Supplementary Materials

Temperature-Responsive Morphology Formation of PS-*b*-PI copolymer: A Dissipative Particle Dynamics Simulation Study

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The size of our models is systemically varied from 10 to 100 monomers forming an amorphous structure in a cubic simulation box in order to check the values of the minimum chain length of each polymer representing the polymeric chains. An experimental density of 1.05 and 0.90 $g \cdot cm^{-3}$ was initially set for constructing of the PS and PI models, respectively. The initial structure was optimized using the conjugate gradient method with a convergence threshold of 0.01 $kcal \cdot mol^{-1} \cdot Å^{-1}$ and an energy convergence of 1.0×10^{-6} $kcal \cdot mol^{-1}$. The optimized amorphous structure was then refined by running an MD in the NVT ensemble for 200 ps. A typically small time step of 1 fs was used for the MD simulation to ensure the stability of simulation. The temperature of the system during the MD simulation was controlled by the Anderson thermostat. Then, the model was annealed at temperatures fluctuating from its glass transition temperature and the melting point, i.e., temperature ranges of 383 K to 433 K and 225 K to 285 K were used in the annealing process of PS and PI systems, respectively. An annealing temperature step of 10 K was used for 25 heating-cooling cycles. Then, the structures were further refined by performing NPT-MD simulations for 200 ps. Anderson thermostat with a collision ratio constant of 1.0 and Barendsen barostat with a decay constant of 0.1 ps were applied to control the temperature and pressure of the systems, respectively. We have repeated the annealing-refining process for four times. Finally, NVT-MD simulations were performed to attain the most stable structure. In this work, the diblock copolymer of PS and PI consisted of 138 and 375 units was selected regarding to the available experimental results. The solubility parameters of pure PS, pure PI, and mixed systems at different temperatures were analyzed from the production runs. These values were further used to calculate the χ parameter.

