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The Distribution of Tgs in Thin and Ultrathin Methacrylate-Based Polymer Films: Percolation of Free Surface and Interface Effects over Tens and Hundreds of Nanometers. RODNEY D. PRIESTLEY, MANISH K. MUNDRA, PERLA RITTIGSTEIN, LINDA J. BROADBELT, JOHN M. TORKELSON, Northwestern University, Evanston, IL 60208 — A multilayer/fluorescence method is used to measure the distribution of Tgs in poly(methyl methacrylate) (PMMA) films. The average Tg increases with decreasing total film thickness (h) below 90 nm. In bulk, bilayer films, the free surface layer exhibits a reduced Tg at a thickness of 30 nm or less. The Tg reduction at the free surface is a fraction of that in polystyrene (PS) films; a 14-nm-thick free surface layer exhibits a Tg reduction of $\sim$ 6 K in PMMA and $\sim$ 32 K in PS. The Tg increase observed in the substrate layer of a PMMA bilayer, bulk film exceeds the Tg reduction observed at the free surface and occurs over longer length scales. An amazing effect of confinement of the total multilayer film on the free surface layer Tg of PMMA has been observed. When h $<$ 250 nm, a 12-nm-thick free surface layer exhibits an increase in Tg with decreasing h, and the free surface layer Tg exceeds that of bulk Tg when h $<$ 90 nm. This is the first demonstration that a free surface layer can exhibit Tg $>$ bulk Tg and means that the perturbation of Tg dynamics at the substrate can percolate over hundreds of nanometers in PMMA films. These results will be contrasted with those of poly(isobutyl methacrylate) films which exhibit no average Tg-confinement effect.

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