High Performance Single-ion Polymer Electrolytes via Macromolecular Engineering

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Single-ion solid polymer electrolytes (SI-SPEs) represent the ultimate solution to the safety issues associated with the use of flammable and toxic liquid electrolytes in commercial Li-ion batteries and for the realization of high energy-density Li-metal batteries. In spite of the considerable research effort in SI-SPEs, the realization of their potential has been hindered by the inability to design materials that possess simultaneously, cation transference number close to unity (i.e. single-ion solid polymer electrolytes, $r_+ = 1$), good mechanical properties, and high ionic conductivity,. In this talk, we introduce the use of novel, stiff/glassy nanostructured polyanion particles, composed of polyanion miktoarm star copolymers of poly(styrene-4-sulfonyltrifluoromethylsulfonyl) imide lithium, PSTFSILi, arms that are complement to longer ion conducting poly(ethylene oxide), PEO, arms, $(PSTFSILi)_n(PEO)_n$, where n ≈ 22, attached to a poly(divinylbenzene), PDVB, crosslinked core as additives to liquid, oligomeric poly(ethylene oxide), PEO, electrolytes for the synthesis of SI-SPEs that are single-ion by design while exhibit an unparalleled combination of high shear modulus and Li-ion conductivity. Key to their performance is the morphology that stems from the ability of the $(PSTFSILi)_n(PEO)_n$ nanoparticles to homogenously disperse within the liquid PEO electrolyte, allowing the development of a highly interconnected network of pure liquid PEO and the profounds effect of mikto arm architecture on the degree of ion dissociations that promotes high ionic conductivity.

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