Contribution ID: **25** Type: **Poster**

Dynamic and Structure of Polymer-Cellulose Composite Electrolyte for Li-ion Battery

Tuesday 19 July 2016 16:50 (2h 40m)

Polymer electrolytes are safer, cleaner, and more flexible than liquid electrolytes that are currently being used in Li batteries. However, polyethylene oxide (PEO) based electrolytes are not stiff enough to prevent dendrite formation, which limits the use of Li metal as an anode. They also do not have high enough conductivity to be practical. In amorphous polymer electrolytes, stiffness and conductivity are inversely related because Li motion is coupled to polymer motion, and any attempt to improve conductivity through faster polymer motion results in decreased stiffness. The crystalline PEO6LiX complex is a tunnel-like polymer/salt structure that promotes fast Li motion. The application is limited because high ion conductivity is only observed with short molecular weight PEO, as the molecular weight increases, growth of crystalline lamellae fold tunnels on themselves and restrict conduction.

We show that high aspect ratio nanofillers based on cellulose nanowhiskers promote the formation of tunnel structures. These fillers offer controllable surface chemistry, degree of functionalization and aspect ratio, thus forming an ideal model system. In this work, we fabricate high molecular weight PEO/cellulose nano whiskers nanocomposites. Compared with unfilled electrolytes, room temperature ion conductivity increases up to 1100%. With wide angle x-ray scattering (WAXS), we observe that the structure transitions from amorphous to crystalline as we add cellulose nano whiskers. This is because the interaction between the acidic cellulose surface and polymer chains enhances crystallization. Based on the temperature dependence of conductivity, acidic cellulose nano whiskers reduce the activation energy for Li+ hopping. Using quasi-elastic neutron scattering (QENS), we find that the rotation of PEO6 channels are stabilized when acidic surfaces are present. We believe this is the origin of the low activation energy and high conductivity.

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Session Classification: Poster Session

Track Classification: Energy storage & transformation