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Dynamics in Confined Systems: Polymer Thin Films and Surfaces¹

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Here we visit the problem of dynamics of polymers confined in ultrathin films and at surfaces. We present results obtained on ultrathin polymer films using a novel nanobubble inflation device to measure the biaxial creep of films as thin as 13 nm. The results show that the viscoelastic response of PVAc in the glass transition regime is similar to that of the bulk material, while polystyrene shows considerable acceleration of the molecular dynamics that corresponds to a 40 K or more reduction in the glass transition. We will discuss the breadth of the transition, as this is related to the presence of a liquid layer at the surface as well as to the thickness of such a layer. We also discuss the effects of film thickness on the rubbery plateau response of these films. An observed stiffening of the material is found that gives an apparent plateau compliance that is over 100 times smaller than that of the bulk material. In the thinnest films a significant fraction of the deformation energy comes from surface tensions. However, in films above approximately 30 nm, this contribution is less than 30% of the total energy of deformation and suggests that much of the stiffening is due to a thin film effect. This is discussed in detail. Finally, we show results for nanoparticle embedment experiments and argue that the mobility increases observed on the polymer surface are insufficient to explain a reduction of 40 K or more of the glass transition in ultrathin polymer films.

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