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Exploring the Parameters Controlling the Crystallinity-Conductivity Correlation of PFSA Ionomers AHMET KUSOGLU, SHOUWEN SHI, ADAM WEBER, Lawrence Berkeley Natl Lab — Perfluorosulfonic-acid (PFSA) ionomers are the most commonly used solid-electrolyte in electrochemical energy devices because of their remarkable conductivity and chemical/mechanical stability, with the latter imparted by their semi-crystalline fluorocarbon backbone. PFSAs owe this unique combination of transport/stability functionalities to their phase-separated morphology of conductive hydrophilic ionic domains and the non-conductive hydrophobic backbone, which are connected via pendant chains. Thus, phase-separation is governed by fractions of backbone and ionic groups, which is controlled by the equivalent weight (EW). Therefore, EW, along with the pendant chain chemistry, directly impact the conductive vs non-conductive regions, and consequently the interrelation between transport and stability. Driven by the need to achieve higher conductivities without disrupting the crystallinity, various pendant-chain chemistries have been developed. In this talk, we will report the results of a systematic investigation on hydration, conductivity, mechanical properties and crystallinity of various types and EWs of PFSA ionomers to (i) develop a structure/property map, and (ii) identify the key parameters controlling morphology and properties. It will be discussed how the pendant-chain and backbone lengths affect the conductivity and crystallinity, respectively. Lastly, the data set will be analyzed to explore universal structure/property relationships for PFSAs.

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