them ideal for applications like electric vehicles and portable electronics. Their low electrochemical potential (−3.04 V vs. SHE) also contributes to higher cell voltage and energy density. However, the use of LiM as anode presents significant safety risks, particularly when combined with highly flammable liquid electrolytes. Therefore, switching to safer electrolytes, such as solid-state ones, is required.[1]

Solid-state batteries (SSBs) offer a promising solution, overcoming many limitations of liquid-based LIBs. The use of solid electrolytes improves safety, stability, and energy density. Among these, polymer electrolytes (PEs) are gaining attention due to their lower cost, greater flexibility, ease of processing, and scalability.[2] Most PEs are dual-ion conductors (DICs), meaning both cations and anions can move freely in the electrolyte. Anions are often more mobile, leading to a low lithium-ion transference number (LTN), which creates concentration gradients during cycling. These gradients cause cell polarization, triggering undesirable side reactions and accelerating battery degradation. A promising solution is the design of single-ion conducting polymers (SICPs), where an anionic group is covalently attached to the polymer backbone. This innovation, emerging in academia in recent years, enables an LTN close to unity, preventing concentration gradients, polarization, and suppressing dendritic growth. The immobilization of anions within the polymer backbone restricts their movement, allowing lithium ions to distribute more evenly during plating and stripping a common issue in LMBs.

Inspired by these challenges our work is devoted to the design and development of new SICPs. In this talk, a general overview of our core research will be given with a special focus on following pillars: a) SICPs as ionically conducting and water processable binders for high voltage application;[3] b) enabling polymer electrolytes working at room temperatures using SICP-based gel polymer electrolytes;[4,5] and c) novel SICP chemistries towards perfluoroalkyl and polyfluoroalkyl substances free structures.

References

[1] M.A. Cabañero Martínez, et al., *Adv. Energy Mater.,* **12** (2022) 2201264.
[2] Song, Ziyu, et al., *Nature communications*, **14**, 1 (2023): 4884.
[3] Santiago, Alexander, et al. *Small*, **18**, 27 (2022) 2202027
[4] Aldalur, Itziar, et al., *Journal of Power Sources*, **624** (2024) 235539
[5] Santiago, Alexander, et al., *Journal of Power Sources*, **653** (2025) 237724

Decoupling Bulk and Interfacial Contributions in Polymer–Ceramic Electrolytes

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Liquid electrolytes have long been used in lithium-ion batteries due to their high ionic conductivity and excellent wettability of composite electrodes. However, their flammability raises significant safety concerns. Solid-state electrolytes are therefore considered a key component for next-generation batteries, offering improved safety and higher energy density, notably by enabling the use of lithium metal negative electrode.

Composite polymer–ceramic electrolytes have attracted considerable attention as they combine the mechanical compliance of solid polymer electrolytes with the thermal stability and fast ion transport of ceramic phases. While the addition of highly conductive ceramic fillers is expected to enhance overall conductivity, experimental observations remain widely dispersed and often contradictory, pointing to an incomplete understanding of the underlying transport mechanisms[1].

Here, we address this issue by identifying the key parameters governing ionic transport in such composites. We develop a simple geometrical model that explicitly incorporates interfacial charge transfer and validate it against well-defined model systems, including PEO/LATP composites with controlled particle sizes and alternative matrices such as PTMC. The model successfully captures the experimental trends and reveals that interfacial resistance plays a dominant role in determining effective conductivity, with a strong dependence on particle size.

To further probe the origin of this behavior, we systematically investigate charge transport across ceramic/electrolyte interfaces using trilayer configurations. Both liquid electrolytes (carbonates and ethers) and polymer systems (PEO, PTMC, SIPE) are examined, providing insight into the mechanisms governing interfacial transport across a wide range of chemistries.

[1] S. C. Sand, J. L. M. Rupp, B. Yildiz, Chem. Soc. Rev., 2025, 54, 178-200.A critical review on Li-ion transport, chemistry and structure of ceramic–polymer composite electrolytes for solid state batteries.

Towards Polymers with High Voltage Stability

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One of the premises of solid polymer electrolytes is the wide electrochemical stability window that would allow operating with lithium metal and high voltage cathodes. This property is often measured with linear sweep voltammetry (LSV) or cyclic voltammetry (CV), generally showing high voltage stability for many materials. However, the cell performance investigation with such materials is often done with LFP (3.5 V vs Li^+/Li), and nowadays more frequently with NMC family (up to 4.4 V vs. Li^+/Li), but it is still rare to demonstrate performance with a higher voltage cathode such as LNMO (up to 4.9 V vs. Li^+/Li).

We are investigating the electrochemical stability of solid polymer electrolytes with an additional technique, synthetic charge-discharge profile voltammetry (SCPV), that applies the real voltage profile of the active material of interest to a cell with SPE and a glassy carbon working electrode instead of the active material. While this technique is not able to reflect the behavior and stability towards the cathode active material, its time-dependent design allows assessment of polymer decomposition at the voltages and durations encountered in real cells.[1]

This presentation will show the oxidative stability study of different polymer families, comparing LSV with SCPV, as well as the cell performance with LNMO cathodes.

References

[1] G. Hernández, I. L. Johansson, A. Mathew, C. Sångeland, D. Brandell, J. Mindemark, *J. Electrochem. Soc.*, **168** (2021) 100523.

In Situ Fabrication of Quasi-Solid Polymer Electrolytes via Photopolymerization-Induced Microphase Separation

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In the context of a significant growth of the battery market, solid polyelectrolytes (SPE) are foreseen to replace the flammable liquid electrolytes. There are two main advantages for all solid-state batteries: bring a higher safety level (no solvent, prevent dendrites growth, etc.) and increase energy density (lithium metal battery). That will ultimately bring improved security and autonomy to battery for end-users. The challenge is then to associate SPE mechanical strength with sufficient ionic conductivity to maintain the performances. Unfortunately, in most cases improving one leads to the degradation of the other. Among polymers, block copolymers (BCP) can be an alternative to decouple those antagonist properties.[1,2] Indeed, in BCP, functional blocks are associated along the polymer chains that self-assembled into nano-domains exhibiting each of them specific properties. Most often BCPs are employed at thermodynamical equilibrium, since it gives stable and well-defined morphologies. More recently, out of equilibrium strategies were reported, such as Polymerization Induced Microphase Separation (PIMS).[3,4] This process, based on liquid formulations (composed of macro-initiator or pre-synthesized BCPs and lithium salt solubilized in monomer), leads after polymerization to kinetically arrested nano-morphologies. In the presentation, we will go through PIMS strategy examples and discuss the advantages and limitations.[5,6]

References:

[1] D. Devaux, D. Glé, T.N.T. Phan, D. Gigmes, E. Giroud, M. Deschamps, R. Denoyel, R. Bouchet, *Chemistry of Materials* **27** (13), 4682 (2015).
[2] K.I. Aniagbaoso, M. Krol, J. Ruokolainen, A. Bousquet, M. Save, L. Rubatat, *ACS Appl Mater Interfaces* **15** (12), 15998-16008 (2023).
[3] M.W. Schulze, L.D. McIntosh, M.A. Hillmyer, T.P. Lodge, *Nano Lett* **14** (1), 122 (2014).
[4] S.A. Chopade, J.G. Au, Z. Li, P.W. Schmidt, M.A. Hillmyer, and T.P. Lodge, *ACS Appl Mater Interfaces* **9** (17), 14561 (2017).
[5] K.I. Aniagbaoso, J. Bitenc, A. Bousquet, M. Save, L. Rubatat, *ACS Appl Mater Interfaces* **17** (2), 3876–3886 (2025).
[6] K.I. Aniagbaoso, M. Krol, J. Ruokolainen, A. Bousquet, M. Save, L. Rubatat, *Appl Mater Today* **47**, 15998 (2025).

The Role of Li^+ ions in Polyzwitterionic Ionogels: Gelator or Mobile Charge Carrier?

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The introduction of zwitterionic (ZI) polymers within lithium-containing, ionic liquid-based electrolytes can change the dynamics and local environment of Li^+ ions, in addition to enabling the formation of non-covalently cross-linked ionogels. In such materials, Li^+ ions can potentially act as free charge carriers or as network formers, and an optimization of Li^+-conducting ionogels requires an understanding of these roles. Therefore, we investigate Li^+ ion coordination and transport in ionogels consisting of the ionic liquid 1-butyl-1-methyl pyrrolidinium bis(trifluoromethyl sulfonamide) (BMP TFSI), LiTFSI, and the ZI polymer poly(2-(methacryloyloxyethyl phosphorylcholine)) (p(MPC)), obtained via *in situ* free radical polymerization. Our findings reveal that the change in ZI polymer content modifies the Li^+ ion coordination and thus affects the ionic transport properties of the electrolyte. Total ionic conductivity was observed to vary with p(MPC) content, reaching a maximum at 8 wt % p(MPC). Self-diffusion coefficients of the IL cation and anion obtained by Pulsed Field Gradient (PFG)-NMR were both observed to increase with p(MPC) content, while the self-diffusivity of Li^+ decreased. Simultaneously, a decrease in ^7Li peak intensity and a spin relaxation enhancement document immobilization of Li^+ ions by p(MPC). The results can be explained by a strong affinity of Li^+ to the negatively charged phosphate group of p(MPC), inducing reduced mobility. Raman spectroscopy confirms decreasing TFSI-Li^+ coordination with increasing p(MPC) content, supporting the concept of preferential Li^+ coordination by the polymer rather than by anions in solution. This shift in coordination has profound effects on the mechanical properties of the gel. Mechanical analysis reveals a drastic increase in elastic modulus, suggesting formation of noncovalent cross-links. From the experimental data the distribution of Li^+ on different sites, such as mobile Li^+, anion-coordinated Li^+, Li^+ coordinated to a single chain, or Li^+ forming noncovalent cross-links by dual chain coordination is analyzed [1]. This allows for an in-depth discussion of the role of Li^+, which is partly acting as a cross-linker and partly as a mobile charge carrier. The results allow us to identify guidelines for optimizing the balance between ionic conductivity and mechanical strength in copolymerized ZI polymer-supported ionogels.

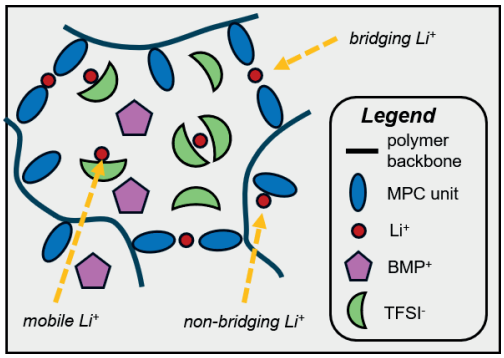

Figure 1. Illustration of different Li^+ species within an ionogel: bridging Li^+ acts as noncovalent cross-linker between ZI units, non-bridging Li^+ coordinated to a single ZI unit, and mobile Li^+ in solution.

