Electronic Supplementary Information

Effects of modified silica on the interfacial behaviour of polypropylene/ammonium polyphosphate composites: A molecular dynamics study

Xinran Dai, [ab] Yueying Sun, [ab] Shuai Li, [ab] Yanchen Li, [ab] Xiaolong Tang, [ab] Chunsheng Li*[b] and Jitao Liu*[ab]

a. Shandong Provincial Key Laboratory of Extreme Environment-tolerant Specialty Chemicals, School of Chemistry and Chemical Engineering, University of Jinan, Jinan 250022, China. E-mail: chm_liujt@ujn.edu.cn

b. School of Chemistry and Chemical Engineering, University of Jinan, Jinan 250022, China.

* Corresponding authors.
E-mail: chm_lics@ujn.edu.cn (C.S.L)
E-mail: chm_liujt@ujn.edu.cn (J.T.L).

Table S1 Lennard-Jones potential parameters for all atoms in MD simulation.

Model	Particles	Description	σ (Å)	ε (kcal/mol)
PP	H	hydrogen, bonded to carbon	2.9950	0.0200
	C	carbon, SP^3, bonded to 1 H, 3 heavy atoms	4.0100	0.0540
	C	carbon, SP^3, bonded to 2 H's, 2 heavy atoms	4.0100	0.0540
	C	carbon, SP^3, in methyl (CH_3) group	4.0100	0.0540
APP	H	hydrogen, bonded to oxygen	1.0980	0.0130
	H	hydrogen, charged cation	1.0980	0.0130
	N	nitrogen, SP^3 in protonated amines	3.2620	0.0650
	O	oxygen, partial double oxygen	3.5960	0.1670
	O	oxygen, SP^3, generic	3.5350	0.2400
	O	oxygen, bonded to hydrogen	3.5350	0.2400
	P	phosphorous, general	4.2950	0.2150
SiO_2	H	hydrogen, atom in terminal hydroxyl group on silicon	2.3541	0.0988
	O	oxygen, siloxane	3.3500	0.2400
	O	oxygen, between two silicons	3.4506	0.1622
	O	oxygen, in terminal hydroxyl group on silicon	3.4618	0.1591
	Si	silicon, in zeolites	3.6900	0.0043
OTES	H	hydrogen, bonded to carbon	2.9950	0.0200
	H	hydrogen, bonded to oxygen	1.0980	0.0130
	C	carbon, SP^3, bonded to 2 H's, 2 heavy atoms	4.0100	0.0540
	C	carbon, SP^3, in methyl (CH_3)	4.0100	0.0540

		group		
	Si	silicon, siloxane	4.2840	0.0700
HDTMS	H	hydrogen, bonded to carbon	2.9950	0.0200
	H	hydrogen, bonded to oxygen	1.0980	0.0130
	C	carbon, SP^3, bonded to 2 H's, 2 heavy atoms	4.0100	0.0540
	C	carbon, SP^3, in methyl (CH_3) group	4.0100	0.0540
	Si	silicon, siloxane	4.2840	0.0700
OTS	H	hydrogen, bonded to carbon	2.9950	0.0200
	H	hydrogen, bonded to oxygen	1.0980	0.0130
	C	carbon, SP^3, bonded to 2 H's, 2 heavy atoms	4.0100	0.0540
	C	carbon, SP^3, in methyl (CH_3) group	4.0100	0.0540
	Si	silicon, siloxane	4.2840	0.0700
KH570	H	hydrogen, bonded to carbon	2.9950	0.0200
	H	hydrogen, bonded to oxygen	1.0980	0.0130
	C	carbon, SP^3, bonded to 2 H's, 2 heavy atoms	4.0100	0.0540
	C	carbon, SP^3, in methyl (CH_3) group	4.0100	0.0540
	C	carbon, non aromatic, next-to-end doubly bonded	4.0100	0.0640
	C	carbon, non aromatic end doubly bonded	3.9000	0.0640
	C	carbon, carbonyl carbon of acid, ester, amide	3.8100	0.1200
	O	oxygen, in carbonyl group	3.3000	0.2670

