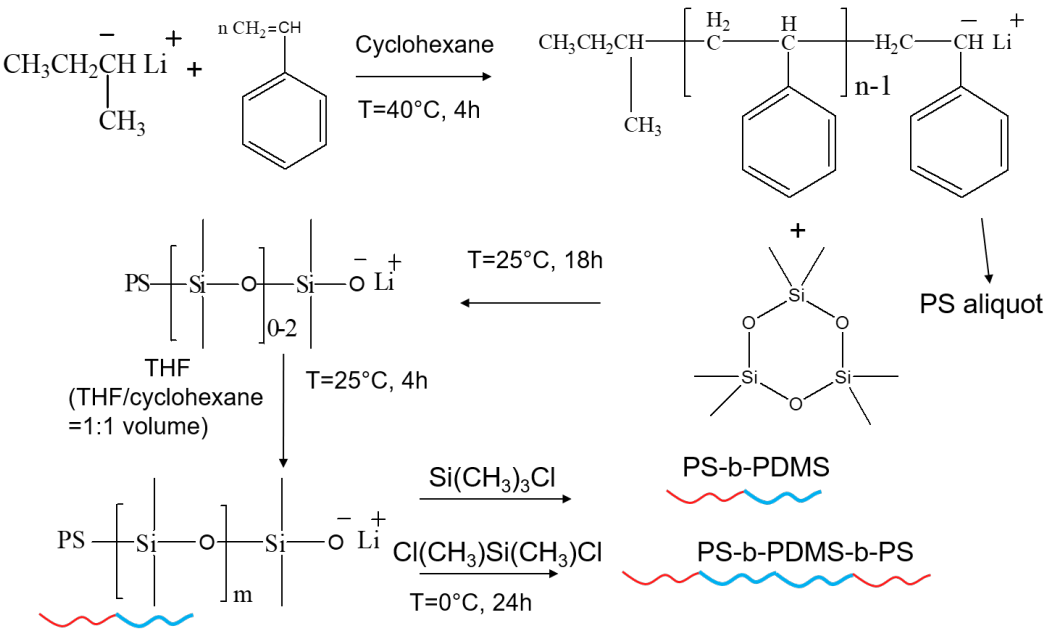
Supporting Information: Easy-processable and Aging-free All-polymer Polysiloxane Composites

Liyang Shen,[†] Tung-ping Wang,[†] Shailja Goyal,[†] Ting-Han Lee,[†] Fang-Yi Lin,[†] Sabrina Torres,[‡] Thomas Robison,[‡] and Eric W. Cochran[*,†]

†Department of Chemical and Biological Engineering, Iowa State University, Ames, Iowa 50011, United States

‡Kansas City National Security Campus, 14520 Botts Road, Kansas City, Missouri 64147, United States

E-mail: ecochran@iastate.edu



Scheme S1: Synthetic route of PS precursor, PS-PDMS diblock copolymer and PS-PDMS-PS triblock copolymer

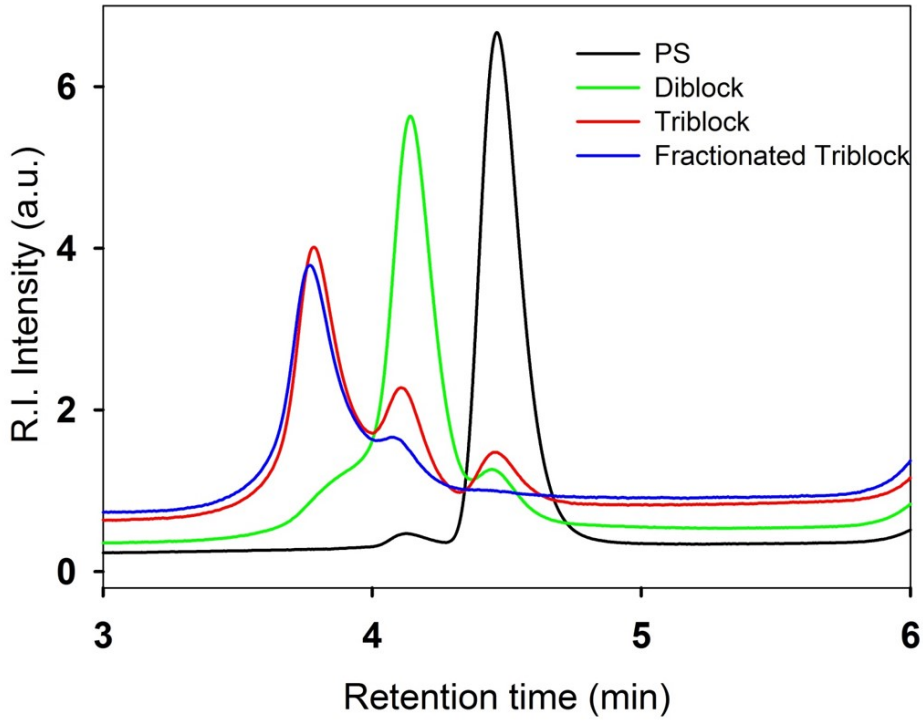


Figure S1: GPC chromatographs of PS precursor, PS–PDMS diblock copolymer and PS-PDMS-PS triblock copolymer