References

[1] S. Arwish, M. K. Alsaedi, R. P. O'Hara, A. Asatekin, M. J. Panzer, M. Schönhoff, *J. Phys. Chem. B*, **130** (2026) 1384–1394.

Phosphonium Poly(Ionic liquid) Electrolytes for Fast Lithium-Ion Conduction

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Polymer electrolytes based on ionic liquids provide a safe solution for future solid-state high-energy-density batteries. In this work, we report a novel class of solid polymer electrolytes for fast lithium ion conduction based on phosphonium poly(ionic liquid)s (poly(IL)s). First, a new family of poly(diallyldimethylphosphonium) poly(IL)s were synthesized by cyclopolymerization of diallyldimethylphosphonium iodide following anion exchange with different sulfonamide-based anions. The characterization of the poly(IL)s confirmed the formation of a stable 5-member ring phosphonium cationic polymer backbone. Then, solid polymer electrolytes (SPEs) were prepared by doping the phosphonium sulfonamide poly(IL)s with different contents of lithium fluorosulfonamide salt (LiFSI). The nature of the ionic interactions and lithium transport of SPEs with different anion systems were deeply investigated by FTIR, DSC, ionic conductivity, solid-state ^{7}Li NMR and molecular modelling simulations. Polymer-in-salt phosphonium SPEs showed excellent properties with high ionic conductivities up to $1.5x10^{-3}$ S cm^{-1} at 80 ºC, high lithium transference numbers up to 0.7 and wide electrochemical stability window superior than state-of-art SPEs. Finally, we showed that phosphonium poly(IL) electrolytes allowed successful Li^{+} plating/stripping as solid electrolyte in lithium metal symmetrical cell, showing constant overvoltage limited to 0.06 V working at of 0.1 mA cm^{-2} current density at 60 ºC, and stable cycling without signs of short-circuiting during 200 cycles at 40 °C.

References

Forsyth, M.; Porcarelli, L.; Wang, X.; Goujon, N.; Mecerreyes, D. Innovative Electrolytes Based on Ionic Liquids and Polymers for Next-Generation Solid-State Batteries. *Acc. Chem. Res.* **2019**, *52* (3), 686–694. https://doi.org/10.1021/acs.accounts.8b00566.

Kondou, S.; Abdullah, M.; Popov, I.; Martins, M. L.; O'Dell, L. A.; Ueda, H.; Makhlooghiazad, F.; Nakanishi, A.; Sudoh, T.; Ueno, K.; Watanabe, M.; Howlett, P.; Zhang, H.; Armand, M.; Sokolov, A. P.; Forsyth, M.; Chen, F. Poly(Ionic Liquid) Electrolytes at an Extreme Salt Concentration for Solid-State Batteries. *J. Am. Chem. Soc.* **2024**, *146* (48), 33169–33178. https://doi.org/10.1021/jacs.4c12616.

Wang, X.; Chen, F.; Girard, G. M. A.; Zhu, H.; MacFarlane, D. R.; Mecerreyes, D.; Armand, M.; Howlett, P. C.; Forsyth, M. Poly(Ionic Liquid)s-in-Salt Electrolytes with Co-Coordination-Assisted Lithium-Ion Transport for Safe Batteries. *Joule* **2019**, *3* (11), 2687–2702. https://doi.org/https://doi.org/10.1016/j.joule.2019.07.008.

Stability-Driven Design of Anion Exchange Polyelectrolytes: A New Versatile Protocol for Functionalisation

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The composition of anion exchange polyelectrolytes (AEPs) used in anion exchange membrane water electrolysers (AEMWE) can be fine-tuned to control ion conductivity, ion exchange capacity, water uptake and swelling.[1] This requires careful design of the cationic head group, the polymer backbone, the tether and the co-monomer. However, balancing the long-term thermal stability and efficiency of AEPs at high pH remains a major challenge which needs to be addressed if AEMWE technology is to become commercially competitive with existing hydrogen production systems.[2-4]

Building upon previous studies, which demonstrated that the choice of cation-backbone tether is a major component in controlling alkaline stability, this work explores a new approach to the synthesis of model quaternary ammonium (QA) head groups and their incorporation into AEPs.[5] A series of QA head groups were prepared, and alkaline stability studies carried out over 84 days at 80 °C, using ^{1}H NMR spectroscopy to monitor degradation. Computational studies were used to investigate the observed degradation pathways and determine activation parameters, providing justification for the relative stability of the QA head groups.

Analysis concluded that increasing the number of 'spacer' units between the backbone of the polymer and the QA centre led to a dramatic improvement in alkaline stability, with the most stable QA head group displaying a half-life in excess of 50,000 h at 80 °C in 3M NaOD, which is competitive with the most stable head groups reported in the literature, under identical conditions. This QA structure was subsequently selected for the synthesis of AEPs to ascertain its suitability for use in an AEMWE. To incorporate the optimum QA head group into an AEP, a new post-polymerisation functionalisation protocol was developed, resulting in a dramatic decrease in reaction time from 72 h to <24 h, a reduction in reaction temperature from 50-70 °C to R.T., and a more straightforward purification procedure (DMSO to THF/MeOH reaction media) compared to the widely adopted S_N2 or Menshutkin reactions.[6]

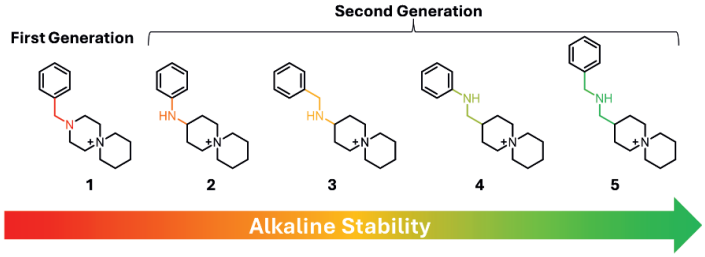

Figure 1: Structures of quaternary ammonium headgroups designed and explored during this study.

References

[1] X. Chu, Y. Shi, L. Liu, Y. Huang and N. Li, *J. Mater. Chem. A*, **2019**, 7, 7717-7727.
[2] M. G. Marino and K. D. Kreuer, *ChemSusChem*, **2015**, 8, 513-523.
[3] Y. M. Lee, *et al., Angew. Chem. Int. Ed.*, **2021**, 60, 19272-19280.
[4] G. W. Coates, *et al., J. Org. Chem.*, **2021**, 86, 254-264.
[5] S. Doherty *et. al., ACS Appl. Energy Mater.* **2025**, 8, 13, 9718-9730.
[6] N. Chen, Q. Jiang, F. Song, X. Hu, *ACS Energy Lett.,* **2023**, 8, 10, 4043–4051.

Thursday
(June 4th)

Thursday, June 4th

Session 1, 9:00−11:30, Chairperson: Daniel Brandell

PL4 9:00-9:40	**Vito di Noto, University of Padova, Italy** Electrical Response and Ion Conduction Mechanisms in Polymeric and Hybrid Electrolytes for Electrochemical Energy Conversion and Storage
K8 9:40-10:10	**Steven Holdcroft, Simon Fraser University, Canada** Towards Robust, Hydroxide Ion-Conducting Polymers
I13 10:10-10:30	**Faezeh Makhlooghi Azad, Deakin University, Australia** Structure–Transport Relationships in Anhydrous Protic Polymerized Ionic Liquid Membranes for Intermediate-Temperature PEM Fuel Cells

Coffee Break (10:30−11:00)

O22 11:30-11:15	**Klaus-Dieter Kreuer, Max-Planck-Institute for Solid State Research, Germany** Nano-phase-separated morphology of proton conducting PFSAs (Nafion): unique but not well-defined
O23 11:15-11:30	**Manabu Tanaka, Tokyo Metropolitan University, Japan** Poly(fluorene)-based Anion Exchange Membranes: Synthesis, Characterization, Molecular Dynamics Simulation, and Water Electrolysis Application

Session 2, 11:30−12:40, Chairperson: Jelena Popovic

K11 11:30-12:00	**Cristina Iojoiu, University of Grenoble Alpes, France** Design of Polymer Electrolytes with Enhanced Stability and Performance for Next Generation Batteries
I14 12:00-12:20	**Jonas Mindemark, Uppsala University, Sweden** PEO – baseline material or a special case?
I15 12:20-12:40	**Jennifer L. Schaefer, University of Notre Dame, USA** Investigation of ionomeric interlayers to mitigate the polysulfide shuttle effect in metal-sulfur batteries with polymer electrolytes

Sponsor Session, 12:40−13:00, Chairperson: Nicolas Goujon

S1 12:40-12:45	**Arkema, France**
S2 12:45-12:50	**Basquevolt, Spain**
S3 12:50-12:55	**Cidetec, Spain**
S4 12:55-13:00	**Biologic, France**

Lunch (13:00−15:00)

Board Meeting

Session 3, 15:00−17:30, Chairperson: Fangfang Chen

K10 15:00-15:30	**Guanglei Cui, Chinese Academy of Sciences, China** Intelligent Design and Film-Forming Process of Composite Solid Electrolytes
I16 15:30-15:50	**Dominic Bresser, Karlsruhe Institute of Technlogy, Germany** Ionic polymers as versatile cell components in lithium and sodium batteries
O24 15:50-16:05	**Hamideh Darjazi, Politecnico di Torino, Italy** Turning Waste into Value: Recycled PVB and Biosourced Polymers for Safe, Sustainable, high-performing Li/Na-based Batteries
O25 16:05-16:20	**Matthew Panzer, Tufts University, USA** An *in situ* Synthetic Approach to Nonvolatile, Ionic Liquid-Enhanced Polyzwitterion/Salt Electrolytes

O26 **Masahiro Yoshizawa-Fujita, Sophia University, Japan**

16:20-16:35 Enhanced Ionic Transport and Electrochemical Stability in Poly(trimethylene carbonate)/LiFSA Electrolytes Using Pyrrolidinium-Based OIPC

Coffee Break (16:35−17:00)

O27 **Yassine Eddahani, University of Grenoble Alpes, France**

17:00-17:15 Decorrelation Between Viscosity and Ion Transport in PMTFSI-based Single-Ion Conducting Polyelectrolytes

O28 **Martino Airoldi, Polymat-EHU, Spain**

17:15-17:30 *Operando* electro-rheology: unraveling decoupled transport-mechanical-morphological properties in nanostructured block copolymer electrolytes

Banquet # Hotel de Londres # (20:30−23:00)

Electrical Response and Ion Conduction Mechanisms in Polymeric and Hybrid Electrolytes for Electrochemical Energy Conversion and Storage

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Electrochemical energy conversion and storage (EECS) technologies are central to current strategies aimed at decarbonizing the energy sector, thanks to their high efficiency, scalability, and flexibility [1]. Among them, fuel cells, electrolyzers, and batteries - including redox flow batteries (RFBs) - play a complementary role across applications ranging from transportation to grid-scale energy storage. In all these systems, the electrolyte is a key component, as it enables ion transport while ensuring the separation of reactive species and the overall stability and efficiency of the device.

