

# Supporting Information

## Effects of Crosslink Density and Distribution on Static and Dynamic Properties of Chemically Crosslinked Polymers

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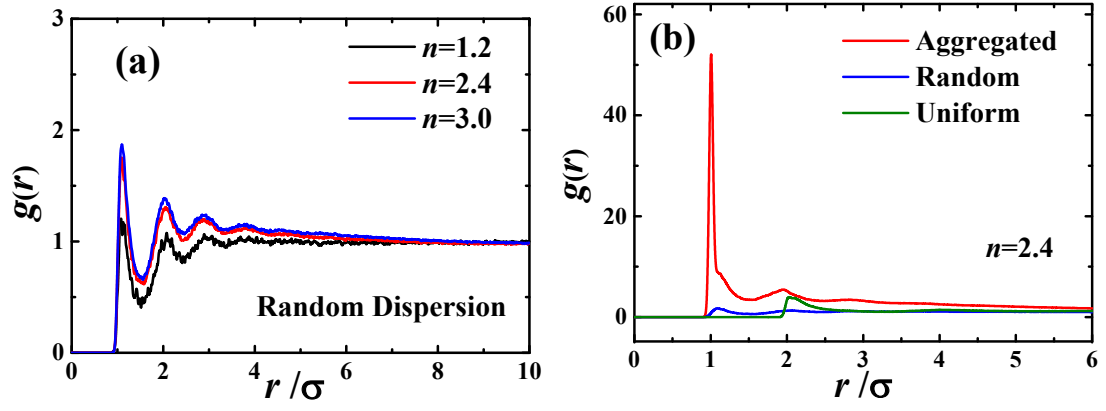
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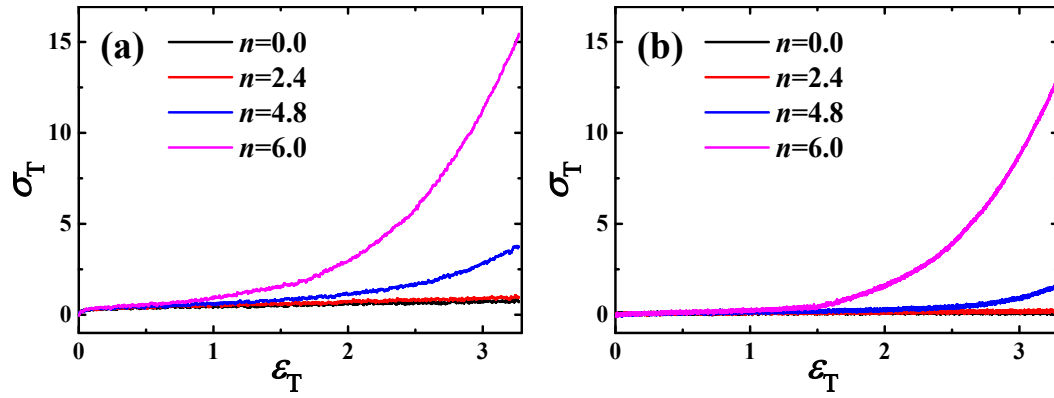
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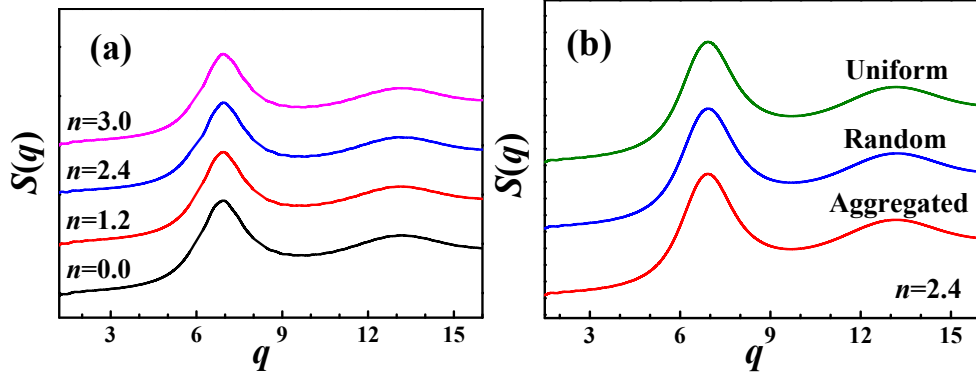
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**Figure S1.** The radial distribution functions of crosslinkers with (a) different crosslink densities and (b) different crosslink distributions.



**Figure S2.** Comparison of the tensile stress-strain curves at the tensile rate of (a)  $0.0327/\tau$  and (b)  $0.00327/\tau$

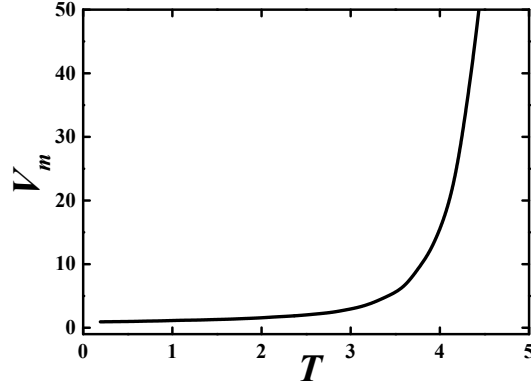


**Figure S3.** The static structure factor  $S(q)$  of polymer chains a set of crosslink densities and crosslink distributions. For visual clarity, curves for higher crosslink densities and dispersion state are shifted upwards.

