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**Simulation of Ionic Aggregation and Ion Dynamics in Model Ionomers**

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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 the currently-used liquid electrolytes. However, to date ionomeric materials do not have sufficiently high conductivities for practical application. This is most likely because the ions tend to form aggregates, leading to slow ion transport. A key question is therefore how molecular structure affects the ionic aggregation and ion dynamics. To probe these structure-property relationships, we have performed molecular simulations of a set of recently synthesized poly(ethylene-co-acrylic acid) copolymers and ionomers, with a focus on the morphology of the ionic aggregates. The ionomers have a precise, constant spacing of charged groups, making them ideal for direct comparisons with simulations. *Ab initio* calculations give insight into the expected coordination of cations with fragments of the ionomers. All-atom molecular dynamics (MD) simulations of the ionomer melt show aggregation of the ionic groups into extended string-like clusters. An extensive set of coarse-grained molecular dynamics simulations extend the results to longer times and larger length scales. The structure factors calculated from the MD simulations compare favorably with x-ray scattering data. Furthermore, the simulations give a detailed picture of the sizes, shapes, and composition of the ionic aggregates, and how they depend on polymer architecture. Implications for ion transport will be discussed.
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