State-of-the-art electrolytes are typically based on polymeric materials such as perfluorinated ionomers, hydrocarbon-based membranes, and ion-conducting polymers designed for both proton- and anion-exchange systems, as well as for ion transport in batteries and RFBs. Despite their high conductivity and chemical stability, these materials still present important limitations, including reduced performance under low-humidity or high-temperature conditions, insufficient mechanical robustness, low conductivity (in batteries) and, particularly in the case of batteries and RFBs, undesired crossover of active species. These issues can severely impact efficiency, lifetime, and energy density.

To address these challenges, extensive research has been devoted to the development of advanced electrolytes, including hybrid organic-inorganic systems and composite materials. The incorporation of functional fillers or secondary phases allows for fine-tuning of transport properties, selectivity, and mechanical behavior. These approaches act by modulating the complex interplay between chemical composition, morphology, and thermal properties, ultimately influencing ion conduction pathways and interfacial phenomena at the nano- and mesoscale.

In this framework, Broadband Electrical Spectroscopy (BES) represents a powerful tool to probe the electrical response of these materials over a wide range of frequencies and temperatures, providing detailed insight into charge transport mechanisms and relaxation processes [2]. This contribution focuses on polymeric, hybrid, and composite electrolytes for applications in fuel cells, electrolyzers, and batteries, with particular emphasis on proton-, anion-, and other ion-conducting systems relevant to RFBs and solid-state devices. By correlating BES results with structural, morphological, and thermoanalytical properties, it is possible to elucidate the relationship between material design and ion conduction mechanisms. Such an understanding is crucial to guide the development of next-generation electrolytes with improved conductivity, selectivity, and stability, ultimately enabling enhanced performance and durability in a wide range of electrochemical energy technologies.

Acknowledgments
This work has been supported under: (a) the National Recovery and Resilience Plan (NRRP), funded by the EU - NextGenerationEU [Number: CNMS named MOST, Concession Decree No. 1033 of June 17, 2022, adopted by the Italian Ministry MUR, Spoke 14 “Hydrogen and New Fuels”]; (b) the project PERMANENT, Italian PNRR, M2C2, Investment Line 3.5; (c) the project AMBITION, Italian PNRR, M2C2, Investment Line 3.5; (d) the project DURALYS funded by the Italian Ministry MAECI; (e) MIUR-PRIN2022 project "Alkaline membrane–electrode assemblies for fuel cells (ALEF)" (Prot. 2022PYXHYL); and (f) the project SAMSARA (FISA-2023-00244) by the Italian Ministry MUR.

References
[1] B. Dunn, H. Kamath, J.M. Tarascon, *Science*, **334** (2011) 928.
[2] V. Di Noto, G.A. Giffin, K. Vezzù, M. Piga, S. Lavina, in *Solid State Proton Conductors: Properties and Applications in Fuel Cells*, **5** (2012) 109-183.

Towards Robust, Hydroxide Ion-Conducting Polymers

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In recent years, the study of cationic polymers possessing hydroxide ion counter ions have gained prominence. However, organic-based polymer cations are prone to nucleophilic attack by hydroxide ions, destroying the anion-exchange capacity and hydroxide ion conductivity. Numerous cationic head groups have been explored with a view to increasing the stability of cationic polymers in highly basic media.

Advancing hydroxide ion-conducting membranes requires an undertaking of rigorous systematic studies on model solid polymer electrolytes and representative materials of known and controllable macromolecular structure. This presentation will focus on recent progress in their preparation and on research trends incorporating these polymers into membranes, catalyst layers, and electrochemical energy storage and conversion devices.

Structure–Transport Relationships in Anhydrous Protic Polymerized Ionic Liquid Membranes for Intermediate-Temperature PEM Fuel Cells

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The global transition to a sustainable energy economy underscores the urgent need for safe, efficient, and high-performance hydrogen technologies. Proton exchange membrane fuel cells (PEMFCs), a cornerstone of hydrogen energy systems, offer compelling advantages including compactness and high energy-conversion efficiency.[1] However, broader deployment is constrained by membrane dehydration at elevated temperatures and continued reliance on perfluorinated membranes and ionomers, which raises environmental and sustainability concerns.

To address these limitations, we have developed new classes of anhydrous proton-conducting membranes and ionomers with tunable chemistries based on protic polymerized ionic liquids (protic poly(IL)s). Proton conductivity is enhanced through the incorporation of targeted additives, including protic ionic liquids (PILs) and protic zwitterions (PZIs), which promote ion dissociation and increase proton mobility. The resulting low-fluorine, free-standing membranes exhibit strong thermal and electrochemical stability, enabling operation at intermediate temperatures (80-120°C) where conventional water-mediated proton transport becomes inefficient. Spectroscopy analysis revealed that a synergistic poly(IL)/additive effect that supports multiple protonic environments and facilitates efficient transport. Atomic force microscopy further confirms membrane mechanical integrity, supporting their suitability for durable PEMFC operation.

Beyond membrane development, we have also implemented the protic poly(IL) as an ionomer in the cathode catalyst layer, demonstrating compatibility with commercial catalysts and improved proton conduction within the electrode. This dual functionality, membrane and ionomer, highlights the platform's versatility and its potential to improve membrane electrode assembly (MEA) integration and cathode performance. Collectively, these results provide a pathway toward PFAS-free membranes and ionomers for next-generation PEMFCs and contribute to broader efforts toward carbon neutrality.

References

[1] Zhang, H.; Shen, P. K. *Chem Soc Rev.* **6** (2012) 2382–2394
[2] George, A. …. Makhlooghiazad, F*, *ACS Applied Polymer Materials.* **7** (2025) 6641-6654

Nano-phase-separated morphology of proton conducting PFSAs (Nafion): unique but not well-defined

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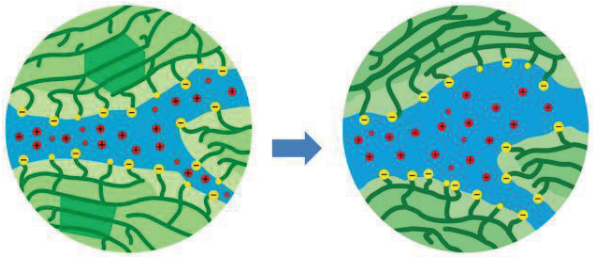
Perfluoro-sulfonic-acid (PFSA) membranes such as Nafion are used in a plethora of electrochemical applications such as fuel cells, electrolyzers and even flow batteries. Main reasons are their unique properties (combination of high ionic conductivity and chemical and mechanical stability) which, to a large extend, steam from PFSA's unique nano-phase-separated morphology. Accordingly, morphological investigations mostly based on small and wide-angle x-ray scattering (SAXS, WAXS) and simulations are an important part of the research on Nafion since the early days. Ever since Gierke et al. [1] presented their "inverse micelle model", many other models (lamellar, sandwich-like, flat ribbons, bundles, parallel-cylinders) tried to approximate PFSAs morphology. Interestingly, the model attracting most attention (parallel cylinder model [2]) was proven wrong [3], while the other models seem to capture certain real aspects of Nafion's morphology.

Recent data from SAXS as function of T and relative humidity (RH), SAXS, WAXS and dynamical mechanical analysis (DMA) after different pre-treatments clearly demonstrate that there is nothing like "the morphology" of Nafion, but complex morphologies reflecting the ionomer's history. Beyond phenomenological descriptions of "memory effects" in Nafion [4], we are now able to attribute the effects to **different competing processes driving the morphology**: While relaxation (structure forming) processes preferentially take place within the polymeric domain at high T and low RH, relaxations within the aqueous ionic domain dominate under wet and cold conditions. Both type of processes are mutually constrained through the presence of fixed-ionic groups which are part of the polymeric structure but also interacting with the aqueous ionic domain through electrostatic forces. This presentation identifies the different processes including the underlying interactions and provides clear rationales for understanding the quenched (frustrated) morphologies of PFSAs as a function of their history.



Example of morphological changes: strong swelling leads to a buckling of the aqueous ionic domain and a loss of crystallinity within the polymeric domain

References

[1] T. D. Gierke, G. E. Munn, F. C. Wilson, *J. Polym. Sci. B: Polym. Phys.* **19** (1981) 1687.
[2] K. Schmidt-Rohr , Q. Chen , *Nat. Mater.* **7** , (2008) 75
[3] K. D. Kreuer, G. Portale Advanced Functional Materials 23 (2013) 5390-5397.
[4] G. Alberti, R. Narducci, M. L. Di Vona; *Solid State Protonic Conductors: Properties and Application in Fuel Cells, eds. P. Knauth, M.L. Di Vona, John Wiley & Sons* (2012) 295-330.

Poly(fluorene)-based Anion Exchange Membranes: Synthesis, Characterization, Molecular Dynamics Simulation, and Water Electrolysis Application

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Anion exchange membrane water electrolysis (AEMWE) is an efficient method for producing green hydrogen using renewable energy, without requiring precious-metal catalysts or highly concentrated alkaline solutions [1]. However, conventional AEMWE systems exhibit lower performance than other water electrolysis systems due to the limited anion conductivity and insufficient alkaline stability of anion exchange membranes (AEMs) [2]. To address these issues, there is a strong demand for anion-conductive polymers that deliver high anion conductivity while maintaining stability under strongly alkaline conditions. In this study, poly(fluorene)-based anion conductive polymers, trimethylammonium-substituted polyfluorene (TMA-PF), were designed and synthesized (Figure 1). The electrolyte characteristics of the AEMs were evaluated, demonstrating higher anion conductivity and alkaline stability than a conventional AEM [3]. The anion transport analysis on TMA-PF was performed by all-atom molecular dynamics simulations [4]. The AEMWE performances using TMA-PF as AEMs and anion exchange ionomers (AEIs) will also be reported in the presentation.

