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Dynamics in ultrathin films[1]

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There is considerable interest in, yet large variability in results on the dynamics of ultrathin films. A question that arises in all cases is whether or not we are measuring dynamics and/or are different measurements probing the same dynamics. Hence, there are multiple experiments reporting changes in the glass transition temperature as based on a "pseudo-thermodynamic" measurement in which a parameter (film thickness, fluorescence intensity, index of refraction) vary as a function of temperature in a way that is reminiscent of the macroscopic volume-temperature in glass-forming liquids. Yet, these are not direct measurements of the dynamics. In the case of supported films, dynamics have been directly probed by dielectric measurements and mechanical measurements, the latter being complicated by contact mechanics problems or limited to extremely thin surface layers. Perhaps, supported films are the most practical problem, but not the most interesting because the effects of apparently increased mobility are small relative to what has been seen in freely standing polystyrene films and similar to the behavior seen in nanopore confined liquids. In the case of essentially freely standing polystyrene films, two types of measurement stand out. Hole growth and bubble inflation measurements. The former seems to give relatively little change of dynamics until the glass transition of the material in the bulk state is reached while the latter, and consistent with the behavior reported for Tg by "pseudo-thermodynamic" methods shows very large decreases in relaxation times corresponding to up to 50 K reductions in the Tg for 10 nm films of polystyrene. Some possible reasons for the differences in behaviors and other contradictory results will be discussed.

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