Physical origin of strain hardening in glassy polymers and constitutive modeling based on molecular dynamics simulations

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Abstract

Glassy polymers typically exhibit strain hardening at large deformations due to their chain-like molecules, effectively increasing the toughness of the materials. The rate- and temperature-dependence of strain hardening increases the difficulty of accurately modeling the mechanical response in the hardening region, which is essential for studying the failure mechanisms of glassy polymers. This contribution presents the study of the micromechanisms underlying strain hardening in glassy polymers based on molecular dynamics (MD) simulations (1). Firstly, the stress response in the hardening region is attributed to the interactions of bond and angle potentials through decomposition of the virial components. Furthermore, the primary contribution to strain hardening is identified as the stretch of polymer bonds by evaluating the microscopic properties, where the disentanglement only plays a small role. Interestingly, rather than the average bond stretch, we find that the key contributions to stress response come from a subset of bonds at the upper tail of the stretch distribution, where the stress in the hardening region can be correlated with the average stretch of the most extended bonds in each polymer chain, independent of temperatures and strain rates. We further incorporate this mechanism into a constitutive model in continuum mechanics with orientation-induced back stress, developing a model that accurately reproduces the stress response of the molecular systems in uniaxial tension over a wide range of temperatures and strain rates in their glassy state. With 13 parameters in the constitutive model, 10 of them can be directly estimated from the MD simulation results. (1) W. Zhao. Rate- and temperature-dependent strain hardening in glassy polymers: Micromechanisms and constitutive modeling based on molecular dynamics simulations. arXiv preprint arXiv:2411.07811.

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