Supporting Information

Thermally resistant thermadapt shape memory crosslinked polymers based on silyl ether dynamic covalent linkages for self-folding and self-deployable smart 3D structures

Zhenjie Ding, Li Yuan, Guozheng Liang* and Aijuan Gu*

State and Local Joint Engineering Laboratory for Novel Functional Polymeric Materials
Jiangsu Key Laboratory of Advanced Functional Polymer Design and Application
Department of Materials Science and Engineering
College of Chemistry, Chemical Engineering and Materials Science
Soochow University, Suzhou 215123, P. R. China

* Corresponding author: Tel: +86 512 65880967; Fax: +86 512 65880089.
E-mail address: lgzheng@suda.edu.cn (GZ Liang); ajgu@suda.edu.cn (AJ Gu).

Table S1 Gel fraction of EPSis in DMF at 150 °C for 24 h

Sample	Gel fraction (%)
EPSi-0.3	97.7±0.08
EPSi-0.5	98.4±0.09
EPSi-0.7	99.2±0.10
EPSi-0.9	99.4±0.06

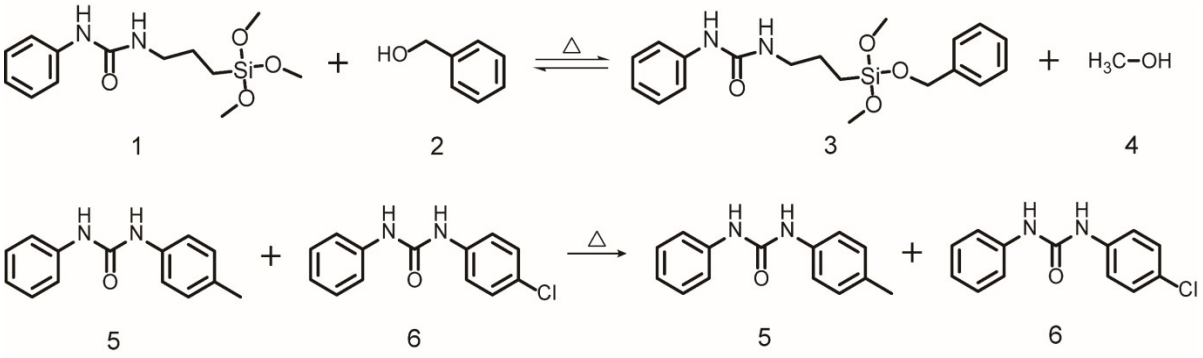

Fig. S1 Structures of model compounds used in the study.