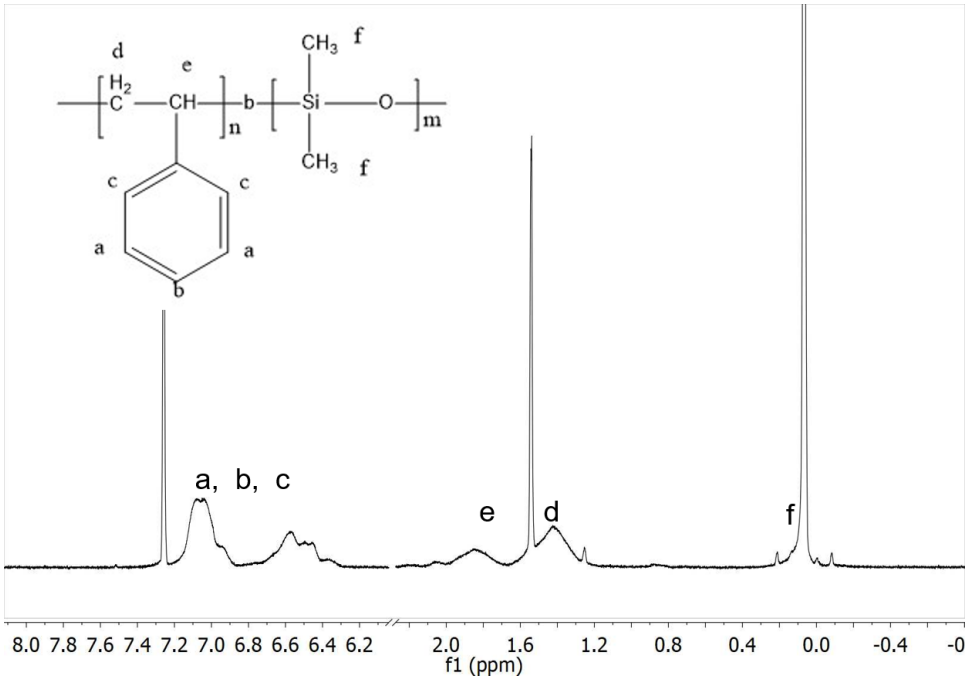


Figure S2: ^{1}H-NMR spectra of diblock copolymer PS-b-PDMS

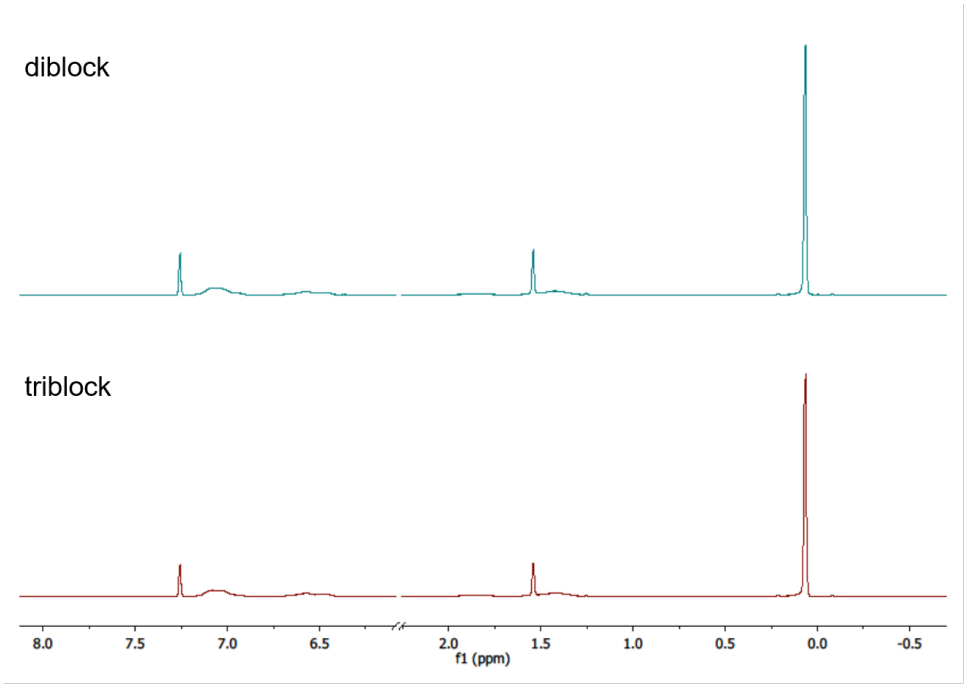
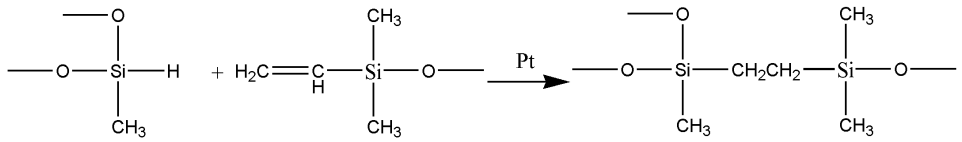


