

Abstract Submitted
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Statistical Mechanical Theory of Penetrant Diffusion in Polymer Melts and Glasses RUI ZHANG, KENNETH SCHWEIZER, University of Illinois at Urbana-Champaign — We generalize our force-level, self-consistent nonlinear Langevin equation theory of activated diffusion of a dilute spherical penetrant in hard sphere fluids [1] to predict the long-time diffusivity of molecular penetrants in supercooled polymer liquids and non-aging glasses. Chemical complexity is treated using an a priori mapping to a temperature-dependent hard sphere mixture model where polymers are disconnected into effective spheres based on the Kuhn length as the relevant coarse graining scale. A key parameter for mobility is the penetrant to polymer segment diameter ratio, R . Our calculations agree well with experimental measurements for a wide range of temperatures, penetrant sizes (from gas molecules with $R \sim 0.3$ to aromatic molecules with $R \sim 1$) and diverse amorphous polymers, over 10 decades variation of penetrant diffusivity. Structural parameter transferability is good. We have also formulated a theory at finite penetrant loading for the coupled penetrant-polymer dynamics in chemically (nearly) matched mixtures (e.g., toluene-polystyrene) which captures well the increase of penetrant diffusivity and decrease of polymer matrix vitrification temperature with increasing loading. [1] R. Zhang and K. S. Schweizer, *J. Chem. Phys.*, 143, 144906 (2015).

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