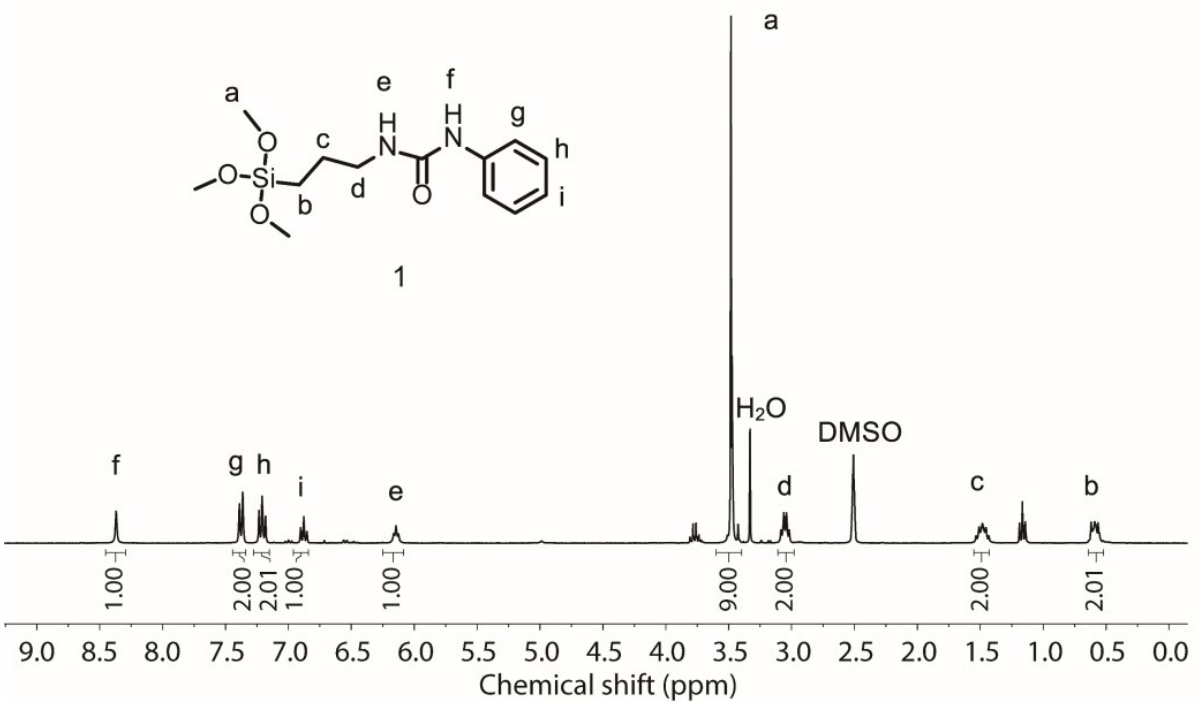

Fig. S2 ^{1}H NMR spectrum of model compound 1.

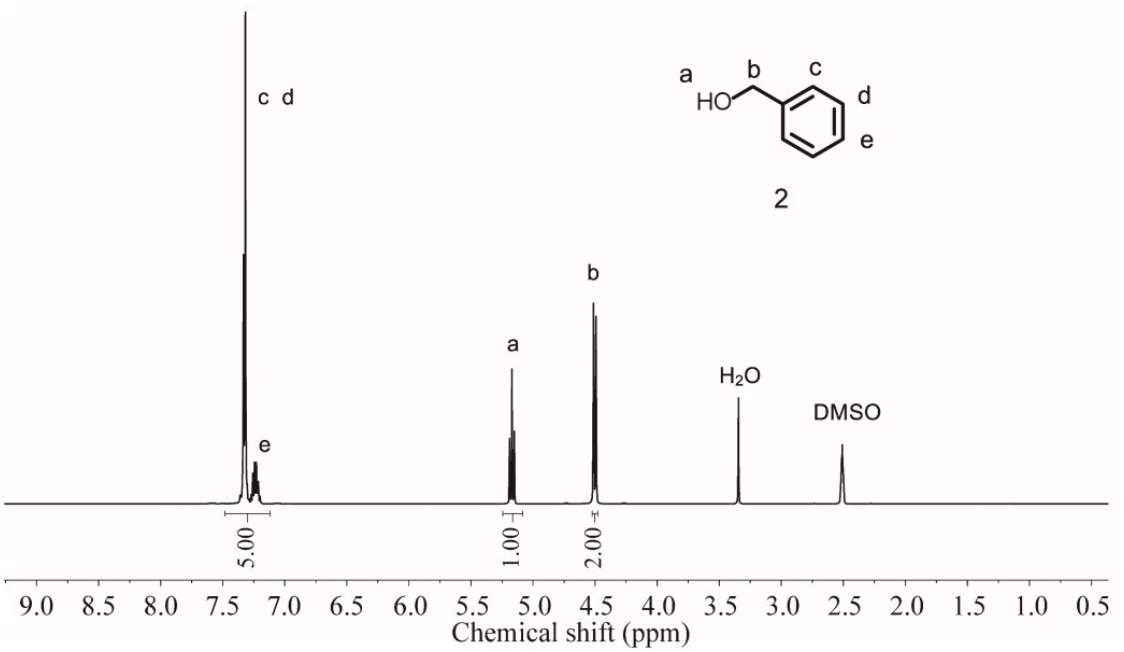

Fig. S3 ^{1}H NMR spectrum of model compound 2.

