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Thermodynamics, Structure and Transport in Model Fuel Cell Membranes¹

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Polymer electrolyte membranes (PEM), used to conduct protons from the anode to the cathode of hydrogen fuel cells, are open systems that exchange water with the surrounding air. Proton conductivity is closely coupled to the presence of contiguous hydrated channels within the membrane. In an attempt to understand the underpinnings of the morphology of these systems, the phase behavior of model PEMs comprising block copolymers in equilibrium with humidified air was studied as a function of the relative humidity of the surrounding air, ion content of the copolymer, and temperature. At low humidity, the copolymers exhibit an order-to-disorder transition as a function of increasing temperature. At high humidity, however, increasing temperature results in a disorder-to-order transition. *In-situ* small angle neutron scattering experiments on the open block copolymer system, when combined with water uptake measurement indicate that the disorder-to-order transition is driven by an increase in the partial molar entropy of the water molecules in the ordered phase relative to that in the disordered phase. This is in contrast to most systems wherein increasing entropy results in stabilization of the disordered phase. The coupling between entropy and proton conductivity will be discussed.

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