

Supporting information

Thermo-reversible Furfuryl Poly(thioether)-*b*-Polysiloxane-*b*-Furfuryl Poly(thioether) Triblock Copolymer as the Promising Material for High Dielectric Applications

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Structure analysis.

FT-IR spectra were carried out on a Nicolet 5700 Fourier transformation infrared spectrometer (Thermo Scientific, USA) from 4000 cm^{-1} to 500 cm^{-1} . An average of about 32 scans was recorded for each measurement.

$^1\text{H-NMR}$ spectra were carried out on a Bruker Advance DMX 400 MHz spectrometer, the CDCl_3 was used as the solvent and tetramethylsilane (TMS) was used as an internal standard. The number-average molecular weight (M_n) and molecular weight distribution of the copolymer was determined by gel permeation chromatography (GPC) with a Waters 2414 refractive index (RI) detection system equipped with a Waters 1525 isocratic high-performance liquid chromatography (HPLC) pump from Waters Technologies and the experiments were carried out at 40 $^\circ\text{C}$ with tetrahydrofuran at a flow rate of 1.0 mL min^{-1} .

Electro-actuation performance.

Electro-actuation performance was conducted using a diaphragm actuator at a prestrain-free condition. The elastomer films were placed on an adhesive acrylate ring with a 10 mm inner diameter, and the ring was stuck tightly onto the top of a diaphragm chamber. A controlled bias air pressure of ~ 400 Pa was used to guide the direction of actuation strain (out of plane). The diameter of the active area was 10 mm (before the actuation and the set of bias air pressure) with two SWCNT electrodes on both sides of an elastomer film. A high-voltage power supply (Trek 610E) was used to drive the actuators. The actuation strain was calculated by analyzing the captured images by Adobe Photoshop.

Quantitative shape memory experiments.

The quantitative shape memory experiments were carried out in a TA-Q800 DMA device. The sample was stretched to 0.45 N at a rate of 0.1 N/min after reaching equilibrium at 50 $^\circ\text{C}$ for 3 min. Then the sample was rapidly cooled to about -10 $^\circ\text{C}$ and the temperature was kept for 3 min to fix the temporary shape. The force applied to the sample was then removed at a rate of 0.1 N/min until a preload of 10^{-3} N was reached. After equilibrium for about 3 min, the temperature was increased to 50 $^\circ\text{C}$ and kept for

3 min to recover the permanent shape. The fixing ratio (R_f) and recovering ratio (R_r) were used to reflect the shape memory behavior quantitatively. The R_f and R_r were calculated by Equation (1) and (2), respectively.

$$R_f = 100\% \times \frac{\varepsilon}{\varepsilon_{load}}, (1)$$

$$R_r = 100\% \times \frac{\varepsilon - \varepsilon_{rec}}{\varepsilon}, (2)$$

Where ε_{load} is the strain under load upon the cooling process, ε corresponds the strain when the load is removed before heating, ε_{rec} represents the strain when the heating process is accomplished under a stress-free condition.

Dissolution experiments.

The dissolution experiments of the PSiPGE-BMI were carried out in DMF at room temperature for 72 h and at 160 °C for about 2 h in a three-neck flask, and then, the solution was cooled to about 60 °C. The sample used in the dissolution experiment was cut into pieces and swollen in toluene for 72 h in advance to make sure that the unreacted BMI had been removed completely, and then the sample was dried in vacuum for 24h.

Table S1. The recipes for the preparation of PSiFGE-BMI with different crosslinking degree.

	PSiFGE (g)	BMI (g)
PSiFGE-BMI-3/1	2.5	0.86
PSiFGE-BMI-10/1	2.8	0.287
PSiFGE-BMI-15/1	2.9	0.204
PSiFGE-BMI-20/1	2.9	0.152
PSiFGE-BMI-30/1	3.1	0.107

Table S2. The molecular weight of the silicon alkoxide and PSiFGE.

