Supporting Information for:

The Effect of Intra-chain Crosslinking on the Thermo-mechanical Behavior of Bulk Polymers Assembled Solely from Single Chain Polymer Nanoparticles

Suwon Bae,[†] Or Galant,[‡] Charles E. Diesendruck,[‡] and Meredith N. Silberstein*,[†]

[†]*Sibley School of Mechanical and Aerospace Engineering, Cornell University, NY 14850 United States*

[‡]*Schulich Faculty of Chemistry and Russell-Berrie Nanotechnology Institute, Technion - Israel Institute of Technology, Haifa, Israel*

*Phone:(+1) 607-255-5063, E-mail:meredith.silberstein@cornell.edu

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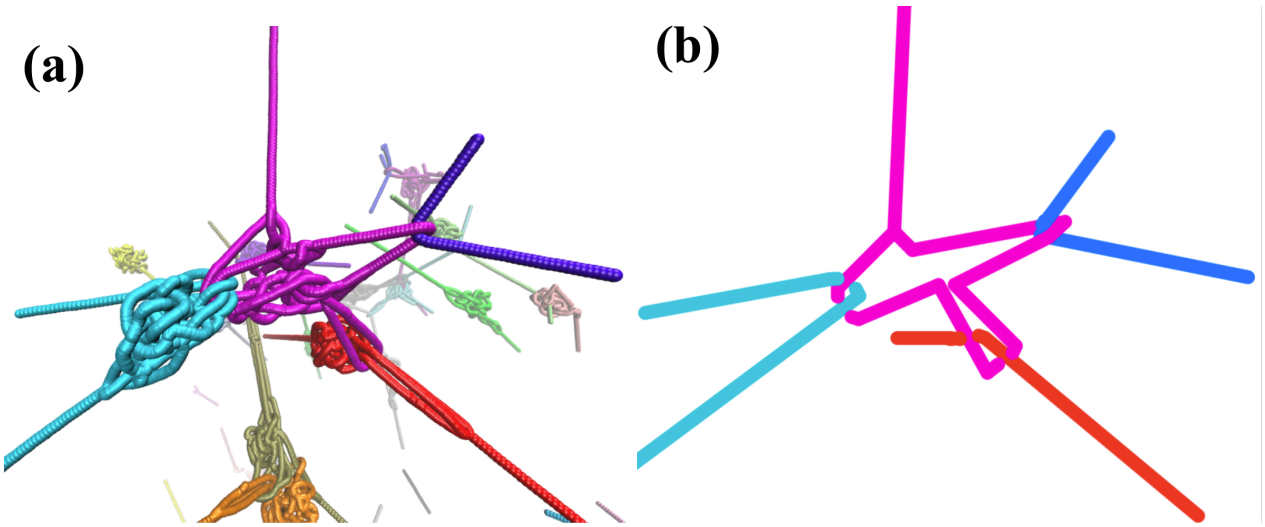

Figure S1: Enlarged view of a primitive path network and simplified primitive paths.

Due to the internal crosslinks, internal segments constructing an individual SCPN may not become taut even in this annealed network (see Figure S1a for example). Summing up the length of all segments constructing a primitive path overestimates the contour path of the primitive path. To avoid counting the length of segments going back and forth and overlapping to each other, we simplified the complex primitive path by introducing a grid (see Figure S1b for example). We created a fine three-dimensional grid for each primitive path and counted the number of grid points on which segments fall. This grid approach allowed us to avoid overestimating the length of a primitive path.

