Comparison of the physicochemical and electrochemical behaviour of mixed anion phosphonium based OIPCs electrolytes for sodium batteries

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Abstract

The physicochemical properties of the phosphonium-based organic ionic plastic crystal (OIPC), trisobutylmethylphosphonium bis(fluorosulfonyl)amide (P$_{1444}$FSI) in mixtures with sodium salts consisting of different anion structures, sodium bis(fluorosulfonyl)amide (NaFSI), sodium bis(trifluoromethanesulfonyl)amide (NaNTf$_2$) and sodium hexafluorophosphate (NaPF$_6$) were investigated. The phase behaviour, ionic conductivity and electrochemical performance at a sodium concentration of 20 mol% for each anion system were compared. 20 mol% P$_{1444}$FSI/NaPF$_6$ displays complicated phase behaviour with additional phase transitions and a higher melting temperature compared to pure P$_{1444}$FSI, indicating the formation of a new compound which is different from pure P$_{1444}$FSI or NaPF$_6$. The system consisting of 20 mol% NaNTf$_2$ exists as a supercooled liquid across the whole temperature range with a glass transition at ~73 °C. At temperatures corresponding to their liquid state, the ionic conductivity values for both 20 mol% NaFSI and NaNTf$_2$ systems are substantially higher than the NaPF$_6$ system and are approximately similar. Na symmetrical cell cycling at room temperature and 50 °C for these two systems at current densities of 0.1 and 0.25 mA cm$^{-2}$ exhibited stable and reversible sodium stripping and plating behaviour with very low polarisation potentials. In contrast, for cells based on the 20 mol% NaPF$_6$ electrolyte, an extended time or higher current density is required in order to form a stable SEI layer before stable cell polarisation behaviour is reached.

Graphical abstract