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Glassy structural relaxation of star-shaped polymers[1] BRADLEY FRIEBERG, EMMANOUIL GLYNOS, University of Michigan, GEORGIOS SAKELLARIOU, University of Athens, PETER GREEN, University of Michigan — Time-dependent changes of thermodynamic properties due to structural relaxations, physical aging, occur in all glasses. In the case of linear polymers, the aging rate is independent of the degree of polymerization at a given aging temperature, relative to the average glass transition temperature. In contrast, we demonstrate that star-shaped macromolecules exhibit average structural relaxations that are dependent on both the number of arms, f, and the degree of polymerization of each arm, Narm. In particular, while increasing f, and/or decreasing Narm, the average segmental relaxation rate decreases, and can be up to a factor of two times lower for star-shaped molecules compared to their linear analogs. We reconcile these differences in terms of the free volume diffusion and its relation to the segmental motions in the glass state. We propose that this ideal class of polymeric materials, star-shaped molecules, can be used in order to tailor the physical properties on a molecular level, by simply changing the polymer architecture.

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