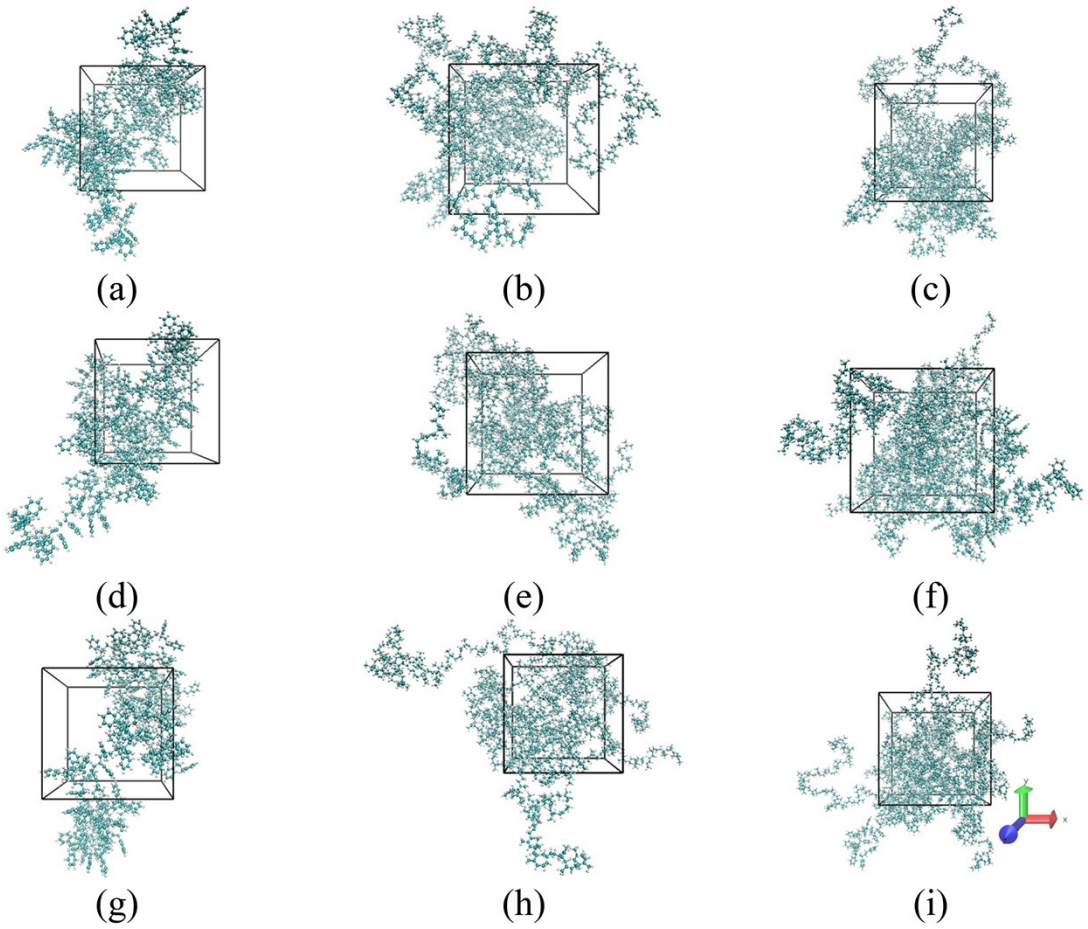

Figure S1. Amorphous unit cells of (a) PS, (b) PI and (c) mixing polymers at 393 K, and (d), (e), and (f) at 493 K, and (g), (h), and (i) at 533 K. The colors are represented carbon and hydrogen atoms by using cyan and grey, respectively. These structures were produced using TopoTools which is implemented in VMD developed by Axel Kohlmeyer (Axel Kohlmeyer, (2016). TopoTools: Release 1.7).

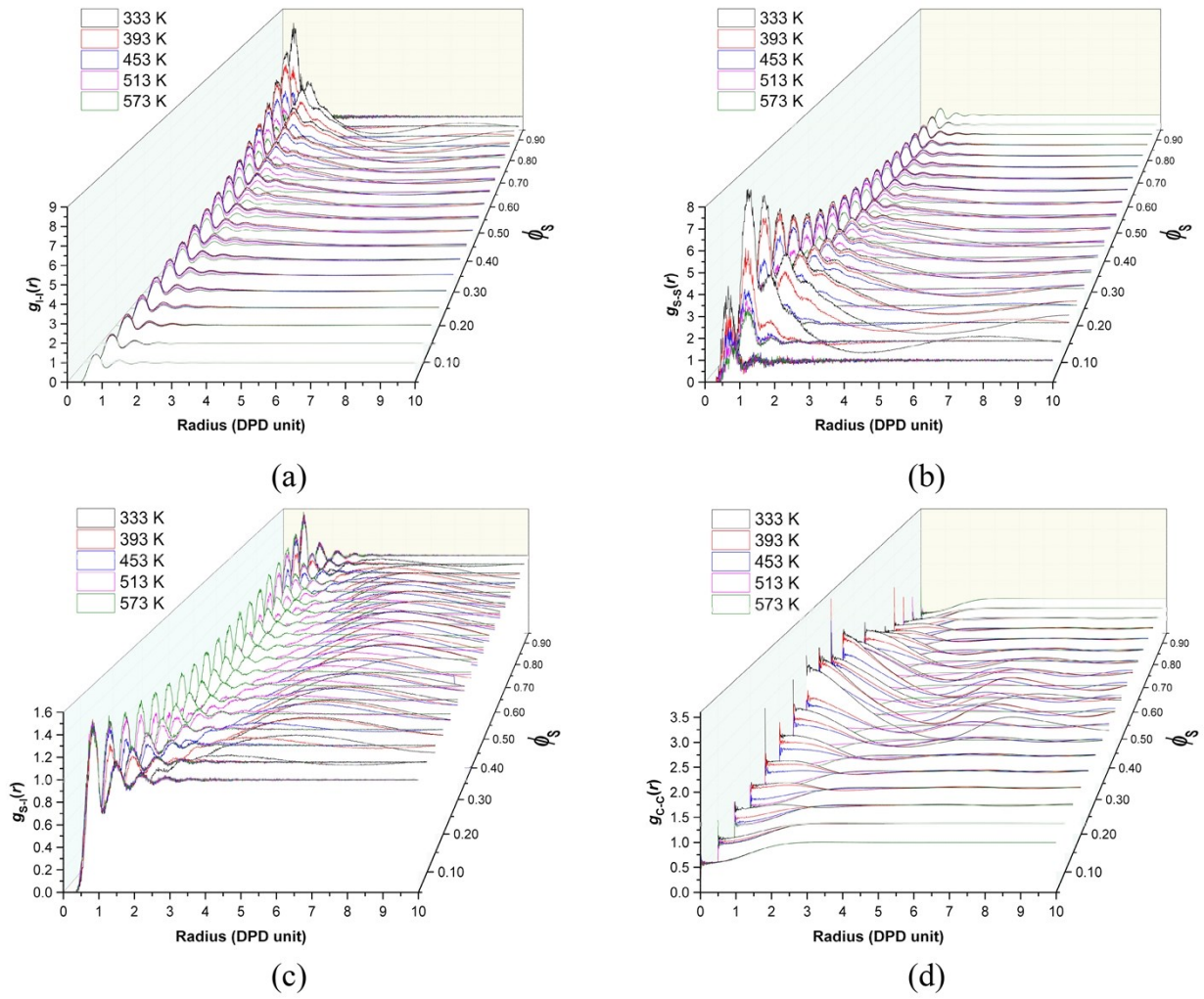

Figure S2. Dependence of RDFs $g(r)$ for all kinds of bead pairs of extended chains in Model B: (a) isoprene-isoprene, (b) styrene-styrene, (c) styrene-isoprene and (d) chain-chain on temperature and volume fraction of styrene. r is plotted in the reduced DPD length unit.