	M_n (kg/mol)	M_w (kg/mol)	PDI
Silicon alkoxide ^a	2.48	-	-
PSiPGE ^b	9.72	18.3	1.88

a: measured by MALDI-TOF-Mass; b: measured by GPC.

Table S3. The detailed values of the mechanical and dielectric properties for the crosslinked

PSiFGE-BMI.

Sample	Tensile strength/MPa a	Elongation at break/%	Elastic modulus (Y)/MPa	ϵ'	Conductivity (10^{-10} S/cm)	$\tan \delta$	$\beta(\text{MPa}^{-1})$
PSiFGE-BMI-3/1	11.17±0.7	83.4±12	79.7	5.63	6.67	0.027	0.07
PSiFGE-BMI-5/1	3.81±0.19	85.7±18	12.6	5.68	5.96	0.048	0.45
PSiFGE-BMI-10/1	0.75±0.07	47.7±1.5	2.39	5.77	3.78	0.103	2.41
PSiFGE-BMI-15/1	0.46±0.08	43.3 ±9	1.36	5.93	2.44	0.068	4.4
PSiFGE-BMI-20/1	0.24±0.05	31±4	0.83	7.64	2.17	0.049	9.2
PSiFGE-BMI-30/1	0.16±0.04	25.6±5	0.6	7.1	1.24	0.028	11.8

Table S4. The detailed values of ϵ , ϵ_{load} , ϵ_{rec} , R_f and R_r for the consecutive shape memory cycles.

	1 st cycle	2 nd cycle	3 rd cycle
ϵ (%)	2.85	2.56	2.46
ϵ_{load} (%)	2.99	2.71	2.59
ϵ_{rec} (%)	0.27	0.1	0.052
R_f (%)	95.3	94.5	95
R_r (%)	90.5	96.1	97.9

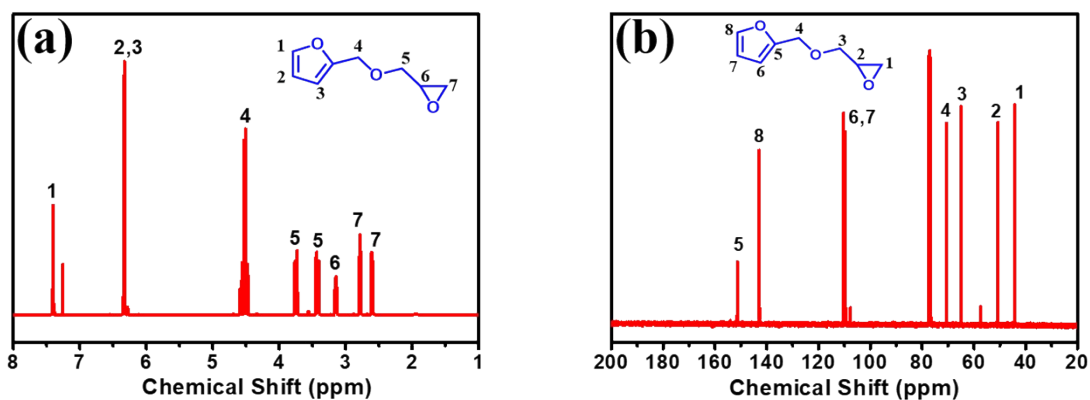


Figure S1. The ^1H -NMR (a) and ^{13}C -NMR (b) spectra of FGE.