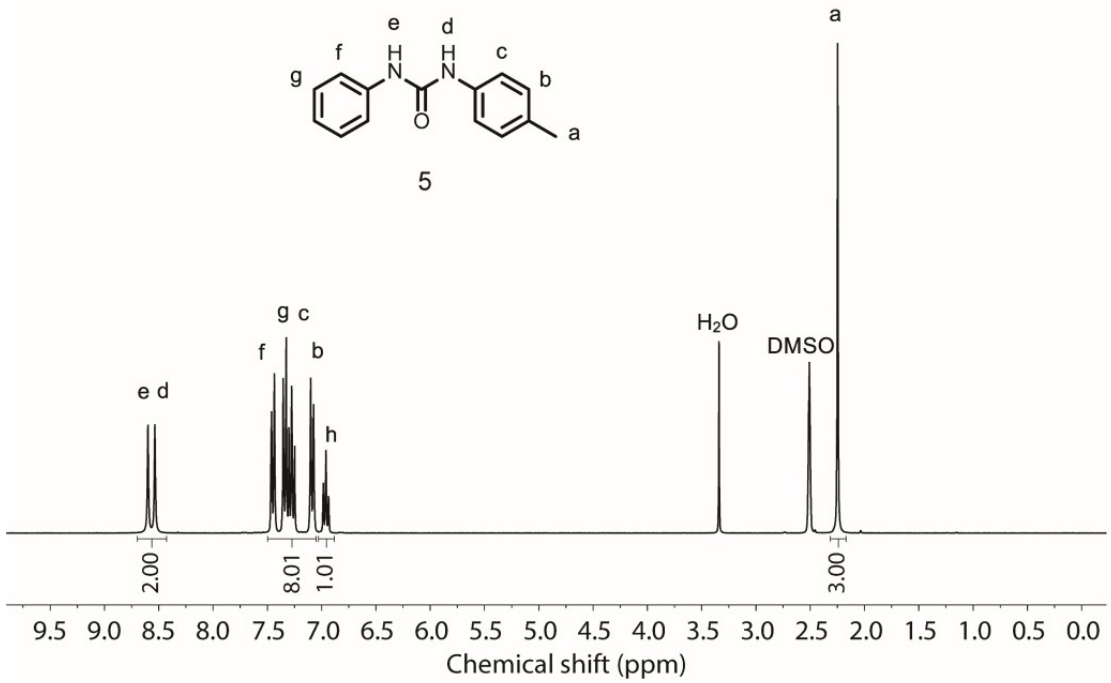

Fig. S4 ^{1}H NMR spectrum of model compound 5.