	O	oxygen, ester	3.4200	0.2400
	Si	silicon, siloxane	4.2840	0.0700

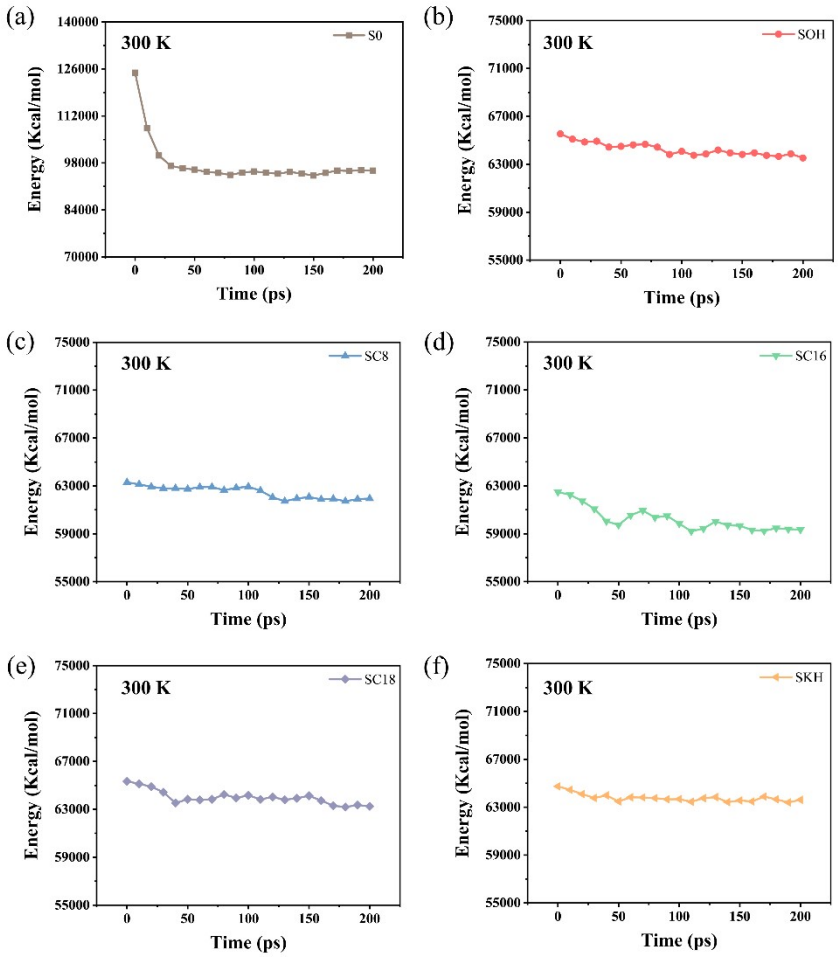

Fig. S1 Total Energy versus Time Curves Demonstrating System Stability at 300 K. (a) S0, (b) SOH, (c) SC8, (d) SC16, (e) SC18, and (f) SKH.