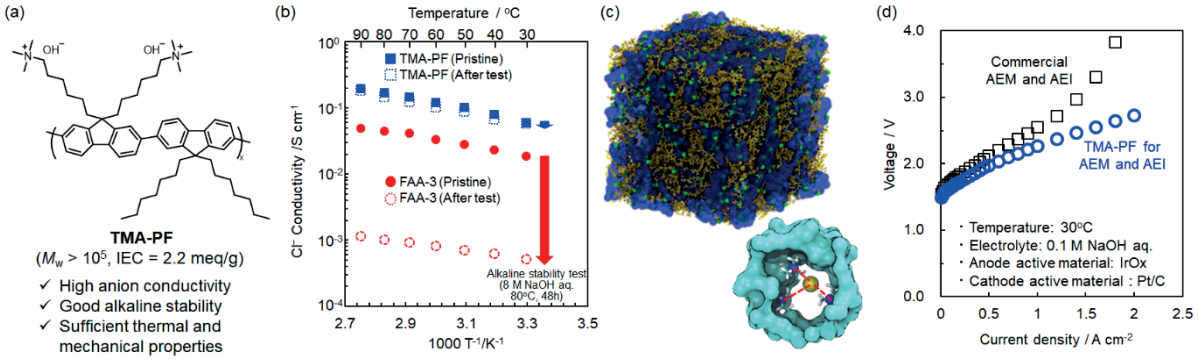

Figure 1. (a) A chemical structure and characteristics of TMA-PF, (b) Anion (Cl^-) conductivity of the TMA-PF membrane and commercial AEM (Fumasep FAA-3) before and after the alkaline stability tests (Immersion in 8 M NaOH aq. at 80°C for 48 hours), (c) All-atom molecular dynamics simulation of TMA-PF (a typical snapshot at λ (water content per anion exchange group) = 10), (d) Water electrolysis performance using TMA-PF as AEM and AEI (anion exchange ionomer) or commercial ones.

References

[1] C. Santoro, A. Lavacchi, P. Mustarelli, V. D. Noto, L. Elbaz, D. R. Dekel, F. Jaouen, *ChemSusChem* **2022**. 15, e202200027. [2] E. J. Park, P. Jannasch, K. Miyatake, C. Bae, K. Noonan, C. Fujimoto, S. Holdcroft, J. R. Varcoe, D. Henkensmeier, M. D. Guiver, Y. S. Kim, *Chem. Soc. Rev.* **2024**, 53, 5704. [3] Y. Nara, M. Tanaka, K. Nagasawa, Y. Kuroda, S. Mitsushima, H. Kawakami, *Polym. Adv. Technol.* **2022**, 33, 2863. [4] Y. Nara, K. Kasahara, K. Yagi, K. Kim, N. Matubayasi, H. Kawakami, M. Tanaka, *ACS Appl. Polym. Mater.* **2025**, 7, 20, 13971. [5] Y. Nara, M. Tomita, K. Nagasawa, Y. Kuroda, S. Mitsushima, H. Kawakami, M. Tanaka, *Adv. Eng. Mater.* **2024**, 2400252.

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Design of Polymer Electrolytes with Enhanced Stability and Performance for Next Generation Batteries

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Metallic lithium, with its exceptional specific capacity (3860 mAh g^{-1}) and the lowest redox potential (−3.04 V vs. SHE), is the ultimate negative electrode material for next-generation high-energy batteries. However, its practical deployment is severely hindered by persistent challenges, including lithium dendrite formation, inhomogeneous solid electrolyte interphase (SEI) formation, and poor Coulombic efficiency. Single-Ion Polymer Electrolytes (SIPEs) offer a promising solution by immobilizing anions within a polymer matrix, achieving a lithium transference number close to unity and effectively suppressing dendrite growth. This presentation reviews the different classes of SIPEs—both in dry and gelled states—highlighting their unique advantages and inherent drawbacks. Despite their promise, SIPEs still react with metallic lithium, forming thick, inhomogeneous SEI layers that increase interfacial resistance and compromise lithium reversibility. To address these issues, this study focuses on improving lithium reversibility and SEI formation in SIPEs through targeted doping with additives such as $LiNO_3$. Electrochemical tests demonstrate that doped SIPEs enable more homogeneous and smooth lithium deposition, lower interfacial resistance, and reduced polymer degradation.

Furthermore, to overcome the limitations of widely used poly(ethylene oxide) (PEO)-based polymer electrolytes—such as narrow electrochemical stability windows and poor compatibility with high-voltage cathodes—this work introduces a novel class of dry solid-state polymer electrolyte based on poly(butyl malonate) (CPBM). Compared to PEO-based systems, CPBM electrolytes deliver comparable ionic conductivity, higher lithium transference numbers (>0.6), and a wide electrochemical stability window (up to 4.7 V vs. Li/Li^+). Long-term cycling stability is demonstrated through lithium stripping/plating tests and full cells with LFP, LMFP, or NMC electrodes at high current densities, underscoring the potential of these materials for future battery applications. Additionally, aromatic gelled or plasticized SIPEs, along with their advantages, will be briefly introduced and compared to dried ones.

References

[1] H. P. K. Ngo, ACS Applied Energy Materials, **8 (5),** (2025), 2819–2827.
[2] Y. Shao, ACS Appl. Mater. Interfaces 18 (4), (2026), 6912–6925.
[3] Y. Shao, Journal of Materials Chemistry A, **12,** (2024), 30032 – 30040,
[4] H–D. Nguyen, Energy & Environmental Science, **11 (11),** 3298–3309, (2018).

PEO – baseline material or a special case?

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Ever since its involvement in the very inception of the field itself [1], the legacy of poly(ethylene oxide) – PEO – as a host material for solid polymer electrolytes lives on. From early studies that established the foundations of ion transport in polymers, through detailed investigations that uncovered the details of the ion transport mechanisms, to being a readily available reference material for just about every study on polymer electrolytes, a lot is owed to this one central material.

There are of course other polymers – that may or may not be more suitable as ion transport matrices [2] – and as investigations continue to uncover a wider variety of coordination chemistries and structural diversity, it is only fair to start asking the question: is PEO representative of polymer hosts in general or is it merely a special case at the edge of the spectrum? Some assumptions on polymer electrolyte properties generalized from PEO, such as having a low cation transference number, are simply not true for a wider range of polymers [3]. When looking at more subtle behaviors, such as how the ion coordination strength and transference number depend on molecular weight, PEO behaves completely opposite to some other polymer hosts [4]. While the basic modes of ion transport seem to be relatively similar [5], recent data also suggests different relaxation behavior for PEO when ions are added, as well as markedly different coordination behavior.

With all this in mind, it may not be so fair to simply generalize from PEO to other polymer materials. If so, that also begs the question: Do we need to find a new go-to polymer, at least for the purposes of creating broadly applicable models for ion–polymer interactions and ion transport mechanisms? And in what ways are PEO really that different from other materials? Or is what we have already good enough?

References

[1] D. E. Fenton, J. M. Parker, P. V. Wright, *Polymer,* **14** (1973) 589.
[2] J. Mindemark, M. J. Lacey, T. Bowden, D. Brandell, *Prog. Polym. Sci.,* **81** (2018) 114-143.
[3] M.P. Rosenwinkel, R. Andersson, J. Mindemark, M. Schönhoff, *J. Phys. Chem. C,* **124** (2020) 23588-23596
[4] R. Andersson, S. Emilsson, G. Hernández, M. Johansson, J. Mindemark, *ChemElectroChem,* **11** (2024) e202400415.
[5] H. Gudla, A. Hockmann, D. Brandell, J. Mindemark, *ACS Appl. Polym. Mater.,* **7** (2025) 4716-4724.

Investigation of ionomeric interlayers to mitigate the polysulfide shuttle effect in metal-sulfur batteries with polymer electrolytes

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The development of new rechargeable battery systems employing alternative chemistries is imperative to meet the increasing energy storage demands of emerging technologies. Metal-sulfur is one promising chemistry for moving beyond lithium-ion, due to the availability of S and the high theoretical energy density of S-based cathodes. However, metal-sulfur batteries with liquid electrolytes typically suffer capacity fade as a result of the polysulfide shuttle effect. Use of an inorganic solid electrolyte can alter the reaction pathway and mitigate the polysulfide movement away from the cathode, but this approach comes with its own challenges. Polymer electrolytes offer processibility and flexibility to accommodate volumetric changes upon battery cycling, however polysulfides are soluble in common polar polymer electrolytes so the polysulfide shuttle effect is not mitigated. Use of a polysulfide rejecting interlayer in conjunction with the polymer electrolyte is a possible solution.

Here, we report on our investigation of the polysulfide-rejection capability of crosslinked ionomeric interlayers for application in metal-sulfur batteries with solid polymer electrolytes. Ex-situ investigation of polysulfide uptake into and crossover through the interlayer is undertaken with both qualitative and quantitative approaches, including colorimetric analysis and elemental analysis.[1] The transport of the supporting salt is also tracked. The polysulfide rejection capability of ionomeric interlayers is found to be highly dependent on composition, including the type and density of covalently bound charges in the interlayer.

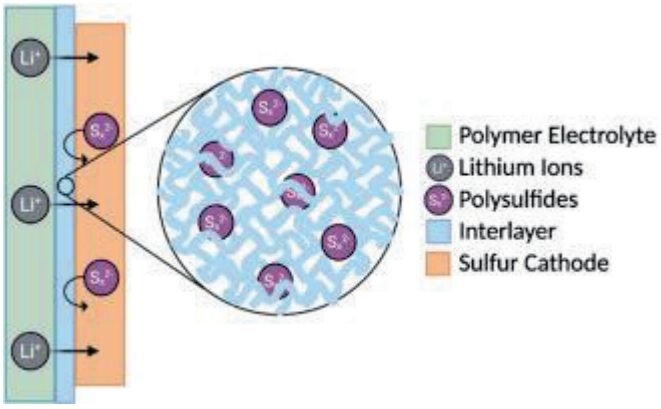

Figure. Schematic of polymer interlayer at the interface of the sulfur cathode and the bulk polymer electrolyte in a solid-state metal-sulfur polymer battery[1]

References

[1] P. Deshpande, B. A. Feczko, J. L. Schaefer, *Polymer,* **343** (2026) 129432.

Intelligent Design and Film-Forming Process of Composite Solid Electrolytes

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Sulfide-based composite solid electrolytes represent a pivotal direction for the future development of solid-state batteries, as they uniquely synergize the flexibility and superior interfacial processability of polymer electrolytes with the ultra-high ionic conductivity of sulfide materials. To accelerate the discovery and optimization of these materials, we have established a comprehensive AI-driven design framework. Guided by this framework, we successfully developed sulfide-based composite solid electrolytes exhibiting exceptional comprehensive performance. Building upon our material innovations, we systematically addressed the engineering challenges of film formation under harsh environmental stress. Specifically, we pioneered a progressive series of scalable manufacturing technologies, including the a new-paradigm melt dry process based on low-viscosity melts, a Hofmeister effect-mediated slurry system, and a benzene-free coating process utilizing multifunctional block binders. These systematic engineering transformations have successfully enabled the large-scale, high-quality mass production of composite electrolyte membranes.

Ultimately, the solid-state batteries fabricated using our scalable composite electrolytes were integrated into a deep-sea solid-state power system, achieving a successful technology demonstration in extreme operational environments. This work bridges the gap from intelligent material design to practical engineering implementation, providing a robust pathway to boost the industrialization of all-solid-state batteries.