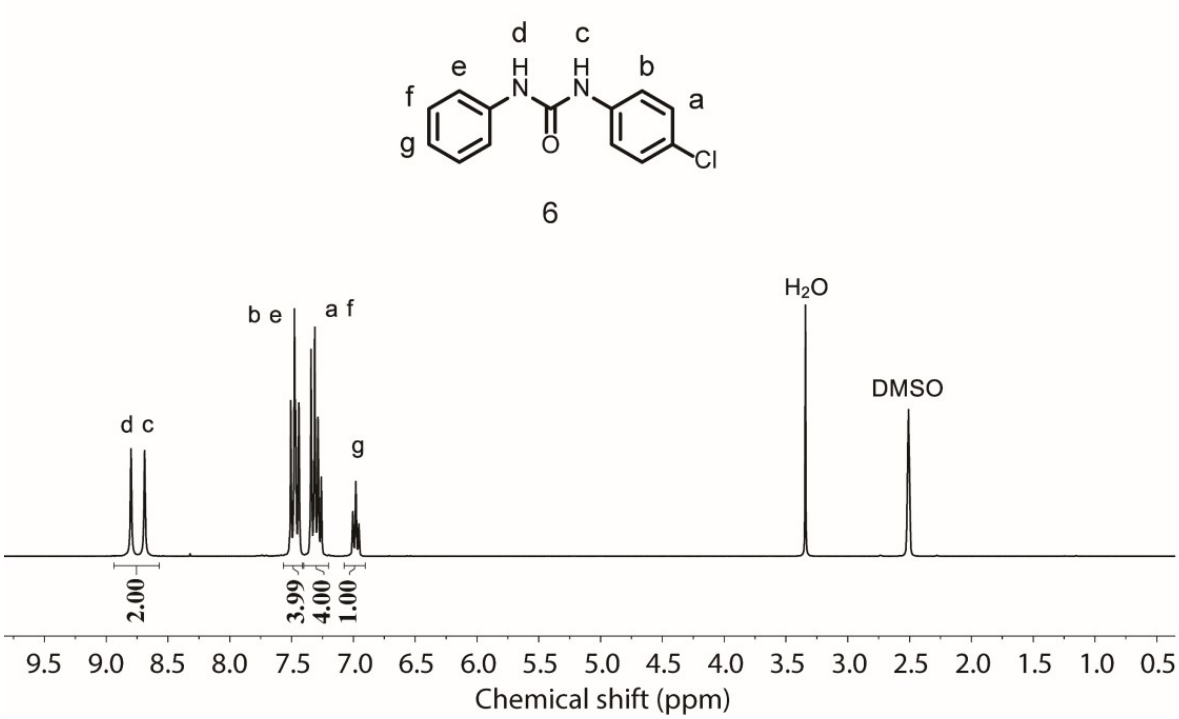

Fig. S5 ^{1}H NMR spectrum of model compound 6.

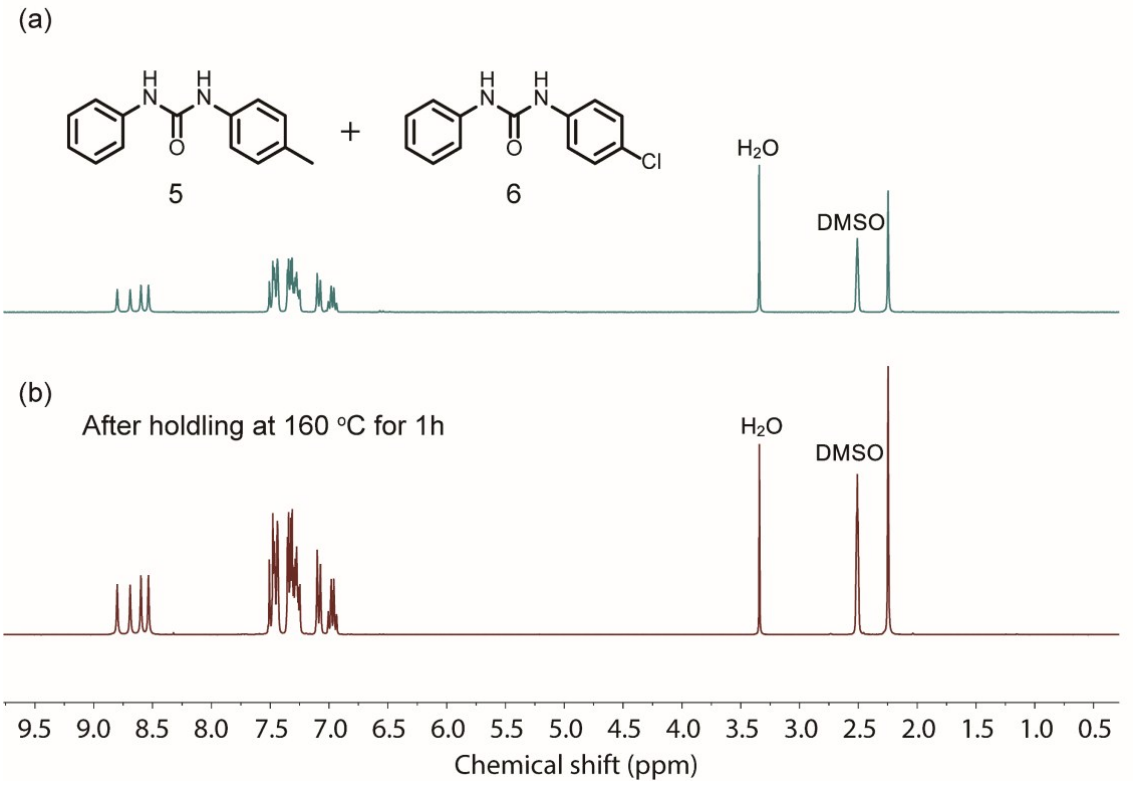

Fig. S6 ^{1}H NMR spectra of model compounds mixture (a) and after holding at 160 °C for 1 h (b).

