

Prediction of Electronic Properties of Radical-Containing Polymers at Coarse-Grained Resolutions

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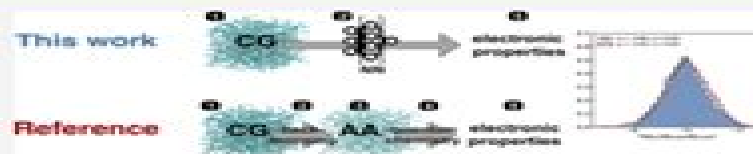


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ABSTRACT: The properties of soft electronic materials depend on the coupling of electronic and conformational degrees of freedom over a wide range of spatiotemporal scales. The description of such properties requires multiscale approaches capable of, at the same time, accessing electronic properties and sampling the conformational space of soft materials. This could in principle be realized by connecting the coarse-grained (CG) methodologies required for adequate conformational sampling to conformationally averaged electronic property distributions via backmapping to atomistic-resolution level models and repeated quantum-chemical calculations. Computational demands of such approaches, however, have hindered their application in high-throughput computer-aided soft materials discovery. Here, we present a method that, combining machine learning and CG techniques, can replace traditional backmapping-based approaches without sacrificing accuracy. We illustrate the method for an emerging class of soft electronic materials, namely, nonconjugated, radical-containing polymers, promising materials for all-organic energy storage. Supervised machine learning models are trained to learn the dependence of electronic properties on polymer conformation at CG resolutions. We then parametrize CG models that retain electronic structure information, simulate CG condensed phases, and predict the electronic properties of such phases solely from the CG degrees of freedom. We validate our method by comparing it against a full backmapping-based approach and find good agreement between both methods. This work demonstrates the potential of the proposed method to accelerate multiscale workflows and provides a framework for the development of CG models that retain electronic structure information.



INTRODUCTION

Radical-containing polymers, also known as open-shell macromolecules, macromolecular radicals, or simply radical polymers, possess intriguing redox, optoelectronic, and magnetic characteristics that make them appealing for applications ranging from energy storage and optoelectronics to spintronics and memory storage.^{1–6} Nonconjugated, radical-containing polymers are organic polymers that have a nonconjugated backbone bearing pendant stable radical sites. They constitute a class of charge-carrying polymers that do not rely on π -conjugation to transport charges successfully. The rational design of radical polymers with enhanced characteristics could be greatly advanced by deriving relationships that connect their molecular structure, morphology, and electronic properties. These relationships are inherently multiscale, involving the coupling of electronic and conformational degrees of freedom over a wide range of spatiotemporal scales. New modeling approaches capable of describing such coupling are needed.

Recent work has shown that coarse-grained (CG) models can be used to probe polymeric material length and time scales reaching the mesoscale.^{7–10} In contrast, explicit quantum-chemical calculations, usually using density functional theory (DFT), which are necessary to access electronic properties,^{11–13} are extraordinarily demanding and can only capture picosecond and Angstrom-level processes. To bridge these two

scales, that is, to take into account large-scale morphological features generated via self-assembly processes (possibly as a function of processing conditions) when computing electronic properties, recent efforts have sought to introduce new multiscale modeling approaches.^{12,14} In such approaches, the soft material morphology generated via CG simulations is backmapped to the atomistic resolution required for the quantum-chemical calculations, and such calculations are then performed on conformations drawn from the backmapped morphologies. These multiscale approaches have been primarily developed in the context of organic semiconductors^{12,15–17} and provide a means to access the conformational dependence of electronic properties. However, considerable computational demands and workflow complexity of such have limited their applicability for high-throughput computer-aided materials discovery.

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Electronic Properties Of Polymers

Michael Hoelscher



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