Notes

Comment on "A Molecular Dynamics Simulation Study of Relaxation Processes in the Dynamical Fast Component of Miscible Polymer Blends"

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Bedrov and Smith[1] performed molecular dynamics simulations of model miscible polymer blends consisting of chemically realistic 1,4-polybutadiene (CR-PBD) (the slow or higher T_g component) and PBD chains with reduced dihedral barriers (LB-PBD) (the fast component with lower T_g). The simulation was designed to study the influence of the slow component on the local segmental (or α-) relaxation and the β-relaxation in blends. Before blending, the α- and β-relaxations of the fast component are too close in frequency, so that the weaker β-relaxation cannot be clearly resolved. However, with addition of the slow component, Bedrov and Smith found a monotonic increase in the separation between the α- and β-relaxations of the fast component, whereby the two relaxations become well resolved. The increased separation between the two is due to the strong increase of the α-relaxation time, $\tau_{\alpha f}$, of the fast component with increasing concentration of the slow component, concomitant with a negligible change of the relaxation time, $\tau_{\beta f}$, of the β-relaxation of the fast component. The results of Bedrov and Smith are an important advance in molecular dynamics simulations of polymer blends, which should impact theoretical models of the component dynamics in blends. We would like to point out that similar results had been derived from the coupling model and compared well with experimental data of polymer blend component dynamics,[2] mixtures of molecular glass-formers,[3−5] and mixtures of a molecular glass-former with a polymer.[6] In these previous works the primitive relaxation time of the coupling model (subsequently identified with the β-relaxation time) was unchanged with blending, and the varying relation between the α- and β-relaxations of both the fast and the slow components with composition was predicted, in agreement with existing experimental data and with the recent simulation of Bedrov and Smith.

Moreover, Bedrov and Smith[1] found that the frequency dispersion of the α-relaxation for the slower CR-PBD becomes *narrower* when blended with the faster LB-PBD. This simulation result is consistent with the coupling model prediction[2−6] that the coupling parameter of CR-PBD in the blend is smaller than in neat CR-PBD. The consequent narrowing by this mechanism (counter to the usual broadening due to concentration fluctuations) yields the result[4−6] also found by Bedrov and Smith. Thus, the purpose of this paper is to note the correspondence between simulation results of Bedrov and Smith and published studies based on the coupling model.

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References and Notes

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