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Vitrification of thin polymer films: from linear chain to soft-colloid like behavior EMMANOUIL GLYNOS, BRADLEY FRIEBERG, University of Michigan, GEORGIOS SAKELLARIOU, University of Athens, ALEXANDROS CHREMOS, Imperial University, PETER GREEN, University of Michigan — The glass transition temperature T_g of sufficiently thin, supported, polymer films is dependent on the film thickness. Based on the nature of the polymer substrate interactions T_g may increase, $\Delta T_g > 0$, or decrease, $\Delta T_g < 0$, in relation to the bulk. We show that for star-shaped macromolecules the value of ΔT_g depends on the functionality f of the molecule, for polymer films supported by the same substrate. Specifically in the case of polystyrene (PS) macromolecules, with arms of molecular weight $M_{\text{arm}} < 10$ kg./mol., supported by silicon oxide substrates, $\Delta T_g < 0$, when $f < 4$. For much higher functionalities, $f \geq 32$, where the polymer exhibits soft-colloid like behavior $\Delta T_g \sim 0$. For values of $4 < f < 32$, $\Delta T_g > 0$. The transition from the linear-chain to the soft-colloid behavior is gradual and occurs with increasing f and/or decreasing M_{arm} . With the help of molecular dynamics simulations we rationalize this behavior in terms of competing entropic effects, associated with changes in f and M_{arm} , which drives the ability of these molecules to efficiently pack at interfaces.

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