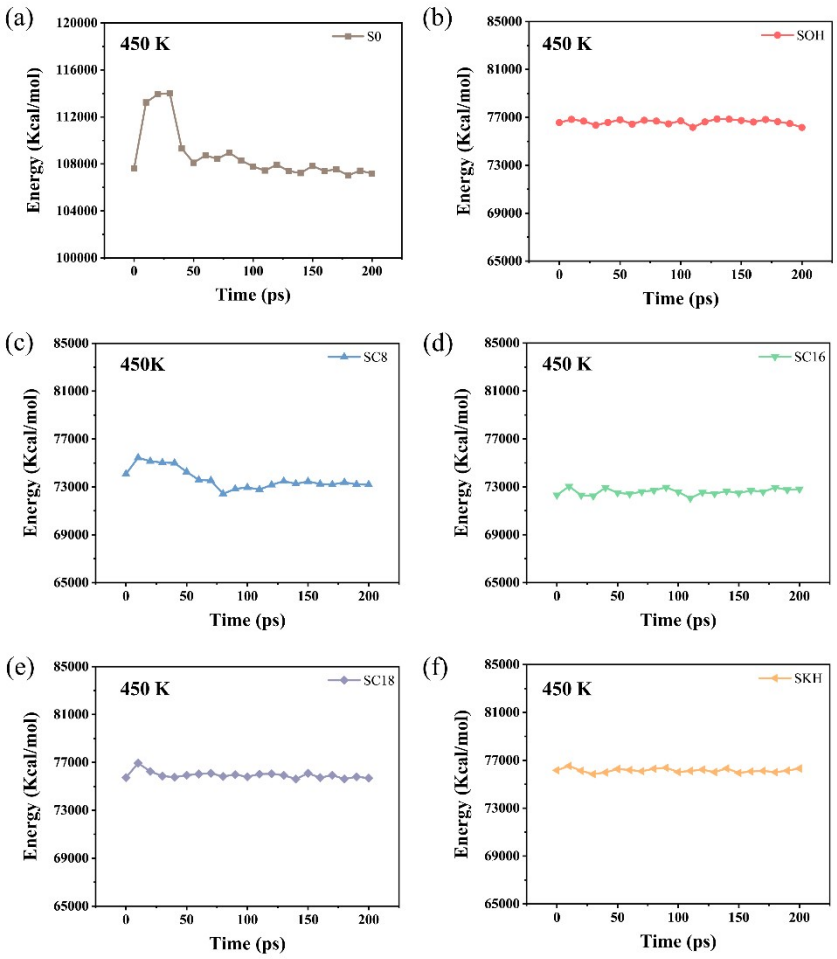

Fig. S2 Total Energy versus Time Curves Demonstrating System Stability at 450 K. (a) S0, (b) SOH, (c) SC8, (d) SC16, (e) SC18, and (f) SKH.

During the subsequent isothermal relaxation at 450 K following the heating phase, the total energy of all systems first shows a brief increase, then decreases, and finally stabilizes. This behavior can be explained by considering the system's approach to equilibrium. Although the gradual heating adjusts the velocity distribution toward that of the target temperature, the atomic configuration initially remains out of equilibrium with the new thermal state. When the simulation switches to constant-temperature conditions, the rapid equilibration of these lagging configurational degrees of freedom causes a short-term rise in energy. Afterwards, the system evolves toward a more stable potential-energy surface through structural rearrangement, which leads to an energy decrease until true thermal equilibrium is attained.

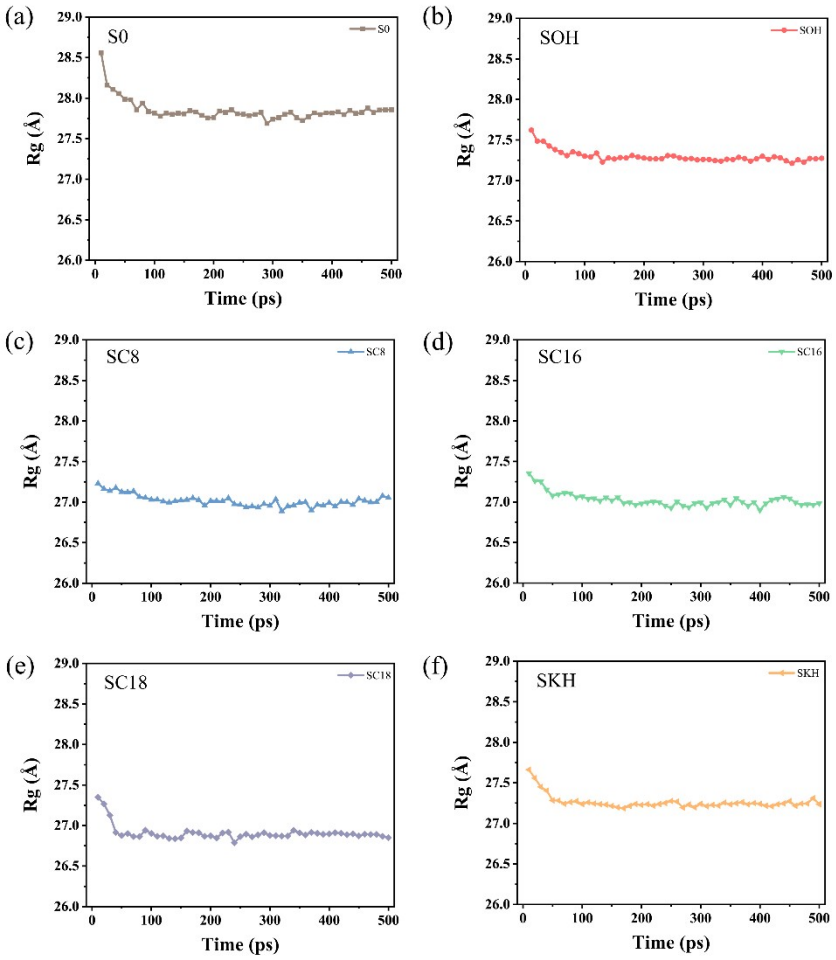

Fig. S3 The convergence profiles of the Rg during the 500 ps equilibration phase for the different systems.

