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**Atomistic Simulations of Aggregation in Ionomer Melts**

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Ionomers, polymers containing a small fraction of covalently bound ionic groups, are of interest as possible electrolytes in batteries. A single-ion conducting polymer electrolyte would be safer and have higher efficiency than currently-used liquid electrolytes. However, to date ionomers do not have sufficiently high conductivities for practical application, most likely because the ions tend to form aggregates, leading to slow ion transport. An understanding of the relationships between ionomer chemistry, morphology, and ion transport is needed to design ionomers with improved conductivity. To provide insight into the ionic aggregate morphology, we have performed molecular dynamics simulations of a series of polyethylene-based model ionomer melts, in which the spacing between functional groups is precisely controlled. We vary the counterion type, the neutralization level, and the length of the spacer. The simulations provide new insights into the shape, size and composition of ionic aggregates. In particular, we observe a wide variety of aggregate morphologies, ranging from small spherical aggregates to string-like shapes and large percolated networks. The structure factors calculated from simulation agree well with X-ray scattering data. Depending on the morphology, the simulation and experimental scattering curves can be well fit with either a modified hard sphere or a modified hard cylinder model. These fits, combined with the simulation data, provide the first (indirect) experimental evidence of string-like aggregate morphologies in ionomer melts. We speculate that stringy, percolated aggregates may enhance ionic conduction.