Figure S3: ^{1}H-NMR spectra of diblock copolymer and triblock copolymer



Scheme S2: Platinum catalyzed silicone crosslinking (hydrosilylation reaction)

Table S1: Summary of mechanical properties of neat PDMS, all-polymer and silica reinforced composites

Filler weight fraction	Elongation at break	Tensile strength (MPa)	Young′s Modulus (MPa)	Toughness (MJ/m^3)
50 wt% diBCP (**D50**)	1.06 ± 0.05	3.38 ± 0.39	1.40 ± 0.03	0.88 ± 0.10
34 wt% homoPS/ 15 wt% diBCP (**H34 D15**)	0.79 ± 0.08	2.19 ± 0.15	2.37 ± 0.25	0.76 ± 0.10
30 wt% diBCP/ 20 wt% triBCP (**D30 T20**)	0.65 ± 0.03	2.37 ± 0.08	3.87 ± 0.10	0.78 ± 0.06
34 wt% homoPS/ 14 wt% diBCP/ 10 wt% triBCP (**H30 D14 T10**)	0.78 ± 0.05	4.39 ± 0.61	5.14 ± 0.74	1.58 ± 0.24
30 wt% silica (**silica30**)	0.85 ± 0.10	3.60 ± 0.84	2.57 ± 0.01	1.16 ± 0.27

Young′s modulus was calculated by linear regression within the elastic region of each curve. Toughness was determined by the integrated area under engineering stress-strain curve.

Table S2: Summary of mechanical properties of all-polymer composites with various triblock copolymer content

Filler weight fraction	Elongation at break	Tensile strength (MPa)	Young′s Modulus (MPa)	Toughness (MJ/m^3)
34 wt% homoPS/ 15 wt% diBCP (**H34 D15**)	0.79 ± 0.08	2.19 ± 0.15	2.37 ± 0.25	0.76 ± 0.10
33 wt% homoPS/ 15 wt% diBCP/ 2 wt% triBCP (**H33 D15 T2**)	0.57 ± 0.03	1.87 ± 0.18	2.75 ± 0.19	0.49 ± 0.05
32 wt% homoPS/ 14 wt% diBCP/ 6 wt% triBCP (**H32 D14 T6**)	0.84 ± 0.06	4.17 ± 0.35	4.27 ± 0.36	1.57 ± 0.29
34 wt% homoPS/ 14 wt% diBCP/ 10 wt% triBCP (**H30 D14 T10**)	0.78 ± 0.05	4.39 ± 0.61	5.14 ± 0.74	1.58 ± 0.24

Young′s modulus was calculated by linear regression within the elastic region of each curve. Toughness was determined by the integrated area under engineering stress-strain curve.

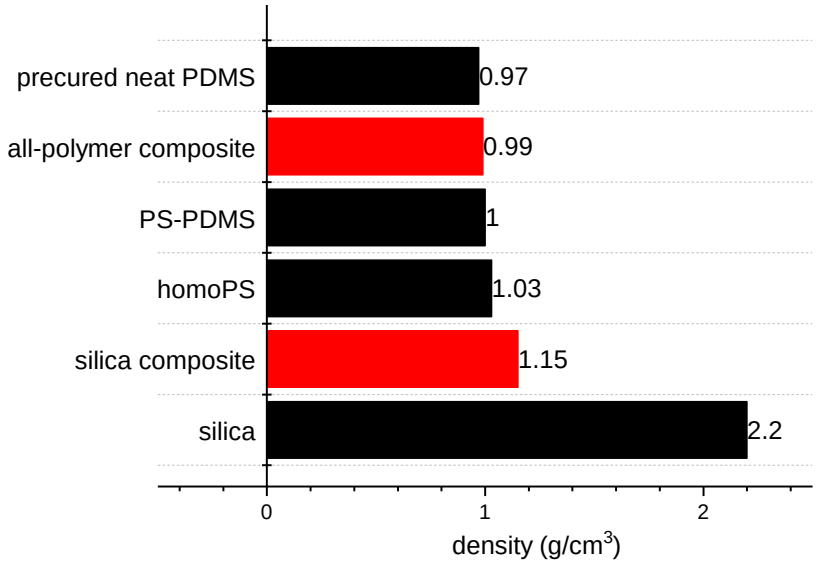


Figure S4: Density comparison of the raw materials, all-polymer (PS 32/ Di 14/ Tri 6 wt%) and silica (30 wt%) composites

The densities of PDMS and silica are from product details; the densities of the rest solid samples were measured according to ASTM D297-15.