Solvent-free vitrimeric poly(ionic liquid) electrolyte for Li-ion batteries

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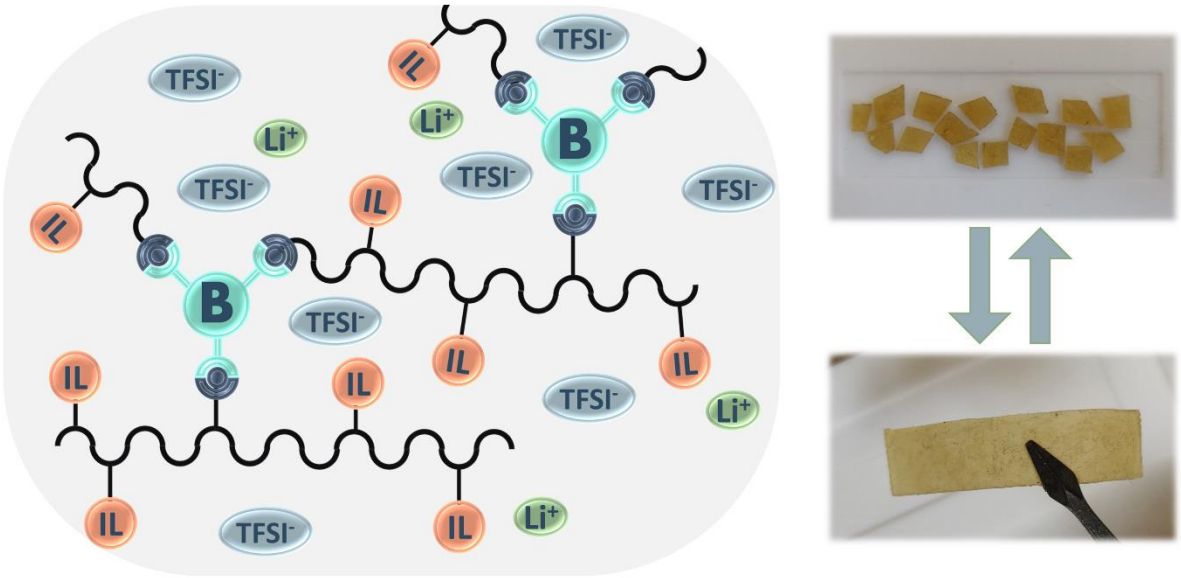

Rechargeable Lithium-ion batteries (LiBs) have become crucial part of our everyday. The drawbacks of current technologies (high price, low energy density, environmental and safety issues etc.) and demand for higher performance motivates the development of next generation LiBs. Currently, main safety concerns are associated with the commercially used flammable electrolytes. Polymer electrolytes (PE) are promising alternatives to overcome shortcomings and make LiBs much safer for users. Introduction of self-healing features in PE leads to prolonged life-time of LIBs, thus tackling cost and environmental issues.

Here we present solvent free, self-healable, reprocessable, thermally stable, conductive Poly(ionic liquid) consisting of pyrrolidinium-based repeating units. PEO-functionalized styrene was used as a co-monomer for improving mechanical properties and introducing pendant OH group in the polymer backbone. Addition of boric acid to a precursor polymer matrix forms dynamic boronic ester bonds, thus yielding in a vitrimeric material. Dynamic boronic ester linkages form non-permanent crosslinking [1] and allowing reprocessing (at 40 °C), reshaping and also the ability to self-heal the mechanical damage.

Pyrrolidinium-based vitrimers with different crosslinking density and content of lithium salt (LiTFSI) were synthesized and characterized. Materials displayed good conductivity (up to 10^{-5} S/cm at 50 °C) and high thermal stability (up to 300 °C). In addition, the PILs rheological properties (above 120 °C) meet the required melt flow behavior [2] for 3D printing *via* fused deposition modeling, offering the possibility to design batteries with more complex and diverse design.

[1] B. B. Jing and C. M. Evans, *Journal of the American Chemical Society* **2019**, *141*, 18932-18937.

[2] a) H. Rupp and W. H. Binder, *Advanced Materials Technologies* **2020**, *5*, 2000509; b) H. Rupp and W. H. Binder, *Macromolecular Rapid Communications* **2021**, *42*, 2000450.