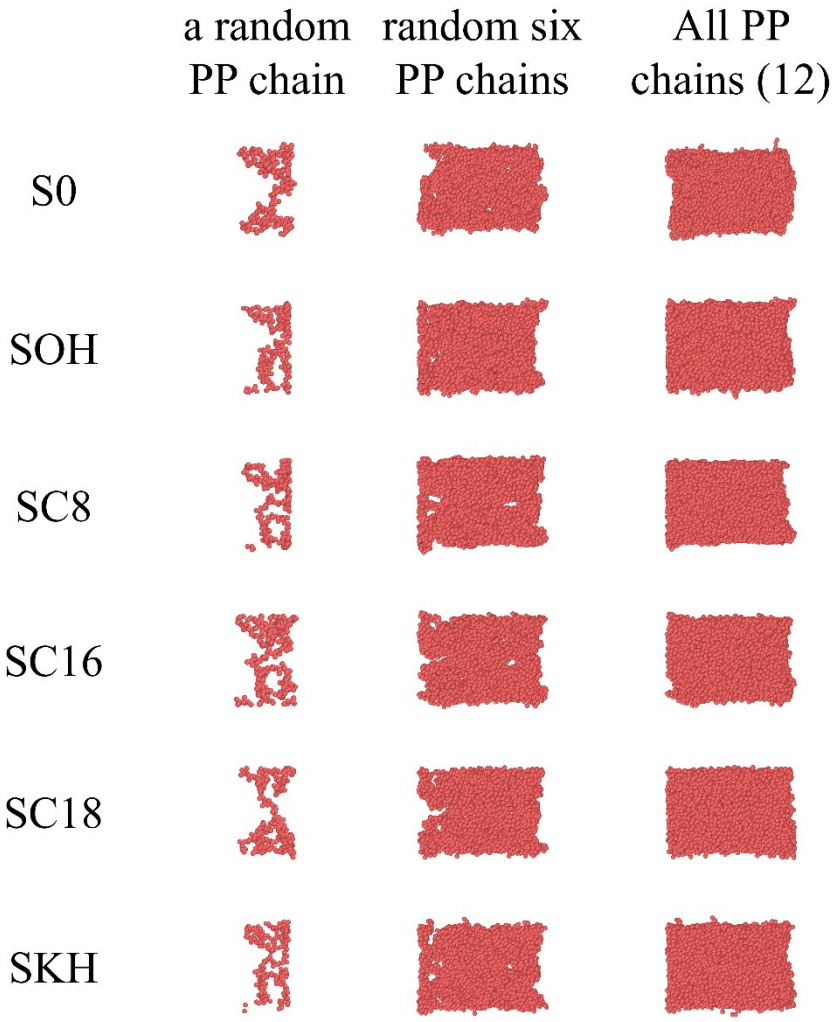

Fig. S4 Conformational snapshots from the equilibrated states of the different systems, showing (from left to right) a single chain (ID 903), six superimposed chains (IDs 1, 903, 9021, 1805, 8119, 7217), and all twelve superimposed chains.