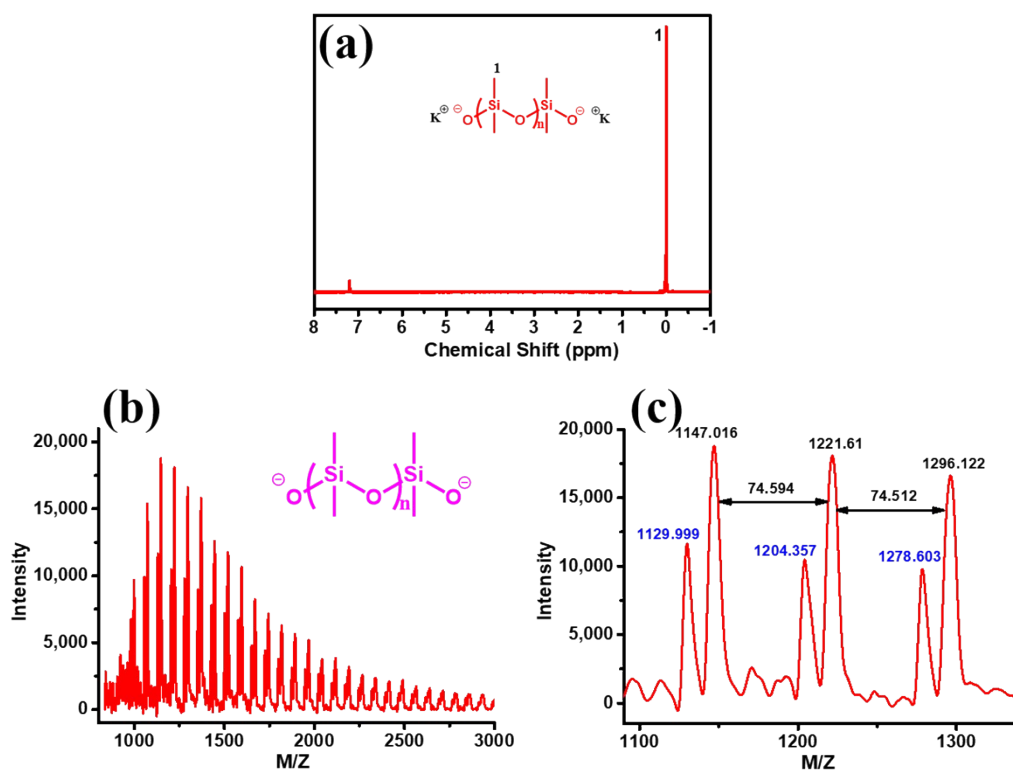


Figure S2. H-NMR (a) and MALDI-TOF MS spectrum (b-c) of silicon alkoxides.

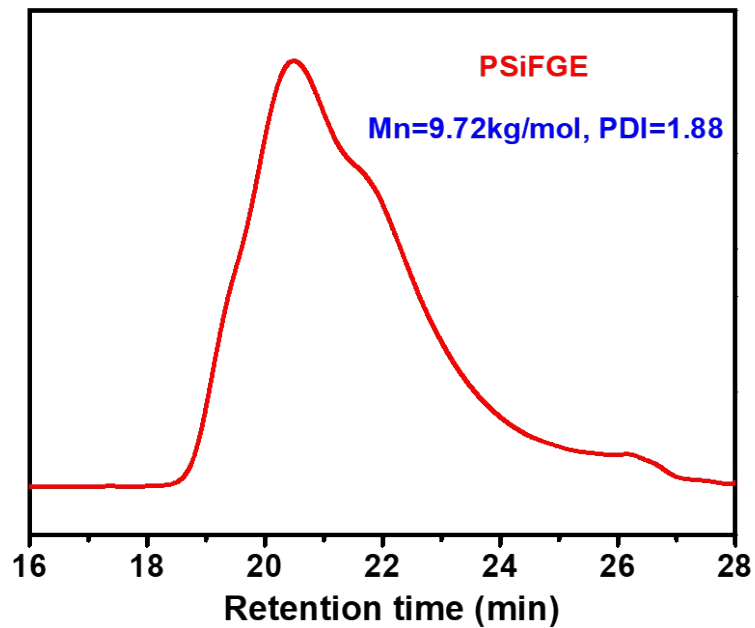


Figure S3. GPC curve of PSiFGE from the copolymerization of FGE and COS catalyzed by silicon alkoxides.

