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The Significance of Incorporating Nanoscale Fluctuations in a Constitutive Description of Glassy Polymers[1]

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The current picture of the glass involves dynamic heterogeneity, where nanoscopic regions of the glass have order-of-magnitude differences in local mobility that evolve with time. Dynamic heterogeneity provides a critical challenge to the traditional nonlinear continuum models, where both temporal and spatial fluctuations are averaged as a result of the continuum postulate. In order to acknowledge dynamic heterogeneity, a Stochastic Constitutive Model (SCM) has been developed to describe the nonlinear viscoelastic behavior of polymeric glasses, where (i) temporal fluctuations are explicitly included and (ii) the *local* mobility depends upon the *local* state of the material (e.g. local stress and local entropy) vs. traditional viscoelastic/viscoelastic models where macroscopic mobility depends upon the macroscopic state. The SCM is able to describe a number of nonlinear relaxation phenomena that cannot be predicted by traditional nonlinear viscoelastic/viscoplastic models, including (i) post-yield stress softening and its dependence on annealing time, (ii) the inversion of the strain dependence of nonlinear stress relaxation with the loading rate, (iii) stress memory and (iv) tertiary creep and creep-recovery. This paper will argue that incorporation of nanoscopic fluctuations is a necessary component for a description of the thermomechanical behavior of polymeric glasses.

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