Glass Transition Dynamics and Surface Layer Mobility in Unentangled Polystyrene Films

ZHAOHUI YANG, Boston University, YOSHIHISA FUJII, F.K. LEE, C.H. LAM, DONGDONG PENG, OPHELIA TSUI — Most polymers solidify into a glassy amorphous state, accompanied by a rapid increase in the viscosity when cooled below the glass transition temperature (Tg). There has been an ongoing debate on whether the Tg changes with decreasing polymer film thickness and the origin of the changes. We measured the viscosity of unentangled, short-chain polystyrene (2.4kg/mol) films on silicon at different temperatures and found that the transition temperature for the viscosity decreases with decreasing films thickness, in agreement with the changes in the Tg of the films observed by thermal expansion measurements. By applying the hydrodynamics equations to the films, we are able to explain the data fully by assuming that a highly mobile layer is present within the top 2.3 nm thick region of the films and follows an Arrhenius dynamics while the remaining of the films is bulk-like.

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