Vitrimeric poly(ionic liquid) electrolyte for Li-ion batteries

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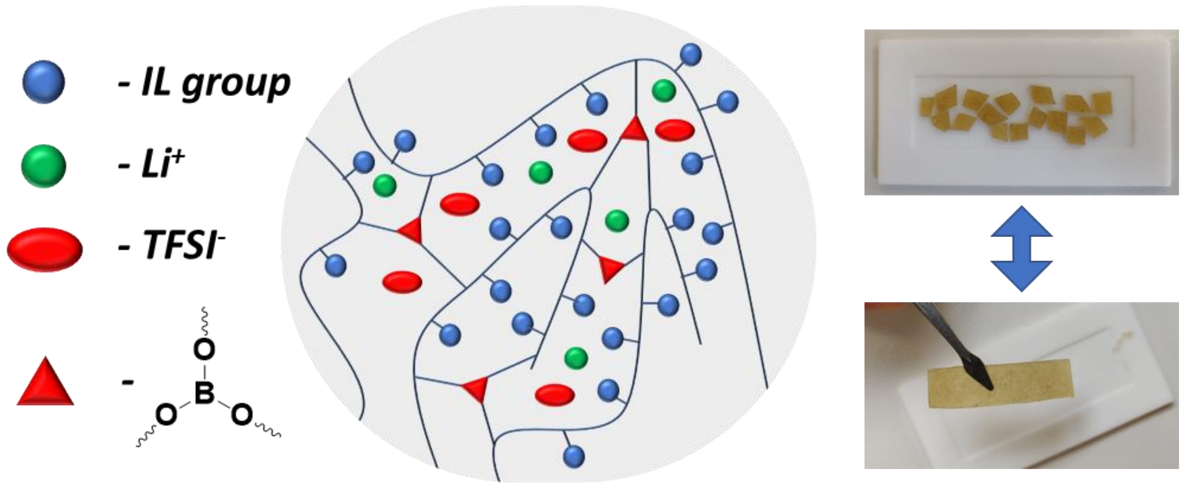

Rechargeable Lithium-ion batteries (LiBs) have become crucial part of our everyday life while covering a wide range of applications. The drawbacks of current technologies (high price, low energy density, environmental and safety issues etc.) and demand for higher performance motivates the development of next generation LiBs. Currently, main safety concerns are associated with the commercially used electrolytes due to presence of flammable organic solvent-based mixtures. Polymer electrolytes (PE) are promising alternatives to overcome shortcomings and make LiBs side much safer for users. Introduction of self-healing features in PE leads to prolonged life-time of LIBs, thus tackling cost and environmental issues. Among different types of polymers poly(ionic liquid)s (PILs) stand out due to their high electrochemical and thermal stability combined with comparably high ionic conductivity.

Here we present solvent free, self-healable, reprocessable, thermally stable, conductive PIL consisting of pyrrolidinium-based repeating units. In addition, PEO-functionalized styrene was used as co-monomer for improving mechanical properties and introducing pendant OH group in the polymer backbone. Addition of boric acid to a precursor polymer matrix leads to formation of dynamic boronic ester bonds, thus forming a vitrimeric material. Dynamic boronic ester linkages form non-permanent crosslinking and allowing reprocessing (at 40 °C), reshaping and also the ability to self-heal the mechanical damage.

Vitrimeric PILs with different crosslinking density and content of lithium salt (LiTFSI) were synthesized and characterized. The conductivity reaches 10^{-5} S/cm at 50 °C in the optimized composition. Furthermore, electrolyte is stable up to 300 °C which is much higher than the operating temperature of batteries. Moreover, the PILs rheological properties fits the required melt flow behavior (above 120 °C) for 3D printing *via* fused deposition modeling, opening the possibility to desing batteries with more complex and diverse design.