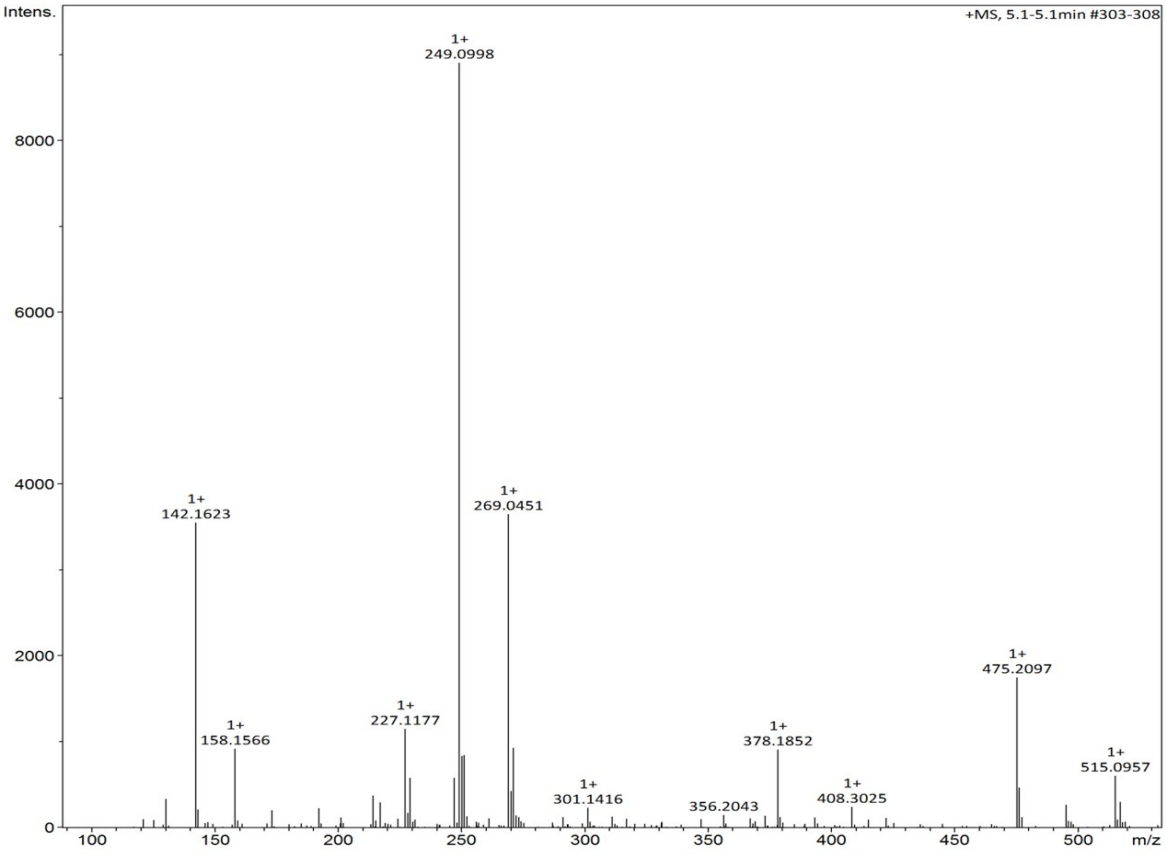

Fig. S7 Mass spectra of model compounds mixture of 5 and 6.

