Organic mixed ion-electron conducting (OMIEC) polymers are capable of transporting both electrons and ions. This unique functionality underpins many emerging applications, including biosensors, electrochemical transistors, and batteries. The fundamental operating principles and structure-function relationships of OMIECs are still being investigated. Computational tools such as coarse-grained molecular dynamics (CG-MD), which use simpler representations than in atomistic modeling, are ideal to study OMIECs, as they can explore the slow dynamics and large length scale features of polymers. Nevertheless, methods development is still required for CG-MD simulations to accurately describe OMIECs.

In this thesis, two CG-MD simulation approaches have been adopted. One is a so-called "top-down" approach to develop a generic model of OMIECs. Top-down models are phenomenological but capable of exploring a broad space of materials variables, including backbone anisotropy, persistence length, side-chain density, and hydrophilicity. This newly developed model was used to interrogate the effect of side-chain polarity and patterning on OMIEC physics. These studies reproduce experimentally observed polymer swelling while for the first time clarifying several molecular factors affecting charge transport, including the role of trap sites, polaron delocalization, electrolyte percolation, and suggesting side-chain patterning as a potential tool to improve OMIEC performance.

The second strategy pursued in this thesis is bottom-up CG-MD modeling of specific atomistic systems. The bottom-up approach enables CG-MD simulations to be quantitatively related to specific materials; yet, the sources of error and methods for addressing them have yet to be systematically established. To address this gap, we have studied the effect of the CG mapping operator, an important CG variable, on the fidelity of atomistic and CG-MD simulations. A major observation from this study is that prevailing CG-MD methods are underdetermined with respect to atomistic training data. In a separate study, we have proposed a hybrid machine-learning and physics-based CG-MD framework that utilizes information from multiple sources and improves on the accuracy of ML-only bottom-up CG-MD approaches.