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Physical Aging of Thin and Ultrathin Free-Standing Polymer Films: Effect of Stress and Reduced Glass Transitions JUSTIN PYE, CONNIE ROTH, Dept. of Physics, Emory University — While great effort has been made in elucidating the effect of confinement on the glass transition (Tg) in polymers, considerably less work has been done on physical aging. Starting with supported films, we have previously shown that the reduced physical aging rates in ultrathin polystyrene (PS) films can be linked to the reduced Tg near the free surface [Macromolecules 2010, 43, 8296]. We then showed that high molecular weight (MW) free-standing PS films have two reduced Tgs suggesting that two separate mechanisms are acting simultaneously to propagate enhanced mobility at the free surface deeper into the film [PRL 2011, 107, 235701]. To help determine the mechanisms of these two reduced Tgs, we performed physical aging measurements on these high MW free-standing PS films. For thick films (220-1800 nm) in which there are no Tg reductions, we find that the physical aging rate depends strongly on stress caused by thermal expansion mismatch between film and support. This stress, applied to the films as they are quenched into the glassy state, can nearly double the physical aging rate when changing the frame material from polycarbonate to silicon [Macromolecules 2013, DOI:10.1021/ma401872u]. Finally, ultrathin high MW PS films held at a temperature between the two Tgs do exhibit physical aging, indicating that at least some of the film is glassy between these two transitions.

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