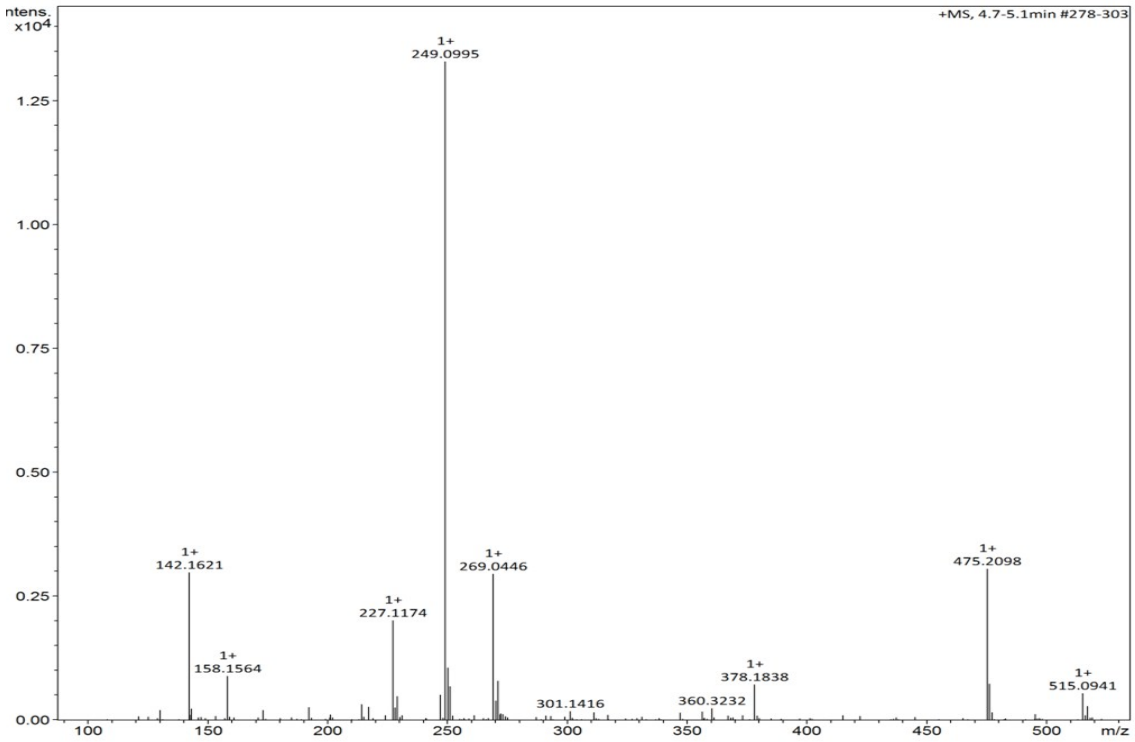

Fig. S8 Mass spectra of model compounds mixture of 5 and 6 after holding at 160 °C for 1 h.