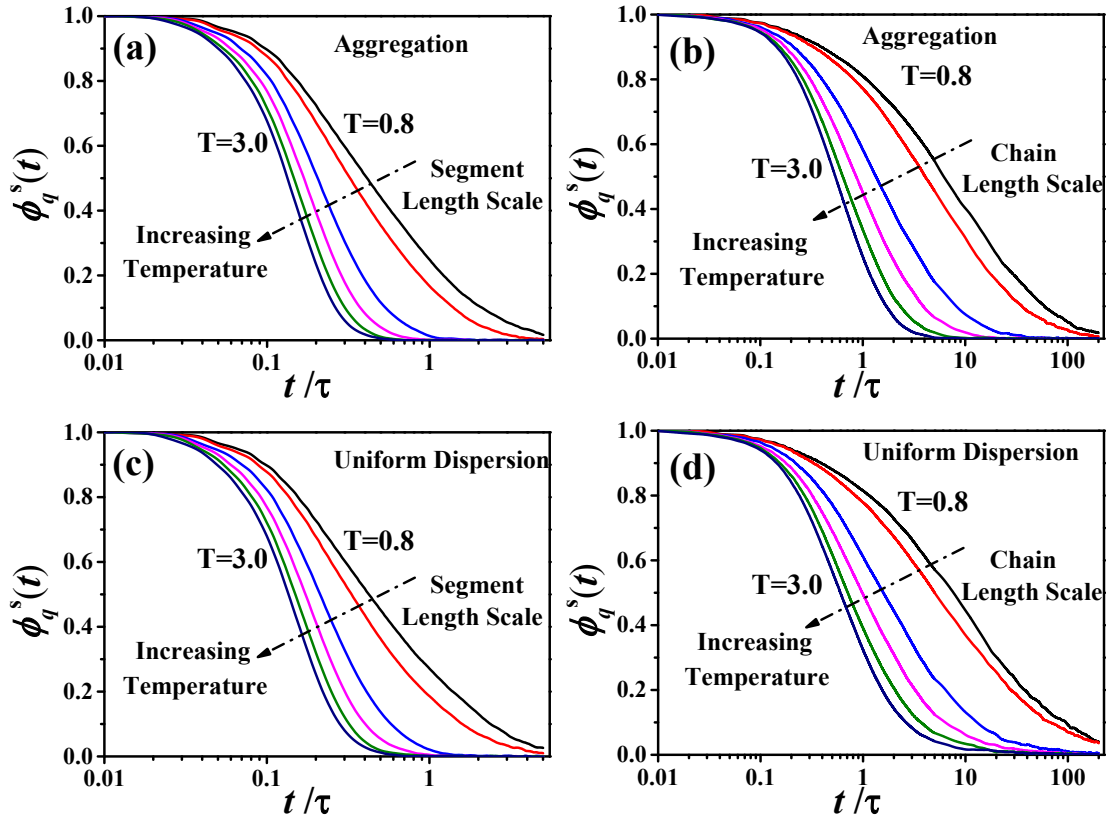
The static property of the elastomeric polymer is investigated by the static structure factor  $S(q)$ , which is calculated through the Fourier transform of the radial distribution function of polymer segments  $g_{pp}(r)$ , given by

$$S(q) = 1 + \rho \int_V \exp(-i\mathbf{q}\mathbf{r}) g_{pp}(r) d\mathbf{r} = 1 + \rho \int_0^\infty 4\pi r^2 \frac{\sin(qr)}{qr} [g_{pp}(r) - 1] dr$$