References

[1] Cui, G. et al., *Adv. Mater.* **2023**, **35**: 2303312
[2] Cui, G. et al., *ACS Energy Lett.* **2023**, **8**: 3733
[3] Cui, G. et al., *ACS Nano* **2024**, **18**: 2475
[4] Cui, G. et al., *Adv. Energy Mater.* **2023**, **13**: 2302411
[5] Cui, G. et al., *Nat. Commun.* **2025**, **16**: 2979
[6] Cui, G. et al., *Adv. Mater.* **2024**, **36**: 2400737
[7] Cui, G. et al., *Angew. Chem. Int. Ed.* **2025**, **64**: e20241275
[8] Cui, G. et al., *Nano-Micro Lett.* **2025**, **17**: 250
[9] Cui, G. et al., *Adv. Mater.* **2025**, **37**: e04360
[10] Cui, G. et al., *Adv. Mater.* **2026**, **38**: e14210
[11] Cui, G. et al., *Angew. Chem. Int. Ed.*,**2025**, **64**: 202512771.
[12] Cui, G. et al., *Adv. Mater.* **2024**, **36**: 202401909.
[13] Cui, G. et al., *Nature Communications,* **2025**, **17**: 706.

Ionic polymers as versatile cell components in lithium and sodium batteries

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Polymer electrolytes based on polyethylene oxide (PEO) are the only commercial solid-state electrolyte system so far. However, the relatively low ionic conductivity at ambient temperatures, the remaining risk of lithium dendrite formation, and the limited electrochemical stability towards oxidation have hindered a widespread use so far.

Single-ion conducting polymer electrolytes (SIPEs) with an enhanced chemical design to withstand electrochemical oxidation, containing small molecules with a high dieletric constant to (partially) decouple the charge transport and the segmental dynamics of the polymer, provide an approach to overcome these limitations – not least as the single-ion conductivity can theoretically prevent a dendritic lithium deposition.

Herein, our progress in this field will be presented, with a specific focus on polysiloxane-based SIPEs. The talk will highlight the great versatility of this electrolyte class, enabling the realization of high-performance SIPEs for lithium and sodium-metal batteries, and the potential use as simple coating layer to stabilize the electrode │electrolyte interface.

Turning Waste into Value: Recycled PVB and Biosourced Polymers for Safe, Sustainable, high-performing Li/Na-based Batteries

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To lower the cost and reduce the environmental impact of modern batteries (Li-ion as well as post-Li batteries), efforts must be devoted at reducing the amount of inactive components in the cell, to substitute synthetic polymer binders/separators and organic solvents with low-cost and biosourced materials and to develop new eco-friendly manufacturing processes, even considering the reuse/revalorisation of recycled materials and components.

In this context, here an overview is offered of the recent developments in our laboratories on the development of innovative solutions including recycled or biosourced polymer binders/electrolytes towards the development of sustainable, safe, high-performing Li-/Na-based batteries. These include the possibility of repurposing in batteries the recycled polyvinyl butyral (PVB) from post-consume laminated glasses (from automotive and construction) that cannot be recycled for its primary purpose in glasses because of degraded optical properties or even bio-manufactured plastics (vitrimeric polyhydroxyalkanoates – PHA). Three strategies were pursued: using recycled PVB as binder in the electrode composition [1], transforming it into a membrane to be used as electrolyte separator [2], and utilizing it in advanced polymer electrolytes for solid-state batteries. In the case of PVB as binder, we investigated the electrochemical and structural properties of polymer blends of PVB with polyacrylic acid (PAA) and poly(vinylidene fluoride) (PVDF). We demonstrated its effective use in the development of various electrodes, including hard carbon (HC) anodes, and high-energy cathodes (NMC and NVP), which showed full capacities even at high C-rates and stable long-term operation at ambient temperature. Regarding solid-state batteries, the incorporation of either PVB or PHA into polyethylene oxide (PEO)-based solid polymer electrolytes was investigated to address both performance and sustainability challenges. This approach enabled the design of composite polymer electrolytes with improved mechanical strength, thermal stability, and electrochemical performance while promoting the use of recycled materials to reduce environmental impact. Overall, in all cases, preliminary results are highly encouraging and pave the way to the development of more sustainable separating electrolytes and binders from waste products for safe, low-cost energy storage devices.

To evaluate the manufacturability and technological relevance of the newly developed electrode/electrolytes, both lab-scale and industrial-format full cells were assembled and tested, showing stable long-term cycling at ambient temperature. Beyond confirming the robustness of these materials in realistic cell configurations, these results underscore a concrete pathway toward the industrial deployment of safer, lower-cost, and truly more sustainable Li/Na-based energy storage technologies.

Acknowledgements:
The SUNRISE project (https://sunrise-project.eu/) has received funding from the EU's Horizon 2020 research and innovation programme under GA 958243. The BIOMAPS project (https://www.biomaps-project.eu) has received funding from the EU's Horizon-RIA programme under GA 101177344.

References

[1] H. Darjazi, A. Piovano, G. Meligrana, G.A. Elia, C. Gerbaldi, *Adv. Funct. Mater.*, (2025) 2426075.
[2] S. Porporato, H. Darjazi, M. Gastaldi, A. Piovano, A. Perez, B. Yécora, A. Fina, G. Meligrana, G.A. Elia, C. Gerbaldi, *Adv. Sustainable Syst.*, (2024) 2400569.
[3] L. Balducci, H. Darjazi, A. Piovano, G.A. Elia, C. Gerbaldi, *J. Power Sources*, **662** (2026) 238704.

An *in situ* Synthetic Approach to Nonvolatile, Ionic Liquid-Enhanced Polyzwitterion/Salt Electrolytes

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The development of highly conductive polymer-based electrolytes having excellent safety characteristics is of great importance to the field of electrochemical energy storage. In this regard, the use of ionic liquids (ILs) as plasticizing components of polymer-based electrolytes is a promising avenue, given that aprotic ILs are nonvolatile and nonflammable liquid salts. It has been known for some time that equimolar combinations of certain zwitterionic (ZI) molecules with lithium bis(trifluoromethylsulfonyl)imide (LiTFSI) can form ion-dense liquid mixtures at room temperature.[1] If the ZI molecule is a reactive monomer, then the synthesis of a polyzwitterion/salt electrolyte through a polymerization reaction is possible. In our recent study,[2] we demonstrated the successful preparation of polyzwitterion-based electrolytes featuring LiTFSI and varying amounts of the aprotic IL 1-butyl-1-methylpyrrolidinium bis(trifluoromethylsulfonyl)imide (BMP TFSI) using a straightforward *in situ* UV-initiated photopolymerization method. The bioinspired ZI monomer 2-methacryloyloxyethyl phosphorylcholine (MPC) was combined with LiTFSI in an equimolar ratio in order to develop liquid phase precursors, to which between 30-90 wt.% BMP TFSI was added. Free radical photopolymerization of those precursor solutions allowed us to synthesize polyzwitterion-based electrolytes *in situ* with a variety of geometrical form factors; room temperature ionic conductivity values between 0.04 and 2 mS/cm were achieved, which increased monotonically with IL content. All electrolytes showed excellent thermal stability up to at least 250 °C prior to degradation via thermogravimetric analysis. Remarkably, the polyzwitterion-supported electrolyte containing approximately 20 wt.% solid poly(MPC) exhibited a room temperature ionic conductivity (~0.4 mS/cm) essentially equivalent to that of its polymer-free analogue (*i.e.* a LiTFSI/BMP TFSI solution containing the same number of moles of LiTFSI (1.6) per liter of IL). Thus, our synthetic approach elaborated here to realize IL-enhanced polyzwitterion electrolytes may prove beneficial for designing new manifestations of nonvolatile, nonflammable electrolytes that can improve the safety aspects of future lithium-ion batteries.

References

[1] A. Narita, SW. Shibayama, H. Ohno, *J. Mater. Chem.,* **16** (2006) 1475-1482.
[2] K. W. Wieck, M. J. Panzer, *ACS Appl. Energy Mater.,* **8** (2025) 11625-11632.

Enhanced Ionic Transport and Electrochemical Stability in Poly(trimethylene carbonate)/LiFSA Electrolytes Using Pyrrolidinium-Based OIPC

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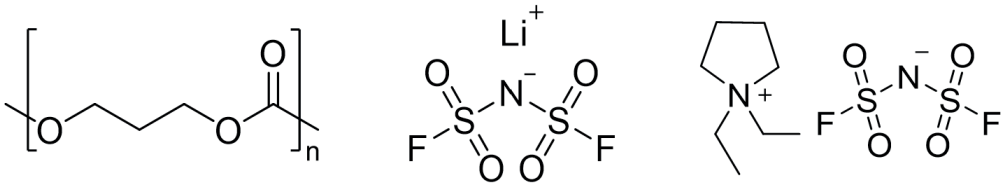
The rapid evolution of technology requires the development of next-generation lithium-based batteries that transcend the energy density and safety limitations of current liquid-based electrolyte systems. Solid-state lithium batteries (SSLBs) have emerged as a potential solution to mitigate the leakage risks, flammability, and dendrite growth typical of commercial lithium-ion batteries. This study explores the potential of poly(trimethylene carbonate) (PTMC) with lithium bis(fluorosulfonyl)amide (LiFSA) salt [1] and pyrrolidinium-based organic ionic plastic crystal (OIPC) *N,N*-diethylpyrrolidinium bis(fluorosulfonyl)amide [C_2epyr][FSA] [2] as an additive. The ternary composite PTMC/LiFSA/OIPC composites were named 22PT*x*, where *x* = wt% [C_2epyr][FSA] from 0-90%, and the mol % of LiFSA was maintained at 30%. Initially, the thermal properties, ionic conductivities, and transference numbers of binary PTMC/LiFSA composites were assessed. Subsequently, the influence of adding OIPC in various concentrations to the polymer matrix was investigated. The glass transition temperature (T_g) decreased monotonically with increasing OIPC concentration, while a melting endotherm was observed at high loadings. All ternary composites exhibited high thermal stability, required for battery applications. The ionic conductivity improved significantly in the ternary composite system, reaching a maximum value of 1.24×10^{-2} S cm^{-1} for 22PT80 at 100 °C. The lithium transference number (t_{Li+}) of 22PT5 achieved a value of 0.61, exceeding that of conventional electrolytes. Structural analysis confirmed a largely amorphous and homogeneous morphology at low OIPC loading. Fourier-transform infrared (FT-IR) and Raman spectroscopy further elucidated the coordination chemistry, indicating that Li^+ and FSA^- interactions are favored over the carbonyl group coordination, as OIPC was added gradually until a specific concentration threshold was reached. Finally, cyclic voltammetry (CV) and linear sweep voltammetry (LSV) demonstrated excellent electrochemical stability up to 5.0 V and improved Coulombic efficiency of up to 94.75% for the ternary composites compared to the binary composites. These characteristics suggest that PTMC/LiFSA/OIPC ternary composites are promising candidates for safe, high-voltage SSLBs.



PTMC LiFSA [C_2epyr][FSA]

Figure 1. Chemical structure of PTMC, LiFSA, and [C2epyr][FSA].

