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Determination of Physical Aging in Thin Polymer Films via Fluorescence: Effects of Confinement and Attractive vs. Neutral Polymer-Substrate Interactions RODNEY D. PRIESTLEY, LINDA J. BROADBELT, JOHN M. TORKELSON, Northwestern University, Evanston, IL 60208-3120 — The effect of nanoconfinement on physical aging of polystyrene (PS) and poly(methyl methacrylate) (PMMA) is studied by fluorescence. Rotor dyes are used as probes ($<$ 0.2wt%) dispersed in the polymer or as labels covalently attached ($<$ 1 label/400 repeat units) to the polymer. Fluorescence intensity increases as local specific free volume/local mobility surrounding the dye decreases, exhibiting a nearly linear change with logarithmic aging time. Thin (500-nm-thick) and ultrathin (20-nm-thick) films supported on silica are annealed above and below the bulk glass transition temperature (Tg,bulk). With PMMA, which has attractive polymer-substrate interactions, ultrathin films exhibit aging at Tg,bulk + 7 K while thin films do not. With PS, which lacks polymer-substrate interactions, thin films exhibit physical aging at Tg,bulk - 10 K while ultrathin films do not. These results are explained by an enhancement (reduction) in Tg of the ultrathin PMMA (PS) film, relative to bulk. Structural recovery is dramatically reduced in ultrathin PMMA films, indicating that attractive interactions significantly affect structural recovery near the substrate.

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