Structure-property-relationships of network ionic polymer electrolytes and their applications to electrochemical devices

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Next generation applications such as roll up displays, wearable electronics, and tactile communication require safe and flexible solid-state electrolytes with diverse functionalities. We have investigated how molecular design controls key properties (e.g. glass transition, ion transport, modulus) of network ionic polymers which also influence their performance as electrolytes in a range of applications. We first systemically showed, in ammonium networks, ion exchange with 1.5 equiv. of the new counterion was insufficient and lead to 10 times lower in ionic conductivity, 50 K decrease in thermal stability, with minimal change in glass transition temperature (Tg). This points to the importance of preparation for fundamental investigations of charge transport in polymers. Next, imidazolium cation was used to synthesize low Tg, electrochemically stable electrolytes. Polar ethylene oxide linkers were found to increase the ionic conductivity and lower the modulus compared to identical length hydrocarbon counterparts. In response to a 3 V stimulus, both decreased crosslinking and increased polarity of the electrolyte improved the bending strain of actuators which scaled inversely with modulus. These materials were also used as electrolytes for flexible supercapacitors with a reduced graphene oxide composite electrodes. They showed stability up to 120 °C, flexible and 3 V, as well as high capacitance (> 300 F/g) exceeding that of existing polymer or ionic liquid systems. In the final investigation, a Li conducting system was prepared by tethering fluorinated anions to the backbone. We showed how systematic tuning of crosslinking density, side chain length, and ion concentration lead to a 70 K fluctuation in Tg and 3 orders of magnitude shift in ionic conductivity. Decoupling between modulus and ionic conductivity was observed at low crosslinking density showing routes towards future design of polymer electrolytes.