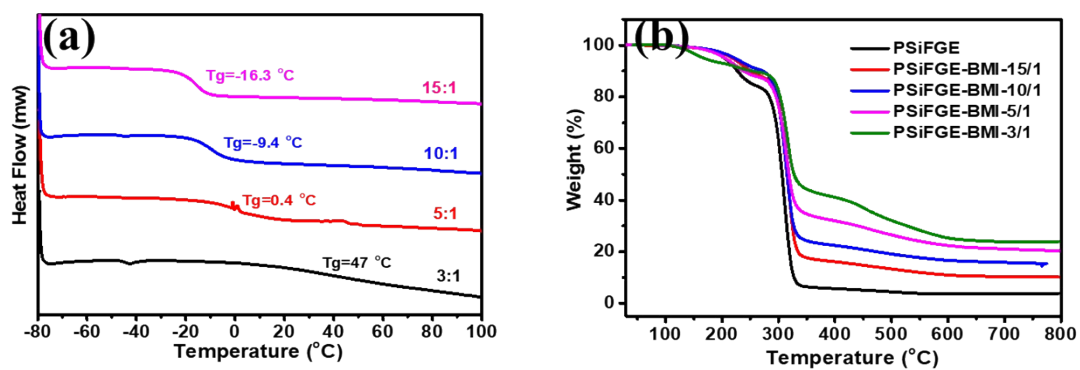


Figure S4. (a) DSC curves and (b) TGA curves for PSiFGE-BMI with different crosslinking degree.

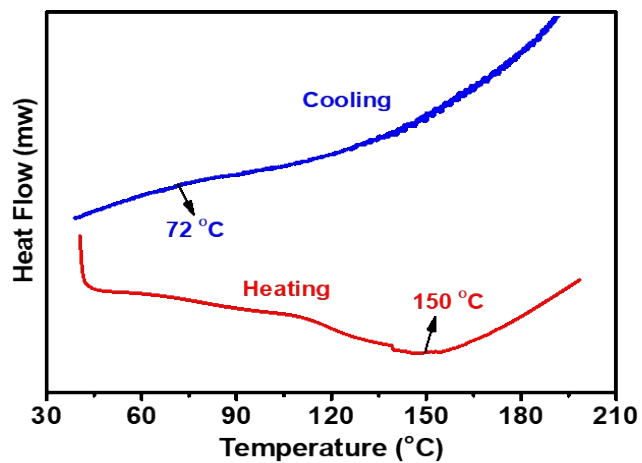


Figure S5. The DSC heating-cooling cycle for the crosslinked PSiFGE-BMI sample.

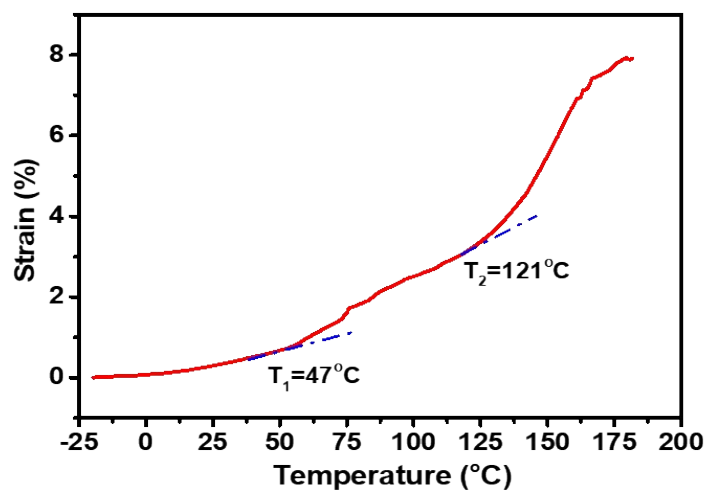


Figure S6. Dilatometry experiments for PSiFGE-BMI sample under the heating rate of 3 °C/min.