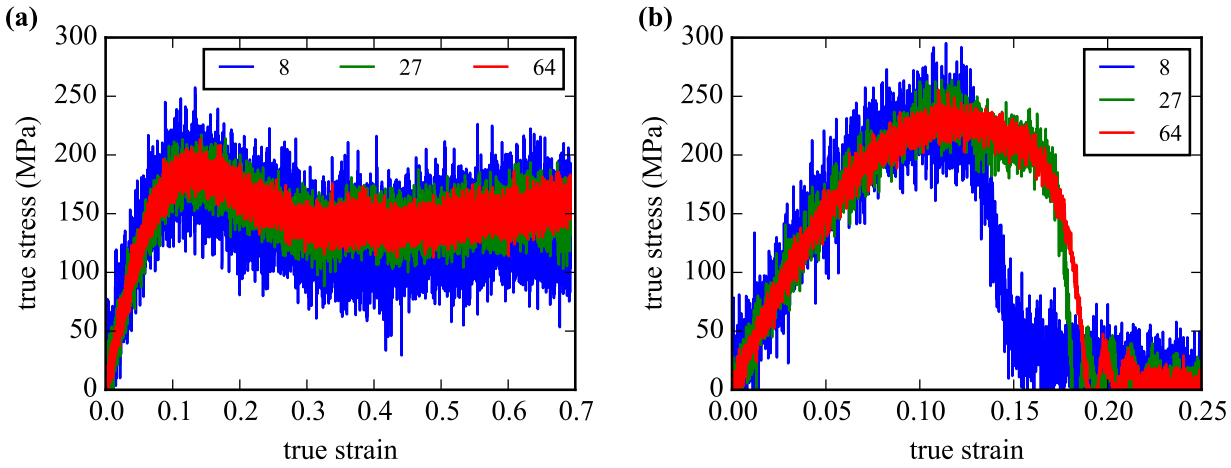

Figure S2: Stress-strain response of the bulk polymer assembled from (a) linear chains and (b) SCPNs with 10% crosslinking ratio at a temperature of 150 K and an engineering strain rate of 10^9 s^{-1} as a function of the number of SCPNs in a periodic simulation cell, showing that the response curve converges.

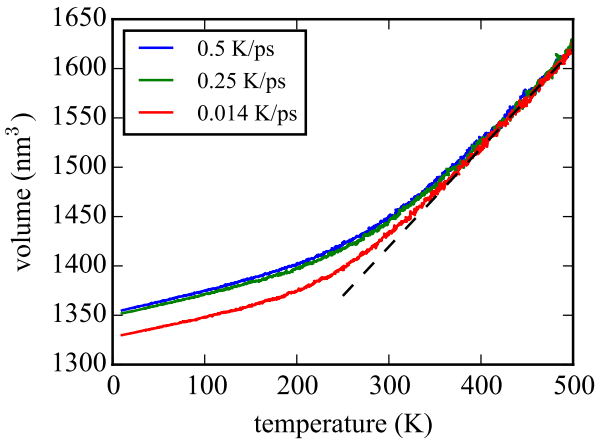

Figure S3: Volume change as a function of cooling rate. The dashed line corresponds to a fitted straight line showing at the rubbery state the volume change does not depend on cooling rate.

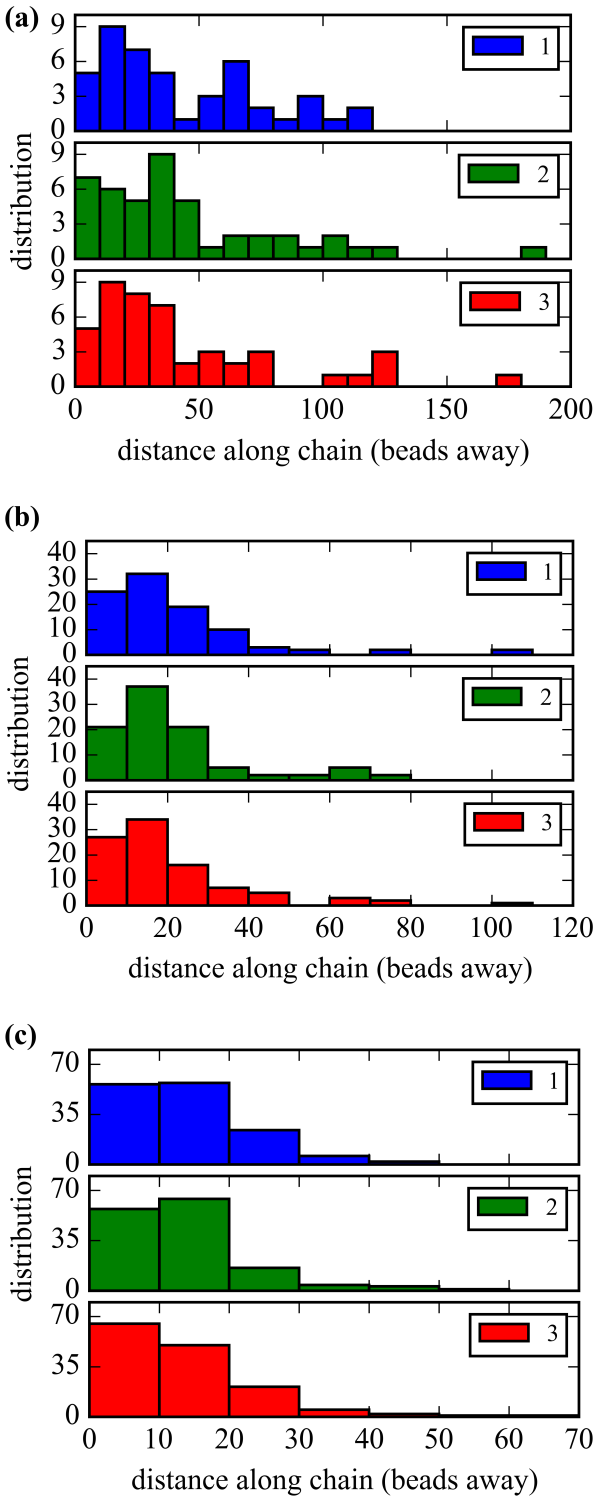

Figure S4: Distribution of distance between adjacent crosslink sites along the backbone of SCPNs for three different identically processed models. (a) 5%, (b) 10% and (c) 15% crosslinking ratios.

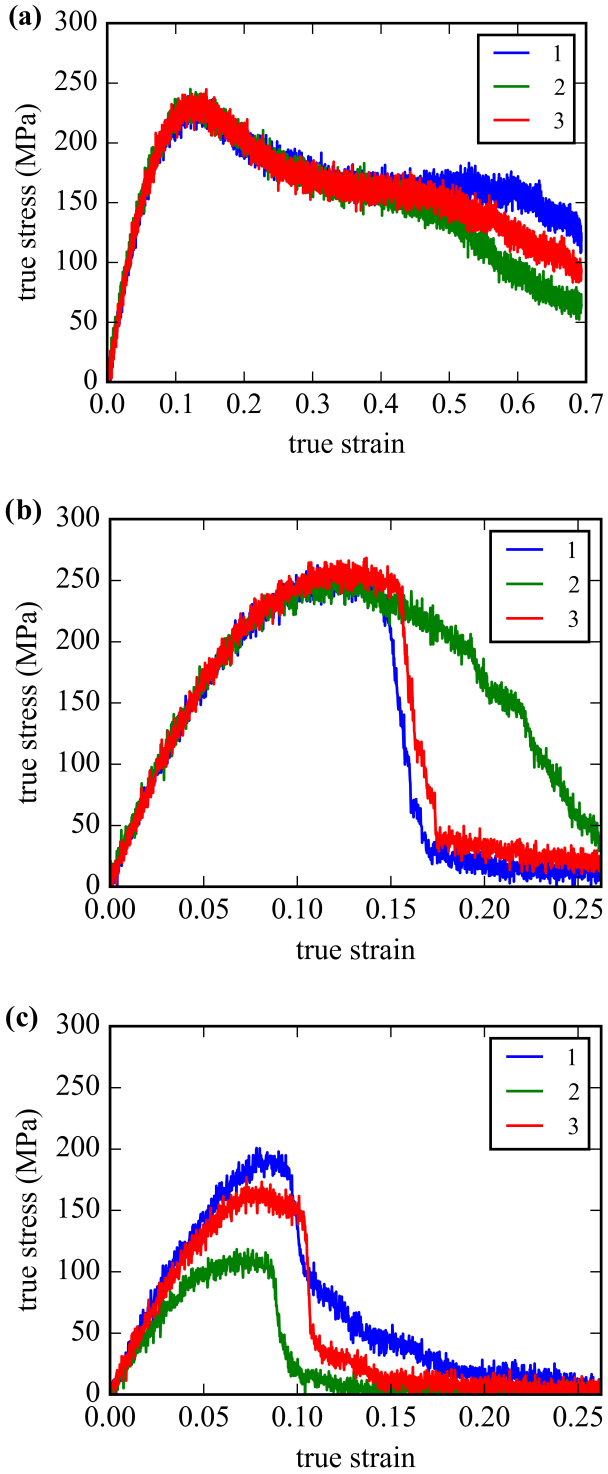

Figure S5: Stress-strain response to uniaxial tension of the bulk polymer for three different identically processed models at a temperature of 150 K and at an engineering strain rate of 10^8 s^{-1}. The bulk polymer is assembled from SCPNs with (a) 5%, (b) 10% and (c) 15% crosslinking ratios.

