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Polymer Crystallization-Driven Gelation of an Ionic Liquid
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Polyethylene glycol dissolves in the room temperature ionic liquid 1-ethyl-3-methylimidiazolium ethyl sulfate [EMIM][EtSO4] when heated above about 60C, the neat polymer's melting temperature. At typical polymer molecular weight and concentration, the homogeneous, slightly viscous solution solidifies during subsequent cooling, forming a semitransparent gel. For example, a 5 wt. percent solution of 6000 MW polymer produces a gel with modulus exceeding 1 KPa at 45C; cooled further to room temperature, the gel's modulus rises to a temperature-insensitive plateau of over 100 KPa. By DSC, rheology, and optical microscopy, gelation of the liquid is traced to kinetically frustrated polymer crystallization, a phenomenon previously reported for many pairings of crystallizable polymer and traditional solvent. Polarized optical microscopy reveals nucleation and growth of fibrillar polymer crystals during cooling, and these crystals, here with largest dimenions of tens to hundreds of microns, act as junction points. Melting is at a temperature higher than for gelation. Surprisingly, gelation can occur even when the starting polymer concentration is an order of magnitude below coil overlap. [EMIM][EtSO4] is hygroscopic, and with water uptake, the modulus drops.

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