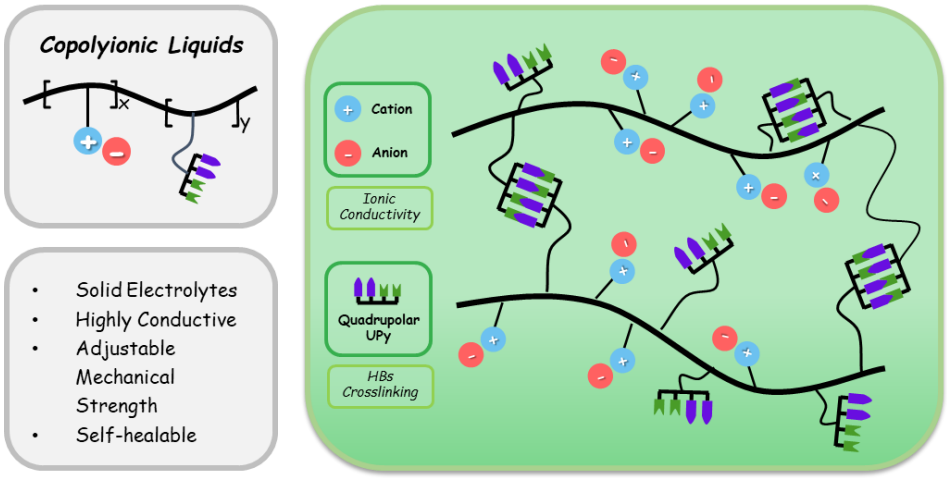
Synthesis and Characterization of Hydrogen Bonded, Self-Healing Polymeric Ionic Liquids as Potential Electrolytes

Authors: Chenming Li, Rajesh Bhandary, Anja Marinow, Wolfgang H. Binder*

Macromolecular Chemistry, Institute of Chemistry, Martin-Luther University Halle-Wittenberg; Von-Danckelmann-Platz 4, D-06120 Halle (Saale), Germany.

Chenming.li@chemie.uni-halle.de
Wolfgang.binder@chemie.uni-halle.de

Keywords: Polymeric ionic liquids; RAFT polymerization; Hydrogen Bonds; Electrolytes;



Abstract:

Batteries are of increasing importance as a cleaner power source compared to traditional carbon-based sources. While solid electrolytes offer promising alternatives to the conventional liquid electrolytes, they could undergo irreversible failures due to, i.e., lithium dendrite growth and volume expansions of the electrodes during cycling. Intrinsic self-healing materials[1-4] bearing dynamic bonding might be proper candidates as potential electrolytes, since the reversible bonding could compensate the micro-failure of the materials consequently extending the lifespan of a battery. Herein, we report the design and synthesis of self-healing electrolytes, able to display enhanced lifetimes and sufficient electrochemical stabilities. By RAFT copolymerization of acrylate-based pyrrolidinium ionic liquids (IL1) with ureidopyrimidinone monomers (SH1), novel polyionic liquids, reinforced by the quadrupolar hydrogen bonding moieties were synthesized. In order to precisely characterize these novel polyionic liquids, a series of homopolymeric ionic liquids with specific molecular weights were prepared, ensuring a proper calibration of the SEC systems in DMF with LiTFSI (0.1 M). Additionally, the thermal behavior of the neat homo/copolyionic liquids was investigated by TGA and DSC. Broadband dielectric spectroscopy (BDS) gave an insight into ionic conductivity of the novel electrolytes and their electrochemical window was determined by cyclic voltammetry (CV). With the increasing content of SH1 group within the polymer chains the consistency of copolyionic liquids alternates from honey-like gels, over brittle solids, and finally to powdery materials, clearly demonstrating an increasing interchain interactions of the ureidopyrimidinone moieties. These materials display self-healing properties and good conductivities, both enabling their embedding as binders in electrode-materials and their use as electrolytes.

Reference:

1. Döhler, D.; Kang, J.; Cooper, C. B.; Tok, J. B. H.; Rupp, H.; Binder, W. H.; Bao, Z., Tuning the Self-Healing Response of Poly(dimethylsiloxane)-Based Elastomers. *ACS Applied Polymer Materials* **2020,** *2* (9), 4127-4139.
2. Campanella, A.; Döhler, D.; Binder, W. H., Self-Healing in Supramolecular Polymers. *Macromolecular Rapid Communications* **2018,** *39* (17), 1700739.
3. Xu, J.; Ding, C.; Chen, P.; Tan, L.; Chen, C.; Fu, J., Intrinsic self-healing polymers for advanced lithium-based batteries: Advances and strategies. *Applied Physics Reviews* **2020,** *7* (3), 031304.
4. Mai, W.; Yu, Q.; Han, C.; Kang, F.; Li, B., Self-Healing Materials for Energy-Storage Devices. *Advanced Functional Materials* **2020,** *30* (24), 1909912.