COMPUTATIONAL MODELING OF POLYMERIC NANOSTRUCTURED MATERIALS ACROSS SCALES: FROM ATOMS TO MACROSCOPIC PROPERTIES

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ABSTRACT

The computational study of complex polymeric materials is a very challenging field, due to the broad spectrum of the underlying length and time scales. Here, we present a hierarchical multi-scale methodology for predicting the macroscopic properties of polymer-based nanostructured systems, which involves atomistic and coarse-grained simulations, as well as continuum models. The coarse-grained (CG) models are derived through a "bottom-up" data-driven strategy, using information from the detailed atomistic scale, for the given chemistry. The systematic linking between the atomistic and the chemistry-specific CG scale, allows the study of a broad range of molecular weights, for specific polymers, without any adjustable parameter [1-2]. At the same time, machine learning (ML) algorithms have been developed to re-introduce atomic detail in the CG scale, and thus obtaining atomistic configurations of high molecular weight polymers [2]. The proposed hierarchical computational scheme allows the study of macromolecular systems, of high molecular weight, over a broad range of time scales, from a few fs up to ms. and the prediction of their (structural, dynamical, rheological, etc.) properties [3]

The proposed approach is applied to provide a fundamental understanding of the mechanism of mechanical reinforcement in glassy polymer nanocomposites, which is of paramount importance for their tailored design. We present a detailed investigation, via atomistic simulation, of the coupling between density, structure, and conformations of polymer chains concerning their role in mechanical reinforcement [4]. Probing the properties at the molecular level reveals that the effective mass density as well as the rigidity of the matrix region changes with filler volume fraction, while that of the interphase remains constant. The origin of the mechanical reinforcement is attributed to the heterogeneous chain conformations in the vicinity of the nanoparticles, involving a 2-fold mechanism. In the low-loading regime, the reinforcement comes mainly from a thin, single-molecule, 2D-like layer of adsorbed polymer segments on the nanoparticle, whereas in the high-loading regime, the reinforcement is dominated by the coupling between train and bridge conformations; the latter involves segments connecting neighboring nanoparticles.

KEYWORDS: Molecular simulations, polymer nanocomposites, machine learning methods

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