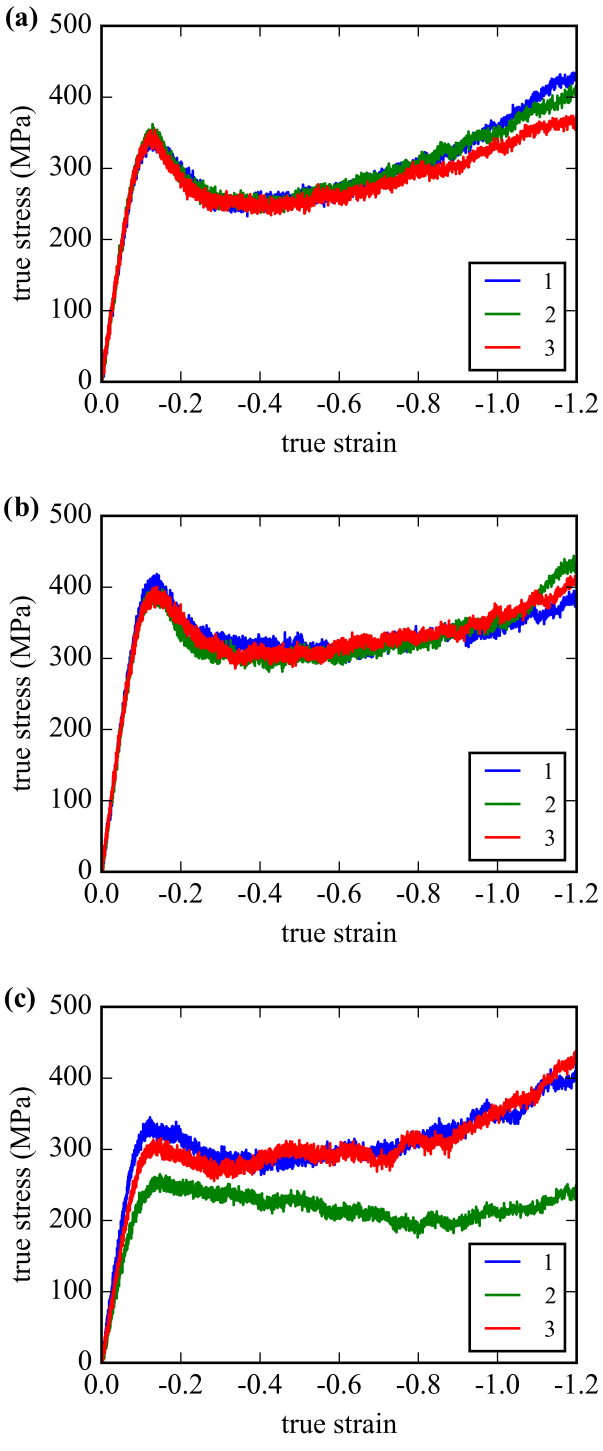

Figure S6: Stress-strain response to uniaxial compression of the bulk polymer for three different identically processed models at a temperature of 150 K and an engineering strain rate of 10^8 s^{-1}. The bulk polymer is assembled from SCPNs with (a) 5%, (b) 10% and (c) 15% crosslinking ratios.