References

[1] Z. Li, J. Mindemark, D. Brandell, Y. Tominaga, *Polym. J.,* **51** (2019) 753-760.
[2] H. Yamada, Y. Miyachi, Y. Takeoka, M. Rikukawa, M. Yoshizawa-Fujita, *Electrochimica Acta*, **303** (2019) 293-298.

Decorrelation Between Viscosity and Ion Transport in PMTFSI-based Single-Ion Conducting Polyelectrolytes

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Lithium metal batteries (LMBs) are promising candidates for next-generation high-energy-density batteries due to the lowest redox potential (-3,05V vs ESH) and high theoretical capacity (3,8Ah/g) of lithium metal. However, their practical implementation has been hindered by non-uniform lithium plating/stripping and dendrite growth during cycling in conventional liquid electrolyte and separators, leading to serious safety concerns [1].

Polymer electrolytes (PEs) have emerged as safer alternatives to liquid electrolytes for LMBs. However, state-of-the-art PEs based on dual-ion conducting PEO suffer from low Li^+ mobility ($t_{Li^+} \approx 0.2$), low ionic conductivity at RT, and uneven growth of Li dendrite [2]. In contrast, Single-Ion Conducting Polyelectrolytes (SIPEs), in which anions are immobilized on the polymer backbone, enable predominantly Li^+ transport ($t_{Li^+} \approx 1$), thereby mitigating dendrite nucleation [3], improving interfacial stability, offering a pathway towards stable lithium metal batteries [4].

In this work, PMTFSI-based SIPEs were synthesized via radical polymerization in two different solvents (water and DMF), yielding homopolymers with identical chemical structures but significantly different molecular weights **(5.2×10^6 and 3.9×10^5 g·mol^{-1}, respectively)**, roughly a factor 10. While most literature studies focus on improving ionic conductivity by swelling polymers with solvent to form gels, we adopt the opposite approach[5].The SIPEs are dissolved in propylene carbonate (PC) to form a "**SIPE in solvent** " system , with polymer content ranging from 1 to 20wt%(ie 0.35 to 0.73 mol·L^{-1}).The transport mechanism in such systems remains poorly understood. In particular, the relationship between polymer chain architecture and lithium-ion mobility in polyelectrolyte solutions has not been systematically investigated to date.To address this , the structural and physicochemical properties of the SIPEs were characterized using rheology , DSC, TGA , Raman spectroscopy , electrochemical spectroscopy (EIS), and PFG-NMR.

The results show that increasing the molecular weight leads to a strong increase in bulk viscosity by a factor 10, consistent with chain overlap and entanglements [6,7]. Surprisingly, despite the large viscosity difference , ionic conductivity remains nearly unchanged and shows weak dependence on molecular weight. At higher polymer content , the systems exhibit highly viscous, gel-like behavior while maintaining liquid-like ionic conductivity (6.05×10^{-4} S·cm^{-1} at 20 °C). Walden analysis confirms a clear **decorrelation between macroscopic viscosity and ionic conductivity** in these electrolytes systems, indicating that ion transport is governed by local dynamics of the surrounding solvent molecule "micro-viscosity " rather than bulk viscosity. PFG-NMR and electrochemical measurements confirm that lithium ions are the dominant charge carriers, with a transference number close to unity (t_{Li+}=0.97). Finally, the electrochemical stability window and cycling performance in Li/Li symmetric and full cells were evaluated, demonstrating the relevance of these polyelectrolytes for lithium metal batteries.

References :

[1] Sanchez, A. J., & Dasgupta, N. P. (2024) Journal of the American Chemical Society, 146(7), 4282−4300.
[2] J. Mindemark, M. J. Lacey, T. Bowden, D. Brandell, Prog. Polym. Sci. 81(2018), 114–143.
[3] J.-N. Chazalviel, Phys. Rev. A, 42 (1990) 7355–7367.
[4] R. Bouchet et al., Nature Materials, 12 (2013) 452–457.
[5] P. Ghorbanzade, L. C. Loaiza, P. Johansson, RSC Adv., 12 (2022) 18164–18167.
[6] P. G. de Gennes, Scaling Concepts in Polymer Physics (Cornell University Press, Ithaca, 1976).
[7] A.V. Dobrynin, M. Rubinstein, Prog. Polym. Sci., 30 (2005) 1049–1118.

Operando electro-rheology: unraveling decoupled transport-mechanical-morphological properties in nanostructured block copolymer electrolytes

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Hierarchically self-assembling block copolymer electrolytes represent a promising class of solid-state ion-conducting materials for next-generation battery technologies.[1] However, the complex interplay among ion transport, polymer dynamics, and self-assembled nanostructure renders the electrochemical-mechanical coupling under galvanostatic conditions insufficiently understood. In this work, a LiTFSI-doped polystyrene-*block*-poly(ethylene oxide) diblock copolymer with a quasi-symmetric, lamellae-forming composition is investigated using combined *in-situ* characterization and a novel *operando* rheo-galvanostatic methodology. *In-situ* measurements enable direct correlation between polymer segmental dynamics and electrochemical transport properties, revealing how ionic translational motion is facilitated by polymer backbone dynamics.[2] The viscoelastic spectrum is analyzed within the framework of Rouse chain relaxation constrained by topological entanglements. Thus, implementing a model based on the Stokes-Einstein equation, strong agreement is established between nanoviscosity, representing segmental friction within copolymer chains, and electrochemistry derived diffusion properties.[3]

Leveraging the *in situ* observation, an *operando* experimental approach is proposed that allows simultaneous real-time monitoring of viscoelastic response, morphological evolution,[4] and ion transport under controlled dc polarization. Supported by transmission electron microscopy (TEM), the results provide a holistic view of how progressively increasing galvanostatic perturbations, associated with evolving salt concentration gradients,[5] induce structural transitions from lamellar to gyroid-like and ultimately hexagonal morphologies. Notably, this approach enables to capture thermodynamics of the copolymer enabling the construction of a phase diagram where current-induced nanomorphologies map directly onto the theoretical phase diagram.[6] In closing, this methodology enables the identification of a mirrored behavior between the rubbery plateau modulus and the interfacial resistance, suggesting a coupling between interfacial electrochemical processes and polymer chain dynamics. These observations support the proposal of a preliminary relationship linking interfacial electrochemical phenomena with polymer chain mobility, thereby highlighting the strong interplay between interfacial processes and electrolyte stability under varying current densities.

References

[1] R. C. Agrawal, G. P. Pandey. *J. Phys. D: Appl. Phys.,* 41 (2008) 223001.
[2] M. Airoldi, M. Fernández, I. Villaluenga. *Journal of Power Sources,* 667 (2026) 239236.
[3] J. Liu, D. Cao, L. Zhang. *The Journal of Physical Chemistry C,* 112 (2008) 6653-6661.
[4] M. B. Kossuth, D. C. Morse, F. S. Bates, *Journal of Rheology*, 43 (1999) 167–196.
[5] E. E. Abdo, N. P. Balsara, *European Polymer Journal*, 204 (2024) 0014-3057.
[6] F. S. Bates, G. H. Fredrickson, *Physics Today*, 52 (1999) 32–38.

Friday
(June 5th)

Friday, June 5th

Session 1, 9:00−10:30, Chairperson: Trang N.T. Phan

PL5 9:00-9:40	**Steven Greenbaum, Hunter College of the City University of New York, USA** Our Most Recent NMR Studies of Ion Transport in Polymer Electrolytes
K12 9:40-10:10	**Jelena Popovic, University of Stavanger, Norway** Understanding Battery Ionics and Electrodics: From Electrolytes to Interphases
I17 10:10-10:30	**Michal Piszcz, Basquevolt, Spain** Degradation study of model electrolytes in Li metal batteries and their impact on electrochemical performance

Coffee Break (10:30−11:00)

Session 2, 11:00−12:20, Chairperson: Jonas Mindemark

I18 11:00-11:20	**Andriy Kashva, Cidetec, Spain** Optimization of high-loading cathodes for practical solid-state batteries
O29 11:20-11:35	**Julia Amici, Politecnico di Torino, Italy** Unlocking Lithium Metal Batteries with Versatile Methacrylate-Based Composite Electrolytes
O30 11:35-11:50	**Melania Kozdra, University of Münster, Germany** Three ion pathways in polymer/ceramic composite electrolytes: a multiscale perspective
O31 11:50-12:05	**Victor Benito Olmos, Luxembourg Institute of Science and Technology, Luxembourg** Direct Visualization of Interfacial Lithium Diffusion in PEO–LLZTO Hybrid Electrolytes using High-Resolution FIB-SIMS
O32 12:05-12:20	**Hermann Pinson, University of Grenoble Alpes, France** Multi-segmented Copolymer-based Membranes for Aqueous Organic Redox Flow Battery

Concluding Remarks (12:20−12:30)

Our Most Recent NMR Studies of Ion Transport in Polymer Electrolytes

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Nuclear magnetic resonance (NMR) has been productively employed to investigate ion transport and solvation in Li ion battery liquid electrolytes and in solid polymer electrolytes based on poly(ethylene oxide) for many years. Future electrochemical power sources require new electrolytes to adapt to disruptive changes in battery chemistry, such as moving to Na ion, or to Li metal. Even when not targeting large scale commercial uses such as electric vehicles, niche applications (for example aerospace) requiring batteries that can operate under extreme conditions also demand new materials approaches. We highlight two recent collaborative activities on polymer electrolytes. In a multi-university (University of Maryland, Brown University, Yale University, City University of New York) effort led by Liangbing Hu , a novel Li^{+}-conducting solid electrolyte based on Cu^{2+}-doped cellulose nanofibers was developed and characterized by a suite of experimental and computational methods. [1,2] Here, we focus on NMR analysis of the ion transport mechanism in this novel material.
Next, we describe a recent collaboration with the Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA) and the University of Rome - Sapienza on ionic liquid based electrolytes with and without a polymer matrix designed for elevated temperature operation. In this presentation, we focus on the characterization of ion transport by NMR methods, in particular diffusometry and broadband relaxometry.

