

Single ion conducting polymer electrolyte for safe and high-performance batteries

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Lithium-based batteries are a key technology for successfully transitioning from depleting fossil fuels to renewable energy sources, particularly in meeting the demands of a highly mobile society. Nevertheless, the commonly employed liquid organic electrolytes are highly flammable and release toxic degradation products.^[1] Solid electrolytes are a promising alternative, since they offer an improved safety and potentially suppress dendritic lithium deposition.^[2] 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 ($>10^{-4}$ S.cm⁻¹ at 25°C), high mechanical strength, and an electrochemical stability window of at least 4.5 V. Conventional polymer electrolytes based on poly(ethylene oxide) (PEO) consist of a lithium salt dissolved in the polymer. The need for a conducting salt, however, results in transference numbers (t_+) far below 0.5. This leads to charge concentration gradient and reversed cell polarization issues, which worsen the long-term cycling stability.^[3] SPEs with $t_+ = 1$, also known as 'single-ion polymer electrolytes' (SIPEs), might overcome these issues. The simplest approach to achieve single-ion conductivity is to covalently tether the anion to the polymer backbone (so-called 'ionomers'), thus ensuring that the only mobile species is the cation. Nevertheless, the conductivity of these systems is typically very low (10^{-8} to 10^{-7} S.cm⁻¹ at 25°C) and/or the mechanical strengths very poor, therefore their employment remains still unfeasible. 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). Therefore, to bring together these two antagonistic properties, it is essential to design polymer electrolytes with well adapted structure and/or nanostructured morphology showing well-defined and co-continuous ionic and hydrophobic domains. In this presentation we discuss on different strategies for the synthesis of SIPE and their characterization in Li and Ca form. The materials properties are nicely tailorable with regard to the ionomer structure particularly backbone and anion side chain structures. We have demonstrated the fluorinated anion or partially fluorinated backbone have a key role in the stabilization of the interface: SIPE/electrode.^[4-7] Remarkable and original results were obtained by incorporating the SIPE in lithium metal batteries.

References

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