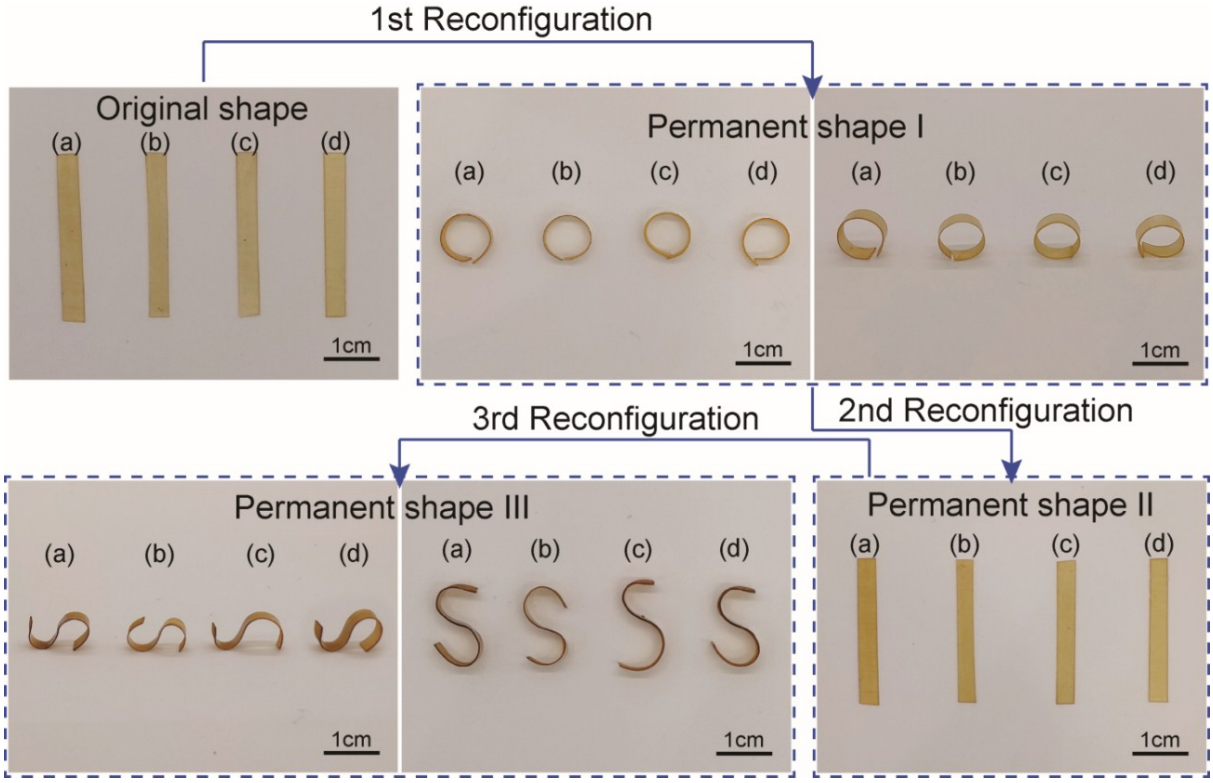

Fig. S9 Multiple reconfiguration of EPSi-0.3 (a), EPSi-0.5 (b), EPSi-0.7 (c) and EPSi-0.9 (d).

Table S2 Integrated performances of thermadapt shape memory polymers (TASMPs) in literatures and this work.

TASMP (sample name)	Dynamic bonds	T_g/T_m (°C)	T_{di} (°C)	Tensile properties			R_f (%)	R_r (%)	R_{ret} (%)	Ref
				σ (MPa)	ε (%)	E (MPa)				
EPSi-0.5	Hydroxyl silyl ether bonds	129.3	314	82.4±1.3	8.0±0.3	1864±52	95.6	99.2	83.3	This work
E51/SA/1%graphene (EP-1wt%)	Transesterification	48.3	348	22.9±1.7	44	1232±23.5	98	99	~100	[S1]
E51/SA (EP)		42.9	345	12.0±0.8	~6	565.9±10.1	--	--	--	
EP/MHHPA/PGE (epoxy 3)	Transesterification	75	--[a]	--	--	--	98	99	--	[S2]
Eu-EP/SA (1:0.5) (R=1:0.5)	Transesterification	53	310	25	8.5	1400	91.8	~100	--	[S3]
Poly(caprolactone) networks	Transesterification	~55	--	--	--	--	>98	>98	~100	[S4]
Polyanhydride networks (PAH/PCL PU)	Transesterification	~30	--	4-5	150-200	--	86-87	78-93	~98	[S5]
Epoxidized natural rubber/carbon nanodot (ENR/CD-35)	Transesterification	~40	--	17.9	452	1.5	>98	>98	~40	[S6]
Thermoset polyurethane	Transcarbamoylation	~41	--	--	--	--	98	99	~98	[S7]