References

[1] Chunpeng Yang, Qisheng Wu, Weiqi Xie, Xin Zhang, Alexandra Brozena, Jin Zheng, Mounesha N. Garaga, Byung Hee Ko, Yimin Mao, Shuaiming He, Yue Gao, Pengbo Wang, Madhusudan Tyagi, Feng Jiao, Robert Briber, Paul Albertus, Chunsheng Wang, Steven Greenbaum, Yan-Yan Hu, Akira Isogai, Martin Winter, Kang Xu, Yue Qi, Liangbing Hu, *Nature* 598, (2021) 590–596. https://doi.org/10.1038/s41586-021-03885-6
[2] Keith Duncan, Qisheng Wu, Mounesha Garaga, Michael McKee, Zixuan Wang, Lin Xu, Xueying Zheng, Yue Qi, Steven Greenbaum, Liangbing Hu, Paul Albertus, *Journal of the Electrochemical Society*, **173**, (2026) 040501. https://doi.org/10.1149/1945-7111/ae4275

Understanding Battery Ionics and Electrodics: From Electrolytes to Interphases

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Ion motion is ubiquitous in battery technology, and the thermodynamics and kinetics of cation transport determine the suitability of specific materials as battery electrodes and electrolytes. Electrodics, focusing on porous electrode responses in electrochemical cells and charge-transfer kinetics, is likewise highly relevant. In this talk, I will discuss how these concepts enable the development and understanding of a wide range of battery materials, with focus on electrolytes. These include Li, Na, K, Mg, and Al metal anodes and their related interphases [1–9]; interphases in Si electrodes [10]; liquid, liquid/solid, semi-solid, and polymeric electrolytes [11–14], and potassium solid-state electrolytes [15,16]. The emphasis is on the development of electrochemical measurement methodologies—particularly electrochemical impedance spectroscopy and galvanostatic polarization—that enable elucidation of transport properties as well as time- and/or current-dependent battery ageing mechanisms, both at the material and full-cell level.

[1] Popovic, *Nat. Comm.*, 12, 6240 (2021), [2] Popovic, *J. Electrochem. Soc.*, 3, 169, 030510 (2022), [3] Lim et al., *ACS App. Mat. Int.*, 13 (43), 51767 (2021), [4] Popovic, *Ener. Tech.*, 9, 4, 2001056 (2021), [5] Lim et al., *ACS App. Mat. Int.*, 11, 5725 (2023), [6] Lim et al., *ACS App. Mat. Int.*, 14, 14, 16147 (2022), [7] Lim et al., *Appl. Phys. Lett.*, 122, 093902 (2023), [8] Löw et al., *ACS Mat. Lett.* 6, 11, 5120 (2024), [9] Grill et al., *Adv. Ener. Mat.*, *online* [9] Grill, Popovic-Neuber, *Commun. Chem.*, 7, 297 (2024), [10] Nojabee et al., *J. Mat. Chem.* A 7 (21), 13331 (2021), [11] Behjat et al. *Electrochim. Acta*, 518, 145792 (2025), [12] Guo et al., *J. Mat. Chem A* 11, 3427 (2022), [13] Grill, Popovic-Neuber, *Electrochim. Acta*, 518, 145792 (2025), [14] Grill, Popovic-Neuber, *Commun. Mat.*, 5 (1), 127 (2024), [15] Grill, Popovic-Neuber*, J. Ener. Chem.*, 111, 274 (2025), [16] Grill, Popovic*, ACS Ener. Lett.* 9, 9, 4465 (2024)

Degradation study of model electrolytes in Li metal batteries and their impact on electrochemical performance.

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The global transition toward renewable energy or electrification of automotive sector demands radical change in battery technology moving towards safe solutions with much higher energy density compared to current state of the art. Basquevolt is developing Li metal batteries that are outperforming existing solutions across multiple metrics. This has been achieved after intense investigation into established electrolyte systems to understand degradation mechanism of Li metal batteries. Understanding the failure modes in model electrolytes provides a critical foundation for further optimization.

In this work, chemically distinct model solvents classes are evaluated in different electrolytes. Their electrochemical performance, transport properties, full-cell behavior, and degradation are systematically compared. Correlation between transference number (t_+), Li cation reduction onset potential, and exchange current are correlated showing best theoretical candidate. However, transporting properties finally were compromised with their electrochemical stability and SEI resistivity change while cells were cycling. The demanding cycling conditions required for electric vehicle applications pose significant challenges for lithium metal anodes, particularly in liquid electrolyte systems. Furthermore, high cathode loadings can introduce transport limitations within porous electrode structures when low-conductivity electrolytes are used.

To address these challenges, Basquevolt has developed a novel strategy based on polymer coating of tailormade anolyte and at the same time developed catholyte with good electrochemical stability and great transporting properties. This approach enables a balanced cell configuration that stabilizes the lithium metal anode while maintaining high performance at the cathode. Figure 1 presents a qualitative comparison of lithium metal electrodes after the same charge throughput, demonstrating that polymer-coated lithium exhibits a more compact and uniform morphology compared to bare lithium electrode.

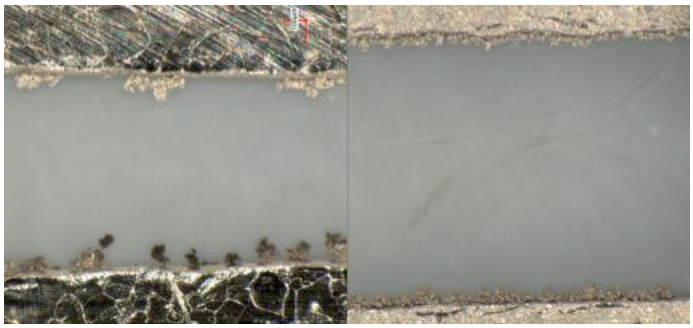

Figure 1. Qualitative comparison of Li metal electrode in symmetrical cells after same charge flow. Left photo – bare Li metal; right photo coated Li

Optimization of high-loading cathodes for practical solid-state batteries

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Achieving high energy density (>300 Wh/kg) in solid-state batteries (SSBs) for highly demanding applications such as road-transport and aviation requires solid-state (SS) composite cathodes able to deliver high areal capacity (high mass loading) under realistic conditions. Increased cathode mass loadings typically amplify issues such as high tortuosity impeding ionic transport and processing-induced heterogeneities. In SS electrode architecture, increasing electrode thickness and mass loading amplifies cracking/delamination issues and worsens wetting and interfacial contact. This often demands higher stack pressure and accelerates interface degradation, leading to poor C-rate capability and rapid capacity fading.

Herein, we report on the development of a SS composite cathode based on the "NMC811+C45+catholyte" system with high areal capacity of ~4 mAh/cm^2. We designed a high-performance SS cathode with polymeric catholyte by optimizing cathode composition (adapted from [1]), catholyte chemistry, and processing conditions to improve mechanical integrity, liquid retention, adhesion to current collector and ionic transport. Further, different post-processing protocols were used to tune the electrode density and boost volumetric energy density. In addition, the effect of the composite cathode formulation on the microstructure, "cathode/electrolyte" interfacial stability and electrochemical performance of "NMC811|SPE|Li-metal" cell at relevant charge-discharge rates has been investigated. In *Figure 1a*, the cross-sectional morphology of a cathode after cell assembly demonstrates a dense microstructure with areal capacity close to 4 mAh/cm^2. Optimized preparation procedure and post-processing protocols yielded composite cathodes (*CAM-74 wt%, C45-3 wt%, catholyte-23 wt%)*, which were tested in solid-state cells with "PVDF-HFP-plasticizer-Li salt" based on solid polymer electrolyte (SPE) system and Li-metal anode, achieving relevant discharge capacity of 3.9 mAh/cm^2, as seen in *Figure 1b*. In this work, we also observed that increasing the catholyte fraction in the cathode formulation can reduce the discharge capacity of solid-state cells. In addition to results summarized here, our contribution will highlight how optimizing electrode formulations enables practical, scalable solid-state cathodes with high energy and high mass loading suitable for practical SSBs.

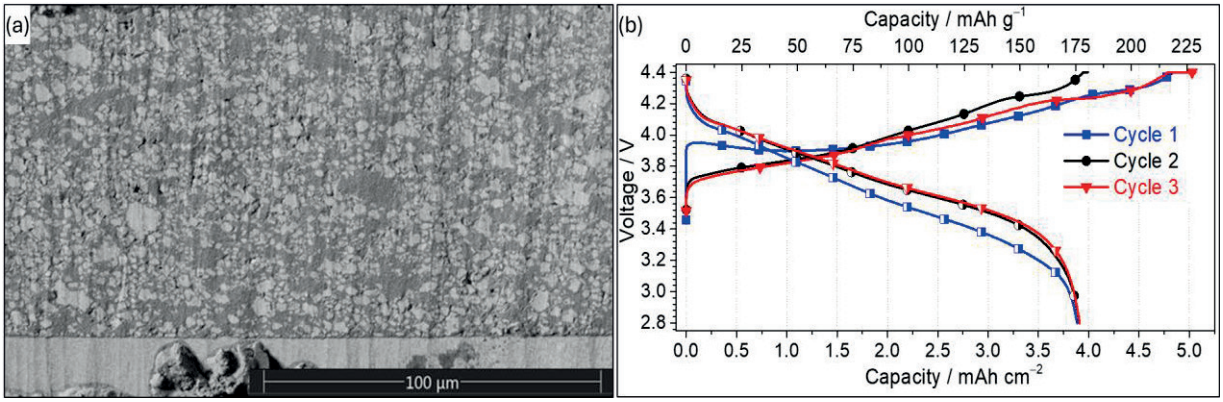

Figure 1. a) Cross-sectional image of high-loading cathode demonstrating dense microstructure, b) Cycling profiles of "NMC811|SPE| Li" solid state coin cell. Cycling conditions: 25 ºC, 0.05C/0.1C, 2.8–4.4 V. Cell configuration: cathode – 4.4 mAh/cm^2, density – 2.8 g/cc; SPE thickness – 238 µm; and anode – Li foil (50 µm).

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References

[1] A. Kvasha, T. Thieu, O. Garcia-Calvo, I. Combarro, M. Cobos, A. Gutiérrez, I. Urdampilleta, "Positive electrode for solid state batteries and solid-state batteries comprising the same," U.S. Patent 20240387816A1, Nov 11, 2024.

Unlocking Lithium Metal Batteries with Versatile Methacrylate-Based Composite Electrolytes

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To decrease carbon dioxide emissions, unprecedented efforts are being undertaken toward the development of efficient and inexpensive electric vehicles and stationary energy-storage systems. Lithium-ion batteries represent an efficient solution, that have transformed personal electronics and enabled the market introduction of electric vehicles. However, the ever-growing energy storage industry imposes great demands that current lithium-ion batteries could hardly satisfy. In this perspective, the use of metallic lithium as anode, would represent the "holy grail" of battery research thanks to its extremely high theoretical specific capacity (3860 mA h g^{-1}), the lowest redox potential (-3.040 V *vs*. the standard hydrogen electrode) and a low gravimetric density (0.534 g cm^{-3}).

However, metallic Li also presents many challenges derived primarily from dendrite formation upon cycling causing both safety issues and poor cycling performance. In addition, liquid electrolytes contain combustible organic solvents that can cause leakage and fire risks during overcharge or abuse conditions, especially in large-scale operation. Therefore, replacement of liquid electrolytes with a solid electrolyte has been recognized as a fundamental approach to effectively address the above problems.