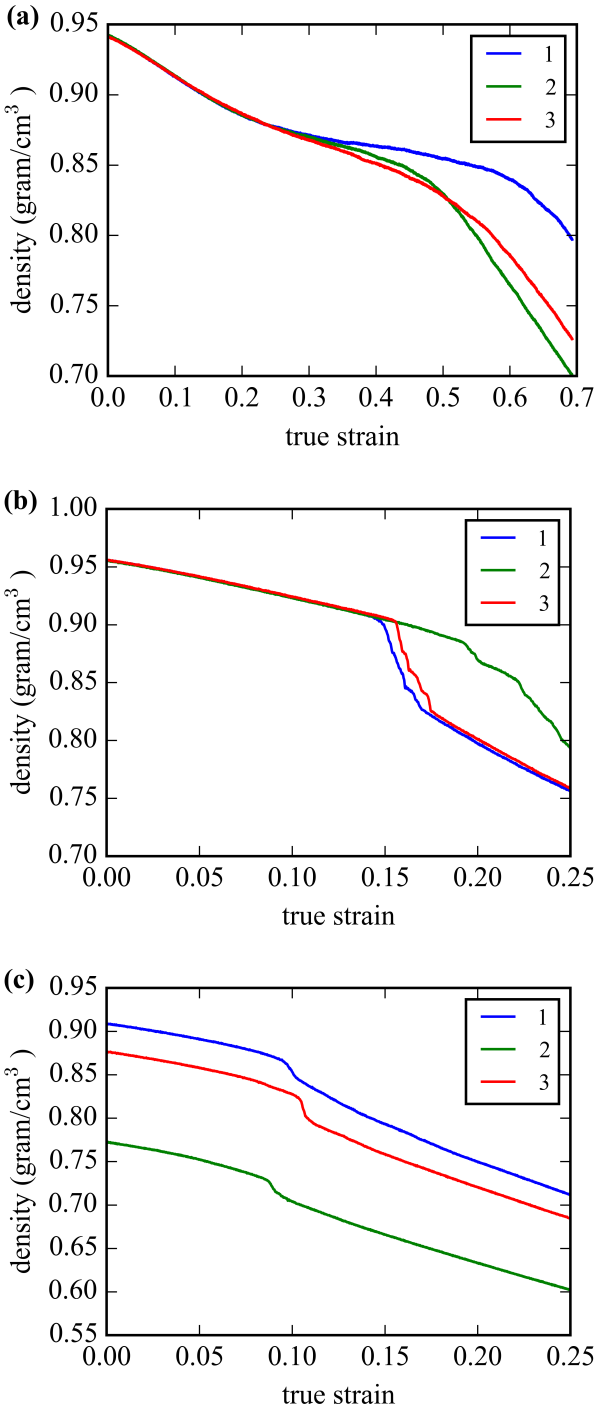

Figure S7: Density change of a bulk polymer during uniaxial tension for three different identically processed models at a temperature of 150 K and an engineering strain rate of 10^8 s^{-1}. (a) 5%, (b) 10% and (c) 15% crosslinking ratios.

