

INSIGHT SERIES

FROM EXPERTISE TO IMPACT



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Single-Ion Polymer Electrolytes for High-Voltage Solid State Batteries

Abstract

Conventional liquid-electrolyte lithium-ion batteries face safety concerns from leakage and flammability, together with limited stability at high operating voltages. Solid-state batteries (SSBs) offer a promising alternative, with the potential to increase energy density without increasing battery size or weight. However, practical implementation remains challenging because long cycle life and rapid identification of degradation mechanisms are still unresolved, particularly at high voltages that place severe demands on the solid polymer electrolyte. Single-ion-conducting polymer electrolytes (SiCPs) offer a potential solution by immobilizing negatively charged anions within the polymer framework, leaving lithium ions as the main mobile charge carriers. This can reduce concentration polarization, support more uniform lithium deposition, and improve compatibility with lithium-metal SSBs. Their key challenge is to combine fast room-temperature ion transport with mechanical strength, high-voltage stability, and scalable manufacturing. The EU-funded HyLiST project addresses these requirements by developing hybrid SiCPs for solid-state cells using lithium metal and a cobalt-free, high-voltage LNMO cathode.

Why solid-state batteries need safer electrolytes

In the global electrification race, lithium-ion batteries (LIBs) dominate portable electronics and electric vehicles space due to their high energy density and long cycle life. However, the **flammability, risk of leakage and thermal runaway** for conventional liquid electrolytes; along with its instability against lithium-metal anode and high-voltage positive cathodes constrain further improvements in **safety and energy density**.

Solid-state batteries (SSBs) offer a promising alternative by replacing liquid electrolytes with solid state electrolytes (SE), potentially **improving safety** while enabling lithium-metal anodes for cells with higher energy density. Among various solid electrolyte classes, **solid polymer electrolytes (SPE)** have several advantages such as being **lightweight, offering good mechanical stability, providing improved thermal stability** compared to liquid electrolytes and enabling scalable processing into thin films. Conventional SPEs have dual-ion conducting nature where both positively charged lithium (Li) ions and negatively charged counter-ions can move. Their movement creates uneven ion concentrations during charging, known as concentration polarization. At high currents, this imbalance can reduce performance and contribute to uneven lithium deposition [1,2]. During repeated charging cycles lithium can be deposited unevenly due to concentration polarization, producing needle-like or mossy dendrites that encourage unwanted reactions and internal short circuits.

Single-ion polymers: directing lithium transport

Single-ion conducting polymer electrolytes (SiCPs) address this limitation by **immobilizing the anion to the polymer framework, making lithium ions the primary mobile charge carriers**. Li ions remain free to travel through the polymer electrolyte during cell cycling, as seen in Figure 1. As a result, the proportion of ionic current carried by Li ions, which is specified as Li -ion transference number, can approach to the max value of "1". This signifies that all the current transferred through the polymer electrolyte will be via the Li^+ ions. This helps minimize concentration polarization and can promote more uniform Li transport [1–3]. Single ion conducting polymers can therefore help to enable practical application of Li metal anodes in rechargeable batteries. [2,4]

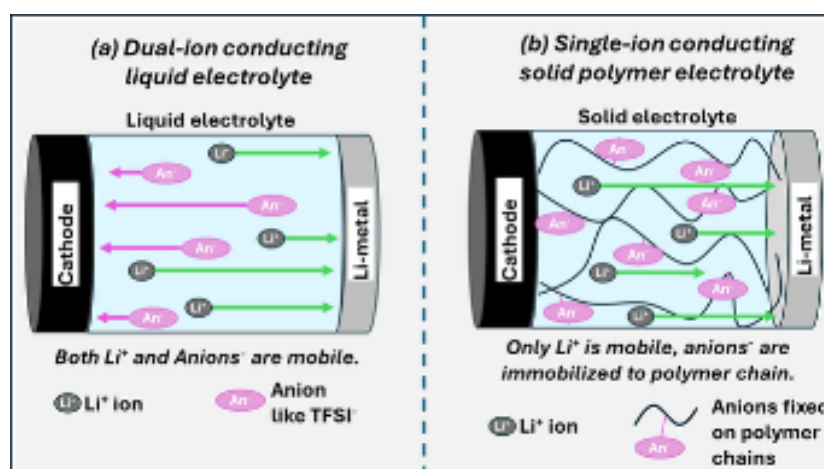


Figure 1 Schematic illustration of (a) DUAL-ion conductors and (b) single-ion conductors.

