Development of Organic Ionic Plastic Crystal-Polymer Nanofibre Composites as Solid State Electrolytes

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Organic ionic plastic crystals (OIPCs) are a class of solid-state electrolyte material with good thermal stability, non-flammability, non-volatility and good electrochemical stability. These materials deform easily under stress and can also allow fast transport of ions such as Li⁺ through the rotational and translational motions of the matrix ions. Doping with lithium salts can increase the conductivity[1], and makes them increasingly promising for future application in lithium batteries[2]. However, waxy or powdery OIPCs cannot form a flexible membrane. Using electrospun polymer fibres as a matrix is a viable way of developing all-solid-state, free-standing and flexible electrolytes. We have recently reported that the plastic crystal N-ethyl-N-methylpyrrolidinium tetrafluoroborate ([C2mpyr][BF4]), doped with lithium tetrafluoroborate (LiBF4) and combined with electrospun PVdF fibers can successfully support stable lithium cell cycling, with a capacity over 140 mAh g⁻¹. The conductivity was also enhanced with incorporation of the PVdF[3].

To further the development of such composite electrolytes, it’s important to investigate the influence of polymer nanofibre on the thermal, structural, morphological, and electrochemical properties of OIPCs. Further, understanding the influence of composite composition on electrolyte parameters such as Li transference number, battery cycling performance and stability is key.

In this study, LiFSI doped [C2mpyr][FSI]–PVdF electrospun nanofibers were developed as composite electrolyte membranes. The effects of PVdF incorporation on the plastic crystal were investigated by DSC, impedance spectroscopy, SEM, synchrotron XRD, and NMR. The optimized ratio was determined to be [C2mpyr][FSI]–10wt%PVdF-10mol%LiFSI, and the transport number in this composite was determined to be 0.11.

This composite electrolyte supported more than 500 cycles of a lithium symmetric cell at a current density of 0.05mA/cm² at 50°C. Use of three layers of electrolyte membrane prevented shorting of the battery through dendrite formation. Thus, this composite is a promising solid state electrolyte for lithium batteries.

References: