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Local Variation of Fragility and Glass Transition Temperature of Ultra-thin Supported Polymer Films PAUL HANAKATA, Department of Physics, Wesleyan University, Middletown, CT 06459, USA, JACK DOUGLAS, Polymers Division, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA, FRANCIS STARR, Department of Physics, Wesleyan University, Middletown, CT 06459, USA — Extensive studies have shown that the properties of ultra-thin polymer films can differ significantly from the bulk. The effect of the film thickness h on the glass transition temperature has been widely examined, but this does not account for the fragility of glass-formation, which quantifies how rapidly relaxation time varies with temperature T . Accordingly, we simulate polymer films of a bead-spring model on a smooth or rough surface and determine both T_g and fragility, both as function of h and film depth. We find that the commonly invoked free-volume layer model does not describe our results. In addition, as opposed to the bulk, we find that T_g and fragility do not generally vary proportionally. Therefore, determination of fragility is essential for the characterization of dynamic changes in film. Finally, we relate these changes of fragility to changes in the cooperative monomer dynamics.

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