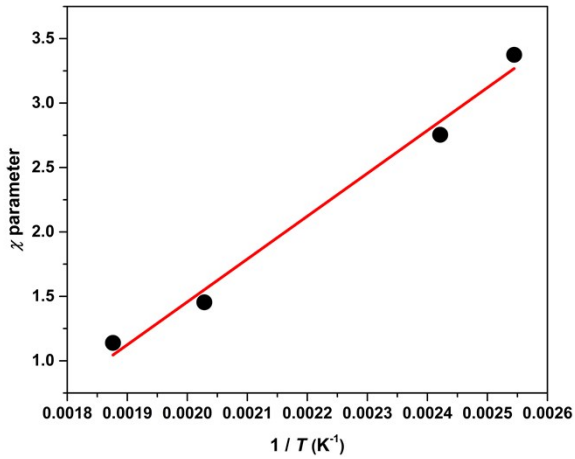

Figure S3. χ parameter *vs.* transpose of temperature in per Kelvin (K^{-1}) unit. The χ parameter of 2.754 was taken previous article reported by Xuejin *et al.* (X. Li, J. Guo, Y. Liu, and H. Liang, *J. Chem. Phys.,* 2009, **130**, 074908) and the value was illustrated in the graph as the third cycle.

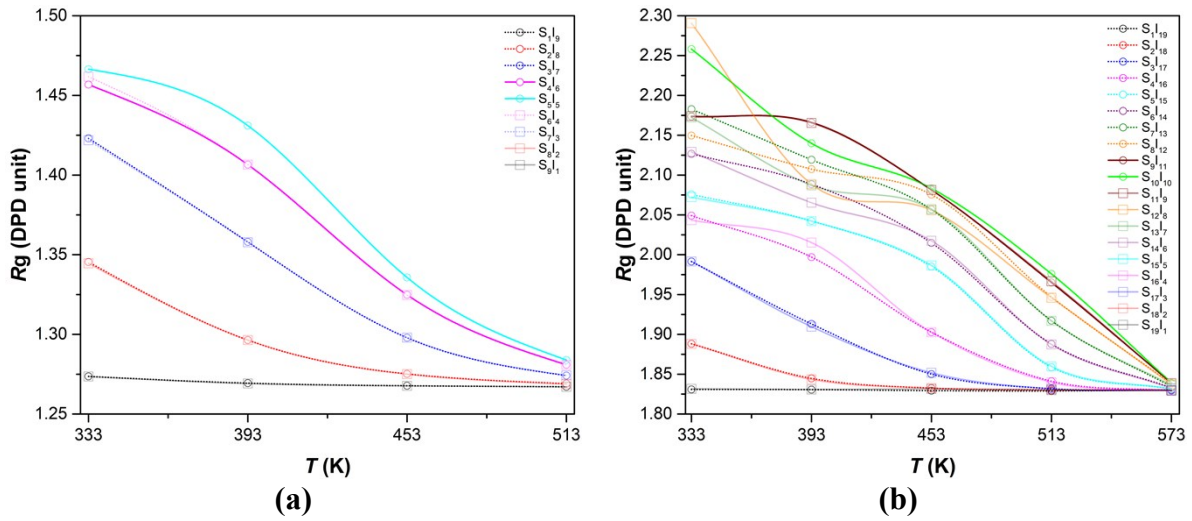

Figure S4. Radius of gyration (R_g) as a function of temperature of (a) finite chains in Model A and (b) extended chains in Model B.

Table S1. Parameters used/obtained in MD simulations.

Parameter	393 K			493 K			533 K		
	Styrene	Isoprene	Mix	Styrene	Isoprene	Mix	Styrene	Isoprene	Mix
Chain per simulation box	2	5	7	2	5	7	2	5	7
No. of repeating units per chain	69	75	*	69	75	*	69	75	*
Final cell parameter (Å)	29.14	37.66	42.50	29.19	38.74	3.47	29.56	38.74	43.99
Final density ($g \cdot mol^{-1}$)	0.9652	0.7944	0.8636	0.9603	0.7296	0.8069	0.9243	0.7301	0.7790
Total simulation time (ns)	71 ns (annealing process of 70 ns and 1 ns production dynamics)								

* Number of repeating units are 69 and 75 units for styrene and isoprene chains, respectively.

Table S2. Parameters used in DPD simulation in dimensionless unit.

Parameter	
Particle number, N	24,000
Simulation box volume	20^3
Coarse-grained degree, N_m	5
Spring constant, C_{ij}	4
Thermal energy, k_BT	1
Repulsion parameter of the same beads, a_{ii}	131.50
Time step, Δt	0.01
Equilibrium step number	1×10^6
Production step number	1×10^7