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Mesoscale simulation of stress relaxation in thin polymer films and the connection to nanocomposites

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ABSTRACT

Key insight into interphase formation and confinement effects in nanocomposites has recently come from studies on polymer thin films supported on solid substrates. In these thin films, both the free surface and the solid supporting layer cause complex changes in the behavior of the polymer. The range and magnitude of these effects have been singled out by systematically varying the boundary conditions (free standing film, supported thin film, and polymer layer confined between two surfaces) and surface/polymer chemistry. Most importantly, the Schadler group and the Torkelson group have shown a quantitative equivalence between nanocomposites and thin films with regards to glass-transition temperature (T_g) via the calculation of an equivalent metric of confinement within the nanocomposite from the distribution of filler surface-to-surface distances. This finding is important because it allows for direct prediction of the T_g of the nanocomposite directly from thin film measurements and microstructural statistics, leveraging current capabilities in accurate computational/experimental characterization of film properties. However, it is currently unknown whether the thin-film analogy can be extended into the constitutive behavior of polymer nanocomposites, most importantly the stress relaxation behavior of the matrix that governs viscoelastic behavior. With an ultimate aim to address this issue, we have begun examining the stress-relaxation in doubly supported polymer thin films through coarse grained simulation using the FENE model. The current study elucidates the connection among film thickness, interfacial energy, and stress relaxation dynamics. In order to characterize the dynamic relaxation behavior of the films at constant temperature, we calculate via an extended, tensorial Green–Kubo relation the linear shear-relaxation modulus from equilibrium coarse-grained simulations of the bulk and of films of varying thickness. We then compare the simulated relaxation moduli to both the Rouse model and the theory of Likhtman and McLeish (originally based on the based on the tube model), with the additional changes proposed by Hou, Svaneborg, Everaers, and Grest. Applications to the continuum mechanics of both thin films and nanocomposites will be discussed.