Mechanical Engineering Doctoral Defense

A Structure-property Relationship Study of Semicrystalline Polyethylene through the Development of Coarse-grained Methods and Representative Systems

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Abstract

"Polyethylene is a common semicrystalline polymer material known for its favorable strength-to-weight ratio, which originates from its complex microstructure at various length scales. However, the stress relaxation mechanisms of polyethylene and other semicrystalline polymers are yet to be fully understood. Molecular dynamics simulation is a frequently used tool to investigate microstructure changes and give insights into the deformation mechanisms, but its space and time scales are highly limited due to the high demand for computational power, but this can be alleviated with coarse-graining. In this work, a coarse-grain model was developed which considered the correlation between the probability distribution of bond--length and bond--angle to improve the representability of the local conformations. In order to parameterize the model, an extended iterative Boltzmann inversion method was introduced that incorporated the mentioned correlation and employed kernel density estimation (KDE) in representing the probability distribution where the bandwidth of the KDE can be used to tune the characteristics of the model. The resulting coarse-grain models successfully reproduced the correlation in the probability distribution of bond--length and bond--angle with only a 10\% increase in the computational cost compared to the models without considering the correlation.

Aside from coarse-grain potentials, semicrystalline representation systems with realistic molecular weights and configurations are also essential in investigating the deformation mechanisms. A hybrid Monte-Carlo method was developed to efficiently generate semicrystalline systems with controllable lengths of the topological segments in the amorphous phases, facilitating investigation into the impact of the topological segments on the mechanical properties of semicrystalline polyethylene. It was discovered that the bridges significantly affect the strain hardening in the tension test while the loops have minimal impact. Shorter bridges are engaged earlier than the longer ones, thus the strain--hardening onsets earlier in the systems with shorter bridges."

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