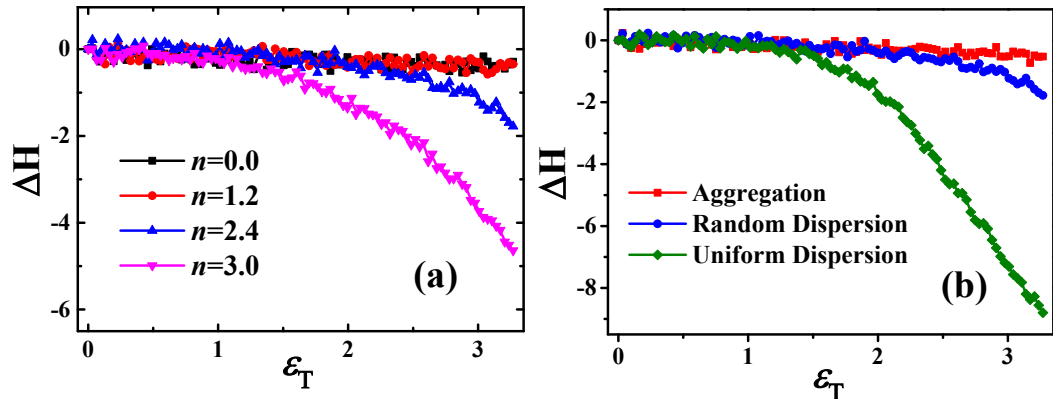
where  $\mathbf{q}$  is the scatter wave vector,  $\rho$  is the overall number density of the system, and  $r$  stands for the inter-particle distance. As the polymer chains are isotropic at equilibrium, the structure factor depends only on the absolute magnitude of the scattering vector  $q = |\mathbf{q}|$ . It is seen from Fig. S3 that the static structure of polymer chains is almost independent of the crosslink density and distribution in the range simulated. The amorphous structure is demonstrated by the amorphous halo around  $q = 6.9$ , corresponding to the nearest neighbor packing of polymer segments. In the low- $q$  regime, as the system is probed over large length scales, the structure factor  $S(q)$  is the order of a few percent or less, reflecting the low isothermal compressibility of rubbery polymer. At higher  $q$ , the structure factor  $S(q)$  oscillates and gradually approaches to 1.



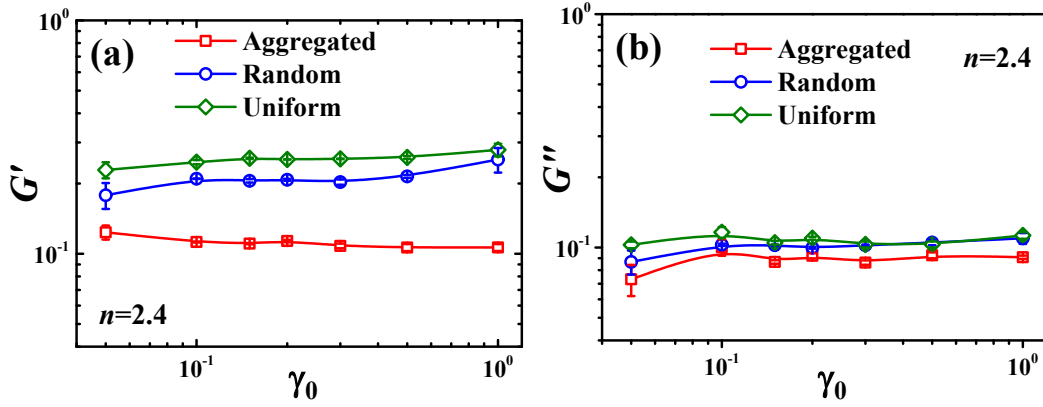
**Figure S4.** Plot of the specific volume for the uncrosslinked polymer with respect to the temperature.



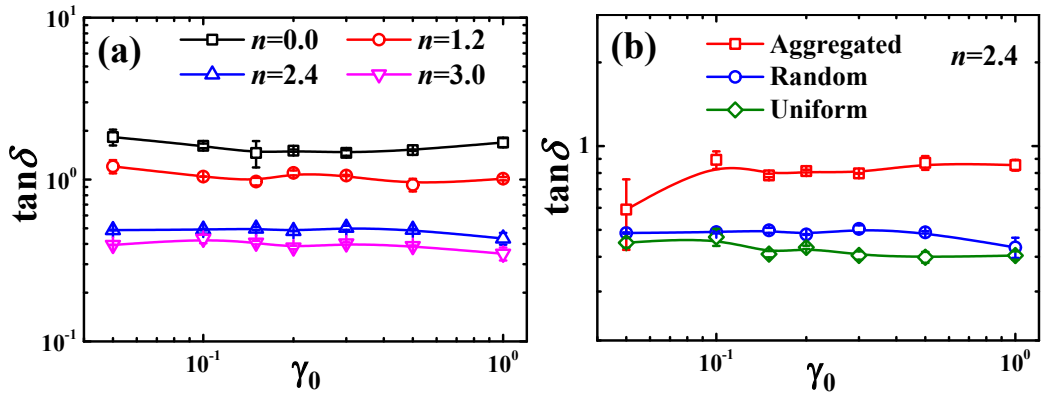
**Figure S5.** The decay of  $\phi_q^s(t)$  for the aggregated dispersion system and the uniform dispersion system, with the crosslink density equal to  $n = 2.4$ , at different temperatures of  $T=0.8, 1.0, 1.5, 2.0, 2.5$ , and  $3.0$ , respectively. (a)(c) The decay of  $\phi_q^s(t)$  at the segment length scale with  $q = 6.9$  and (b)(d) the decay of  $\phi_q^s(t)$  at the chain length scale with  $q = 2.0$ .



**Figure S6.** The change of system enthalpy  $\Delta H$  against the tensile strain.



**Figure S7.** (a)(b) The loss modulus  $G''$  plotted against the shear amplitude.



**Figure S8.** The loss tangent  $\tan \delta$  plotted against the shear amplitude.