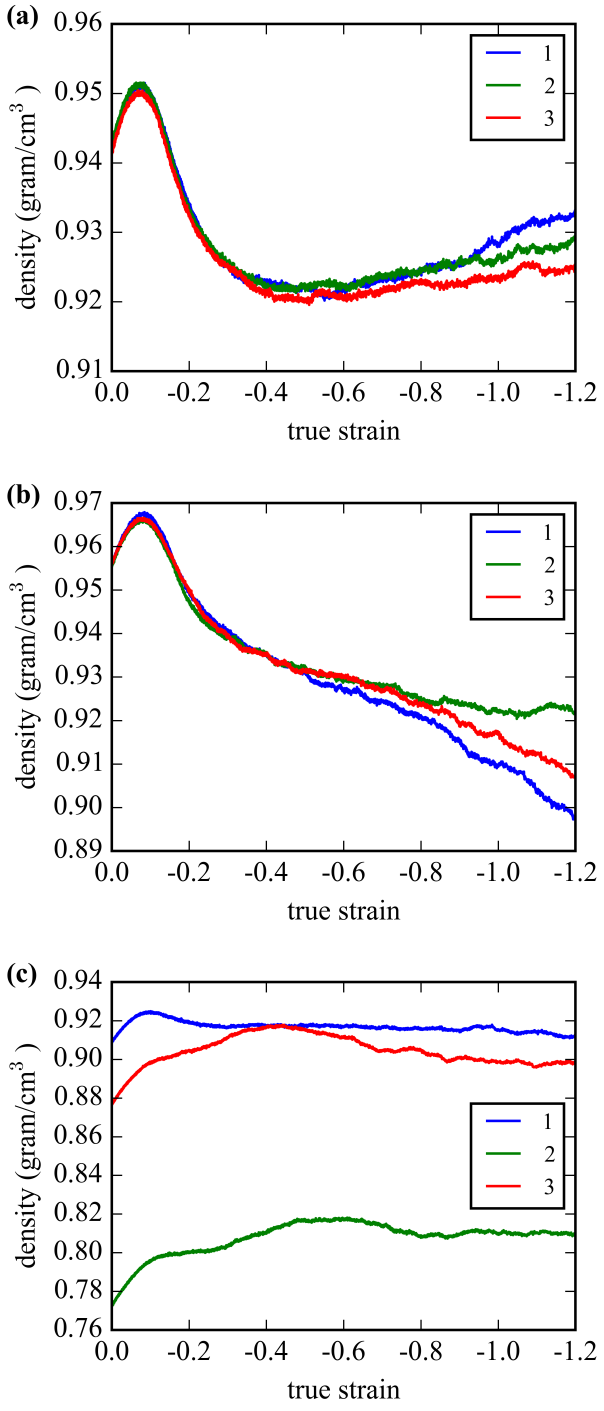

Figure S8: Density change of a bulk polymer during uniaxial compression for three different identically processed models at a temperature of 150 K and an engineering strain rate of 10^8 s^{-1}. (a) 5%, (b) 10% and (c) 15% crosslinking ratios.

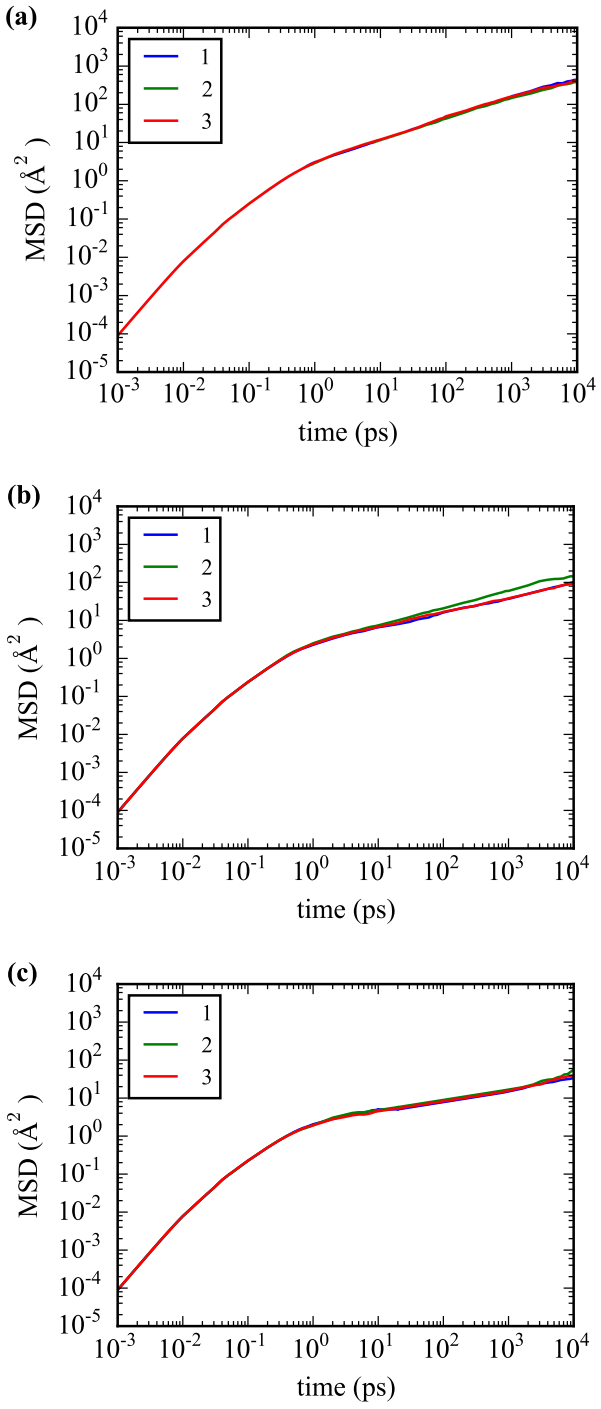

Figure S9: Mean square displacements (MSD) of all beads of the bulk polymers assembled from SCPNs for three different identically processed models. (a) 5%, (b) 10% and (c) 15% crosslinking ratios.