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Confinement and the Glass Transition Temperature in Supported Polymer Films: Molecular Weight, Repeat Unit Modification, and Cooperativity Length Scale Investigations MANISH K. MUNDRA, JOHN M. TORKELSON, Northwestern University, Evanston, IL 60208-3120 — It is well known that the glass transition temperatures, Tgs, of supported polystyrene (PS) films decrease dramatically with decreasing film thickness below 60-80 nm. However, a detailed understanding of the cause of this effect is lacking. We have investigated the impact of several parameters, including polymer molecular weight (MW), repeat unit structure, and the length scale of cooperatively rearranging regions in bulk. There is no significant effect of PS MW on the Tg-confinement effect over a range of 5,000 to 3,000,000 g/mol. In contrast, the strength of the Tg reduction and the onset of the confinement effect increase dramatically upon changing the polymer from PS to poly(4-tert-butylstyrene) (PTBS), with PTBS exhibiting a Tg reduction relative to bulk at a thickness of 300-400 nm. PTBS also shows a Tg reduction relative to bulk of 47 K in a 21-nm-thick film, more than twice that observed in a PS film of identical thickness. Characterization of the length scale of cooperatively rearranging regions has been done by differential scanning calorimetry but reveals at best a limited correlation with the confinement effect.

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