Among the solid-state systems under study, polymer-based electrolytes represent a good compromise in term of room temperature ionic conductivity, thermal and electrochemical stability, and above all, interfacial contact. In lithium metal batteries, the preparation of methacrylate-based polymer matrix, in a one pot, solvent free polymerization, is an inexpensive and quick method to obtain versatile membranes. Meanwhile, eventual activation with small amounts of ionic liquids can allow to obtain composite electrolytes with outstanding room-temperature conductivities, while preserving the non-flammability of the system thus enhancing the safety. The simplicity of the formulation and the preparation method open the road to highly versatile electrolytes, adaptable in function of the final application. Additionally, the insertion of macromolecular groups in the matrix can introduce self-healing abilities (for example through dynamic hydrogen bonding), further improving the safety features of the cells.

Three ion pathways in polymer/ceramic composite electrolytes: a multiscale perspective

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The integration of ceramic ion conductors into polymer electrolytes offers a promising route toward composite solid electrolytes (CSEs) that combine high ionic conductivity with favorable mechanical properties. However, the mechanisms governing Li^+ transport across polymer–ceramic interfaces remain poorly understood. In this work, we investigate Li^+ transport in a polyethylene oxide (PEO) doped with lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) salt and $Li_7La_3Zr_2O_{12}$ (LLZO) composite system using a multiscale approach that combines atomistic molecular dynamics simulations with mesoscale modeling [1]. The focus is on three possible pathways, shown in Figure 1, and their impact on the Li^+ transport in the CSE is systematically evaluated. Here, phase-exchange barrier (PEB) rate for Li^+ transfer between polymer and ceramic phases is of crucial importance as shown in our previous work [2]. Building on this insight, we introduce the concept of a critical PEB rate required for effective utilization of fast ion transport through the ceramic phase, pathway 2 in Figure 1. An analytical expression for this threshold is derived, revealing its dependence on ceramic size, aspect ratio, intrinsic conductivity and interfacial width. For the LLZO/PEO:LiTFSI system, the estimated PEB rate lies close to this critical value, indicating that small changes in interfacial properties can determine whether conductivity enhancement, as compared to pure solid polymer electrolyte, is achieved. Furthermore, our results show that interphase-mediated transport, pathway 3 in Figure 1, does not contribute significantly to conductivity enhancement. These findings provide both physical insight into Li^+ transport mechanisms in CSEs and a framework for assessing when and how to achieve ionic conductivity enhancement.

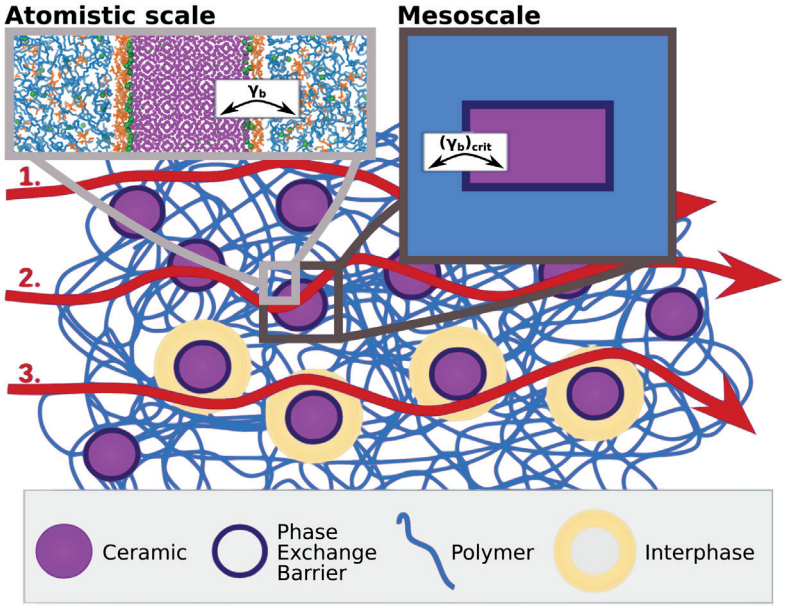

Figure 1. Schematic representation of a composite solid-state electrolyte. The ceramic-polymer interface studied in this work at the atomistic and mesoscale level are enlarged.

References

[1] M. Kozdra, L. Hölzer, D. Brandell, A. Heuer, *https://doi.org/10.26434/chemrxiv-2025-kxkz2*
[2] M. Kozdra, L. Hölzer, D. Brandell, A. Heuer, *Phys. Chem. Chem. Phys.*, **27** (2025), 24918-24931.

Direct Visualization of Interfacial Lithium Diffusion in PEO–LLZTO Hybrid Electrolytes using High-Resolution FIB-SIMS

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The development of advanced polymeric materials has attracted growing attention across a wide range of disciplines, with particular emphasis on battery technology. This is especially relevant for solid electrolyte systems such as polymer electrolytes and hybrid solid electrolytes (HSEs). To optimize both electrochemical and mechanical performance of next-generation polymer-based batteries, significant efforts have been devoted to the design of hybrid solid electrolytes (HSEs) with increasingly refined micro- to nanoscale architectures. However, the characterisation of these sophisticated materials poses a significant analytical challenge, as the required spatial resolution and sensitivity often exceed the capabilities of conventional techniques. Consequently, the development of advanced characterisation methods is essential to support further progress in polymer-based electrolyte research.

Secondary Ion Mass Spectrometry (SIMS) is a powerful surface analysis technique that enables the identification of characteristic secondary ions emitted upon irradiation of a sample with a high-energy primary ion beam. It offers outstanding sensitivity for all elements and their isotopes, including light species critical to battery research, such as lithium. Recently, advances in ion source technology—specifically in Focused Ion Beam (FIB) platforms—have delivered exceptionally bright ion beams, yielding enhanced spatial resolution for Secondary Electron (SE) imaging. By directly integrating a magnetic sector SIMS analyzer onto a FIB platform (FIB-SIMS), we unlock a highly attractive and powerful multimodal tool. This approach enables simultaneous high-resolution SE morphological imaging and true nanoscale chemical mapping, pushing lateral resolution down to the sub-20 nm regime[1][2].

In this study, we developed a comprehensive multimodal workflow for the characterisation of polymer-based HSEs (PEO(LiTFSI)–LLZO). Specifically, we exploited the unique capability of SIMS to distinguish light isotopes by utilizing a ^{6}Li-enriched electrode to investigate ion mobility. Simultaneous SE and high-resolution SIMS imaging were performed across the entire cross-section, previously polished via ultra-microtome. This method enabled the unambiguous identification of the distinct organic and inorganic phases, alongside high-resolution isotopic tracing of the Li across the HSE. Ultimately, this methodology enabled the direct determination of preferred diffusion pathways, local diffusion coefficients (D), and activation energies (E_a).

In this presentation, we will demonstrate the capabilities of this multimodal FIB-SIMS platform for advanced polymer characterization. Beyond conventional chemical and morphological mapping, we will showcase its unique power to spatially resolve lithium-ion transport pathways at the nanoscale, offering researchers a decisive new tool to engineer the next generation of solid-state batteries. This work was co-funded by the Luxembourg National Research Fund (FNR) through the grant INTER/DFG/22/16558792/MINABATT and the German Research Foundation (DFG) HE8586/MINABATT.

References:

[1] J. N. Audinot, et al., *Reports Prog. Phys*. 84, (2021) p.105901.
[2] Benito Olmos, V., et al., *Surfaces and Interfaces*, 84, (2026) p.108606.

Multi-segmented Copolymer-based Membranes for Aqueous Organic Redox Flow Battery

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Aqueous organic redox flow batteries (AORFBs) are promising candidates for large-scale electricity storage due to their safety, environmental friendliness, economic feasibility, and high storage capacity. In these systems, the membrane plays a critical role: it facilitates ion transport, separates the anolyte and catholyte, and prevents electrical short circuits between the electrodes. To maintain the system's charge neutrality, the membrane must allow the circulation of counterions that balance the charge during oxidation and reduction reactions. Additionally, it must exhibit selectivity toward the charged active species present in the aqueous solution. Consequently, ideal membranes should combine high ionic conductivity and selectivity with robust chemical and mechanical stability. The non-fluorinated membranes have poor stability when exposed to RFB electrolytes due, generally, to the presence of ethereal oxygen atoms [1]. High stability ether-free membranes were reported [2] but the synthesis usually requires costly metallic catalysts. Recently, the superacid-catalyzed polymerization reaction (Friedel-Crafts-type polyhydroxyalkylation reaction), characterized by a short reaction time without the requirement of heating and expensive catalysts was achieved [3]. However, copolymers synthesized via a Friedel-Crafts reaction typically yield random copolymers. In this study, we propose an alternative synthetic route to produce multi-segmented block copolymers, allowing for a direct comparison between the two architectures [4], leading the way to polymers with promising properties. These copolymers are subsequently functionalized through thioacetalization reactions followed by oxidation [5], introducing sulfonated ionic groups (SO_3^-). Multi-segmented copolymers with varying lengths of hydrophilic and hydrophobic segments—and thus different ion exchange capacities (IECs)—were investigated as membranes both ex situ and in AORFBs. The multi-segmented block copolymer membranes exhibit well-phase-separated nanostructures at two distinct length scales, as demonstrated by Scanning Electron Microscopy (SEM) and Small-Angle X-ray Scattering (SAXS). The SAXS profiles exhibit two distinct peaks: the first corresponds to the correlation distance between hydrophobic regions (d_{hyd} = 15.3 nm), while the second corresponds to the separation between ionic groups and the polymer backbones (d_{ion} = 2.3 nm). Compared to statistical copolymers with same IEC (1.7 mmol.g^{-1}) the multi-segmented copolymers achieve significantly higher sodium conductivity (7.5 mS·cm^{-1} vs 3.5 mS·cm^{-1}) with only slightly higher water uptake (35% vs 30%). In AORFBs, this results in a reduced area-specific resistance (ASR) (2.6 Ω·cm² vs 5.8 Ω·cm²), enabling operation at higher current densities and thereby contributing to improved system power density.

Additionally, the influence of membrane casting conditions (e.g., solvent type, solvent evaporation time) on membrane morphology and functional properties was thoroughly investigated. For instance, membranes cast from dimethylacetamide (DMAc) exhibit a d_{hyd} = 19.0 nm with a much narrower peak (Full Width at Half Maximum (FWHM= 0.104 nm), whereas those cast from dimethyl sulfoxide (DMSO) show a d_{hyd} =15.3 nm with a broader peak (FWHM= 0.149 nm**)**. While these morphological differences do not significantly affect ionic conductivity, they do influence selectivity: no crossover of redox molecules was observed for the DMAc-cast membrane, whereas a slight crossover was detected for the DMSO-cast membrane (diffusion coefficient$_{210h}$ = 7.4 m²·s^{-1}).

References

[1] M. Adamski, et al., *Materials Advances,* **2** (2021) 4966-5005.
[2] J. Miyake, et al., *Science Advances,* **3** (2017).
[3] L. I. Olvera, et al., *Macromolecules,* **46** (2013) 7245-56.
[4] X. Li, et al., *Macromolecules,* **55** (2022) 10607-17.
[5] M. K. Pagels, et al., *ACS Macro Lett.,* **9** (2020) 1489-93.

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