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Development of Constitutive Models for the Long-term Performance of Glassy Polymers

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Abstract

The development of accurate constitutive models for glassy polymers remains an outstanding problem in polymer engineering, where there is a need to both describe the diversity of nonlinear relaxation behavior exhibited by these materials as well as acknowledge the underlying molecular deformation processes. Both experiments and molecular simulation clearly indicate that glassy polymers exhibit fluctuations at the nanoscale, where there are order-of-magnitude differences in the mobility between neighboring meso-domains, i.e. dynamic heterogeneity. A new constitutive approach has been developed that explicitly acknowledges the nanoscale fluctuations present in the glassy state. This stochastic constitutive model (SCM) only has a single relaxation time, but can describe the complex nonlinear relaxation behavior exhibited by glassy polymer including yield. Of particular significance is the ability of the SCM to predict post-yield softening – a phenomena that heretofore has not been predicted by existing constitutive models. The evolution of the fluctuating internal variables provides a mechanistic picture of what is controlling the macroscopic behavior of glassy polymers in complex thermal-deformation histories. Because fluctuations are explicitly included, the SCM may provide a bridge between molecular simulations of polymer behavior and traditional continuum mechanics.