Balancing conductivity, stability and manufacturability

Current research in **SiCPs explore block co-polymers, cross-linked networks, plasticizers and ceramic fillers**. Early block copolymer based single-ion polymer electrolytes demonstrated Li transference numbers close to unity and stability extending towards 5 V [3]. Other polymer electrolyte systems have achieved ionic conductivities close to 10^{-3} S cm⁻¹ at room temperature and oxidative electrochemical stability limit close to 4.7 V [4,5]. Therefore, a **practical SiCP must conduct Li-ions quickly**, remain **mechanically strong**, being **stable against Li-metal** and **resist oxidation** versus cathode at high charge cut-off voltages. These operational requirements often compete against each other.

A **polymer electrolyte film which has a soft nature may move Li-ions faster** but provide less mechanical support, on the other hand a **rigid polymer electrolyte can be mechanically stronger yet slower to conduct Li-ions** [1,6]. Another facet of mechanical stability of polymer electrolyte is the interface stability where even a highly conductive electrolyte can fail if electrolyte-electrode contact is poorly developed at both electrodes. At Li-metal side, the electrolyte contact must support uniform plating and stripping. At high-voltage cathode side, the electrolyte must resist breakdown while allowing rapid lithium movement. This matters in solid-state batteries because a solid cannot flow into every pore or crack as a liquid does [5,6]. The **upscaled processing of solid electrolytes is another hurdle**. A laboratory-scale polymer electrolyte must be eventually transformed into a thin, uniform film made over long lengths and integrated into practical mono- and multi-layer pouch cells.

Why high voltage raises the stakes?

Another important aspect of improving the energy density without being volumetrically larger or gravimetrically heavier, is to increase the operating voltage of batteries. This makes **high-voltage cathodes especially attractive for next-generation batteries**. However, higher charge cut-off voltages place greater stress on the electrolytes, accelerating unwanted reactions and degradation in conventional liquid systems. **Solid-state batteries offer a promising route** to overcome these limitations by using non-volatile solid electrolytes that can potentially **provide improved safety and wider electrochemical stability window**.

HyLiST's role: from laboratory chemistry to scalable cells

The above-mentioned requirements and challenges for solid electrolyte development are central to EU-funded **HyLiST project - Hybrid Lithium Metal-based Scalable Solid State Battery Manufacturing**. HyLiST is developing Gen4c solid-state cells combining a lithium-metal anode, a cobalt-free lithium nickel manganese oxide (LiNi_{0.5}Mn_{1.5}O₄ - LNMO) based composite cathode and a hybrid single-ion conducting polymer electrolyte (HSiPE) [7,8].

HyLiST targets **room-temperature ionic conductivity of at least 0.5 mS cm^{-1} and a lithium-ion transference number close to "1"** for HSiPE electrolytes. In HyLiST solid-state cells, to achieve competitive energy density values requires LNMO cathodes that remain stable above 4.7 V, so HSiPE electrolyte stability at high voltage is a key requirement [7,9]. Importantly, the project also focuses on manufacturing, dry processing is being developed for the electrolyte, and the cell design is intended to be transferable to existing European production infrastructure [7,8,9]. The project shows the direction for HSiPE research, heading beyond laboratory processing and characterization towards a scaled-up HSiPE electrolyte that works with a high-voltage cathode and lithium metal, which survives repeated cycling and can be manufactured at pilot-plant scale.

Conclusion: a pathway to safer high-energy batteries

To summarize, **single-ion polymer electrolytes immobilize anions within the polymer matrix, allowing lithium ions to carry most of the ionic current.** This can reduce concentration polarization and promote more uniform conditions in lithium-metal batteries. Their operational potential is particularly relevant for high-voltage solid-state batteries, where ionic conductivity, electrochemical stability, mechanical properties, interfaces and manufacturability must be balanced. HyLiST project brings these elements together by combining polymer electrolyte formulation design with high-voltage cathodes, lithium metal and scalable processing. Successful integration could make single-ion polymer electrolytes a key component enabling safer, lighter and higher-energy batteries for electric vehicles, aviation and other demanding applications.

About the author

Kevin Vattappara is a researcher and project manager at the Materials for Energy Unit of CIDETEC Energy Storage, Spain. He finished his joint Ph.D. in 2026 at CIDETEC Energy Storage, Forschungszentrum Jülich and University of Duisburg-Essen. He specialized in "Ceramic-rich composite separators for lithium metal solid-state batteries". His research spans solid-state electrolyte systems, while his current work is dedicated to the research and development of advanced cell components and scalable processes for high-energy solid-state batteries.

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