Supporting Information

Heterogeneous Polymer Dynamics Explored Using Static ¹H NMR Spectra

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Figure S1. Loss tangent versus temperature obtained from dynamic mechanical analysis (DMA) for fully cured BTT-TCDDA networks with different thiol-acrylate stoichiometry ($R = (SH)_0/(C=C)_0$). With decreasing R, T_g and the breadth of the transition increases.



Figure S2. Simulation of the variation of the spectral second moment (M_2) for different pre-exponential correlation time, τ_0 , assuming the Arrhenius temperate dependence shown in Eqn. 2 as a function of **(a)** temperature and **(b)** the reduced temperature. The activation energy, E_a , was held constant at 33 kJ/mol.



Figure S3. Simulated probability distribution assuming (a) a Gaussian distribution (Eqn. A8) of the activation energy E_a for different distribution widths σ (b) the corresponding correlation time τ distribution using Arrhenius relationship (Eqn. 2), and (c) a Davidson-Cole distribution with the characteristic correlation time τ_z and the distribution parameter ε (Eqn. A9). Simulations obtained assuming a mean $E_a = 31 \text{ kJ/mol}$, $\tau_0 = 0.5905 \text{ ns and } T = 423 \text{ K}$.



Figure S4. Correlation between thiol-acrylate stoichiometry ($R = (SH)_0/(C=C)_0$) and **(a)** activation energy (E_a) and **(b)** the natural log of the pre-exponential correlation time (τ_0) obtained from the analysis of the Arrhenius temperature behavior in **Figure 11**.