Thermoset polyurethane (PU-4)	Transcarbamoylation	~80	--	--	--	--	99.7	97.9	~98	[S8]
Thermoset polyurethane (PUU3)	Transcarbamoylation	~40	--	~1.1	~520	--	95	95	~98	[S9]
Thermoset polyurethane (P1)	Transcarbamoylation	~50	--	8.16±0.57	13.04±2.90	--	>94	>94	>98	[S10]
BGPP/FA/BM (DA0.2)	Diels-Alder	35	--	--	--	--	88.8	87.5	95.6	[S11]
SBS-Fu20/CNTs	Diels-Alder	-73, 75	--	15.3-18.5	600~890	--	Photo	Photo	--	[S12]
MDS-EPO	Disulfide	41.4	268.8	10.9±2.2	0.60±0.17	1990±130	--	100	--	[S13]
Polysulfide networks (poly(S-PTMP)-51)	Disulfide	36.4	252.9	~5	~15	--	Photo	Photo	--	[S14]
Thermoset polyurethane (PU10)	Diselenide	57		17	100	--	91	97	~90	[S15]
PCL networks (PCL-6Indole)	Reversible TAD Chemistry	50	--	--	--	--	99	96-99	~80	[S16]
Metallosupramolecular networks (CP2-Ni)	Metal–ligand interactions	~50	--	--	--	--	99	95	~98	[S17]

a: Data not given in the reference.

References

[S1] Z. Yang, Q. Wang and T. Wang, *ACS Appl. Mater. Interfaces*, 2016, **8**, 21691-21699.

[S2] J. Zhu, G. Fang, Z. Cao, X. Meng and H. Ren, *Ind. Eng. Chem. Res.*, 2018, **57**, 5276-5281.

[S3] T. Liu, C. Hao, L. Wang, Y. Li, W. Liu, J. Xin and J. Zhang, *Macromolecules*, 2017, **50**, 8588-8597.

[S4] Q. Zhao, W. Zou, Y. Luo and T. Xie, *Sci. Adv.*, 2016, **2**, e1501297.

[S5] M. I. Lawton, K. R. Tillman, H. S. Mohammed, W. Kuang, D. A. Shipp and P. T. Mather, *ACS Macro Lett.*, 2016, **5**, 203-207.

[S6] Z. Tang, Y. Liu, B. Guo and L. Zhang, *Macromolecules*, 2017, **50**, 7584-7592.

[S7] N. Zheng, Z. Fang, W. Zou, Q. Zhao and T. Xie, *Angew. Chem. Int. Ed.*, 2016, **55**, 11421-11425.

[S8] N. Zheng, J. Hou, Y. Xu, Z. Fang, W. Zou, Q. Zhao and T. Xie, *ACS Macro Lett.*, 2017, **6**, 326-330.

[S9] Z. Fang, N. Zheng, Q. Zhao and T. Xie, *ACS Appl. Mater. Interfaces*, 2017, **9**, 22077-22082.

[S10] Y. Wang, Y. Pan, Z. Zheng and X. Ding, *Macromol. Rapid Commun.*, 2018, **39**, 1800128.

[S11] G. Zhang, Q. Zhao, L. Yang, W. Zou, X. Xi and T. Xie, *ACS Macro Lett.*, 2016, **5**, 805-808.

[S12] J. Bai and Z. Shi, *ACS Appl. Mater. Interfaces*, 2017, **9**, 27213-27222.

[S13] Z. Ma, Y. Wang, J. Zhu, J. Yu and Z. Hu, *J. Polym. Sci., Part A: Polym. Chem.*, 2017, **55**, 1790-1799.

[S14] S. Zhang, L. Pan, L. Xia, Y. Sun and X. Liu, *React. Funct. Polym.*, 2017, **121**, 8-14.

[S15] S. Ji, F. Fan, C. Sun, Y. Yu and H. Xu, *ACS Appl. Mater. Interfaces*, 2017, **9**, 33169-33175.

[S16] T. Defize, R. Riva, J.-M. Thomassin, M. Alexandre, N. V. Herck, F. D. Prez and C. Jérôme, *Macromol. Rapid Commun.*, 2017, **38**, 1600517.

[S17] L. Yang, G. Zhang, N. Zheng, Q. Zhao and T. Xie, *Angew. Chem. Int. Ed.*, 2017, **56**, 12599-12602.