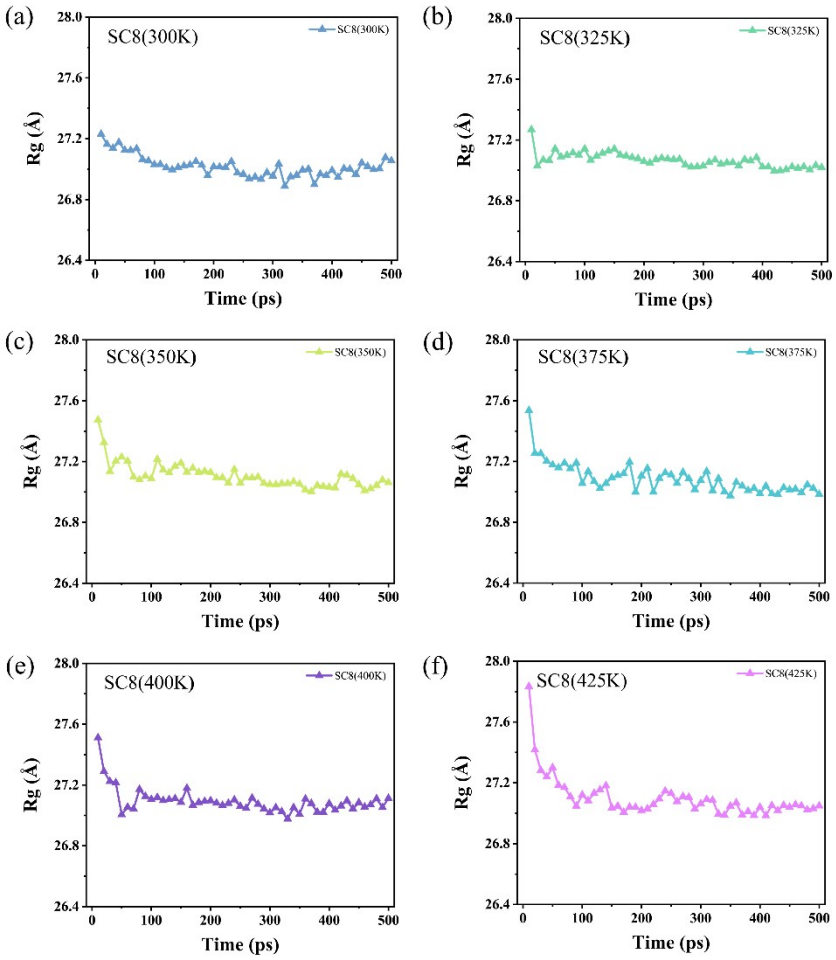

Fig. S5 The convergence profiles of the Rg during the 500 ps equilibration phase for the SC8 system at different temperatures.

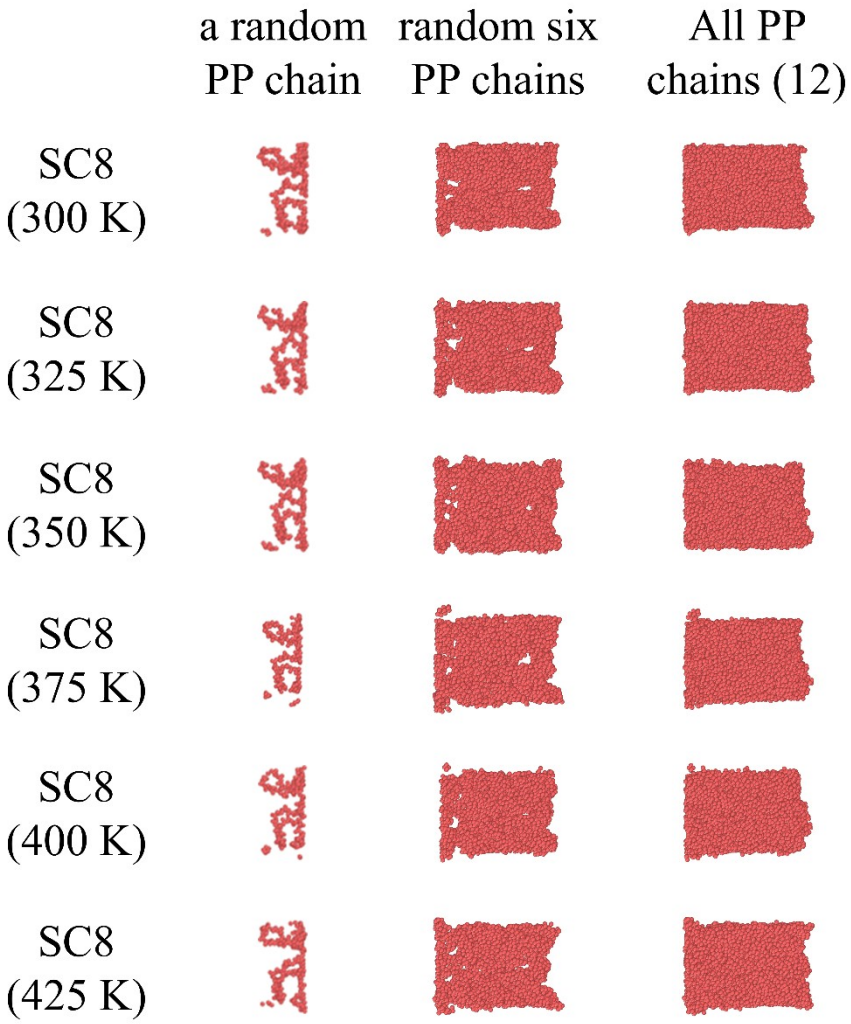

Fig. S6 Conformational snapshots from the equilibrated states of the SC8 system at different temperatures, showing (from left to right) a single chain (ID 903), six superimposed chains (IDs 1, 903, 9021, 1805, 8119, 7217), and all twelve superimposed chains.