Mechanical Engineering Doctoral Defense

Multiscale modeling and molecular dynamics simulation of phase-segregated polymers: polyethylene and polyurea

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abstract

The exceptional physical and mechanical properties of phase-segregated polymers, such as the high toughness of high density polyethylene and the excellent blast-protecting capability of polyurea, are strongly related to the morphologies and nanoscale structural features of polymers.
These features permit significant molecular reorganization to take place, such as the relaxation of macromolecules through crystalline/amorphous interface in polyethylene, and the shock-induced chemical reaction in polyurea. Owing to the large spatial and long temporal scales related to the complex reorganization process, modeling and simulation of phase-segregated polymers remain a challenge. To study molecular relaxation in semicrystalline polyethylene, a coarse--grained models of polyethylene is calibrated from supercooled atomistic melt, which can accurately reproduce the structural properties including the local structural distributions of amorphous phases, thermal properties including the glass transition and melt temperatures, and dynamic properties including the vast difference between the relaxation time scales in different phases of polyethylene. A hybrid Monte Carlo routine is further developed to generate semicrystalline configurations of polyethylene. The structural and morphological properties of the simulated crystalline/amorphous interfaces are validated and verified with available experimental measurement, theoretical predictions, and simulation results. The generated systems accurately predict the overall activation energy for the relaxation of crystalline stems. Further, it gives prediction of range of the lengths of amorphous segments that relates to the highest and lowest activation energy of the connected crystalline stems. These predictions can guide the development of tougher semicrystalline polymers by providing a fundamental understanding of how nanoscale morphology contributes to chain mobility. Moreover, as a first step toward understanding how the strength and viscoelastic property of polyurea change under shock, the initial chemical event including bond dissociation/association is probed and the fidelity of classical and density functional theory (DFT) simulations in predicting shock Hugoniot is studied.

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