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Glass Transition Temperature Reductions in Freely-Standing Films of Different Polymers[1]

JOHN DUTCHER, University of Guelph

The effects of confinement and free surfaces on the dynamics of polymers in thin films have been studied extensively since the original observation of reductions in T_g with decreasing film thickness h in thin polystyrene (PS) films [1]. One particularly striking result, which is yet to be understood in detail, is the observation of very large, molecular weight (MW) dependent reductions of T_g in very thin, freely-standing PS films using Brillouin light scattering and ellipsometry [2]. We have recently measured T_g (h, MW) for freely-standing PMMA films [3] and we find that the results are in qualitative agreement with those obtained for freely-standing PS films. However, the overall magnitude of the T_g reduction is much less (by roughly a factor of three) for the high-MW freely-standing PMMA films than for freely-standing PS films of comparable MW and h. The observed differences between the freely-standing PMMA and PS film data suggest that differences in chemical structure determine the magnitude of the T_g reduction and we discuss the possible origins of these differences. Our analysis of the MW-dependence of the T_g reductions suggests that the mechanism responsible for the MW-dependent T_g reductions observed in the high-MW freely-standing films is different than that responsible for the MW-independent T_g reductions observed in low-MW freely-standing and supported films. [1] Keddie et al., Europhys. Lett. **27**, 59 (1994); [2] Dalnoki-Veress et al., Phys. Rev. E **63**, 031801 (2001); [3] Roth and Dutcher, Eur. Phys. J. E **20**, 441 (2006).

[1]work performed with Connie Roth, Adam Pound, Stephen Kamp and Chris Murray