Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Three-dimensional Graphene Coated Shape Memory Polyurethane Foam with Fast Responsive Performance

Tianjiao Wang,[a,b] Jun Zhao,*[a,b] Chuanxin Weng,[a,b] Tong Wang,[a,b] Yayun Liu,[a] Zhipeng Han,[a,c] and Zhong Zhang*[a,b]

[a] CAS Key Laboratory of Nanosystem and Hierarchical Fabrication, CAS Center for Excellence in Nanoscience, National Center for Nanoscience and Technology, Beijing 100190, China

[b] University of Chinese Academy of Sciences, Beijing 100049, China

[c] Tianjin Key Laboratory of Molecular Optoelectronic Sciences, Department of Chemistry, School of Science, Tianjin University & Collaborative Innovation Center of Chemical Science and Engineering (Tianjin), Tianjin 300072, China

* Corresponding authors: National Center for Nanoscience and Technology, Beijing 100190, China

E-mail: zhaoj@nanoctr.cn, zhong.zhang@nanoctr.cn

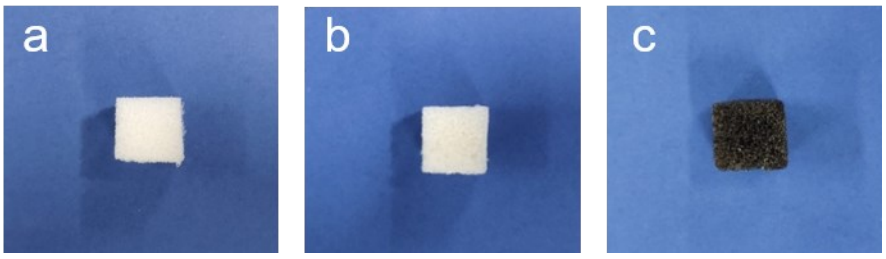

Fig. S1 The digital photographs of PU foam (a), SMPU/PU foam (b) and rGO/SMPU/PU foam (c).

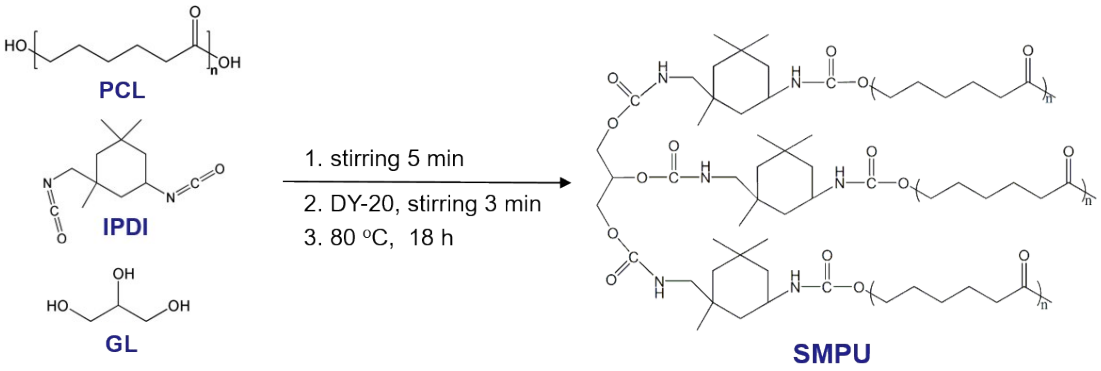

Fig. S2 Schematic representation of the synthetic route for SMPU.

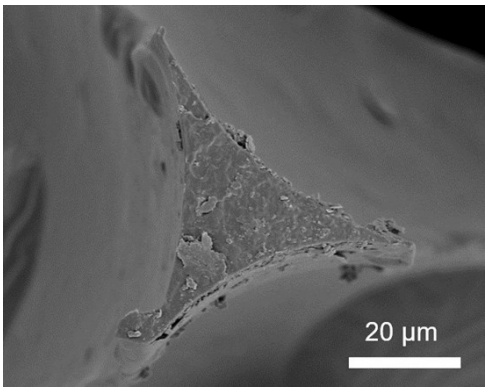

Fig. S3 The cross-sectional SEM images of rGO/SMPU/PU foams.

As shown in **Fig. S3**, the rGO is uniformly coated on the SMPU/PU foam skeleton and the thickness of rGO coating is about 0.3 μm.

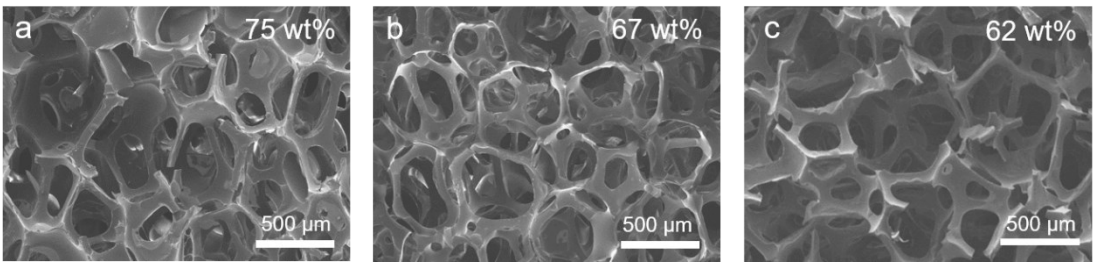

Fig. S4 The cross-sectional SEM images of SMPU/PU foams with SMPU contents of 75 wt% (a), 67 wt%(b), and 62 wt% (c), respectively.

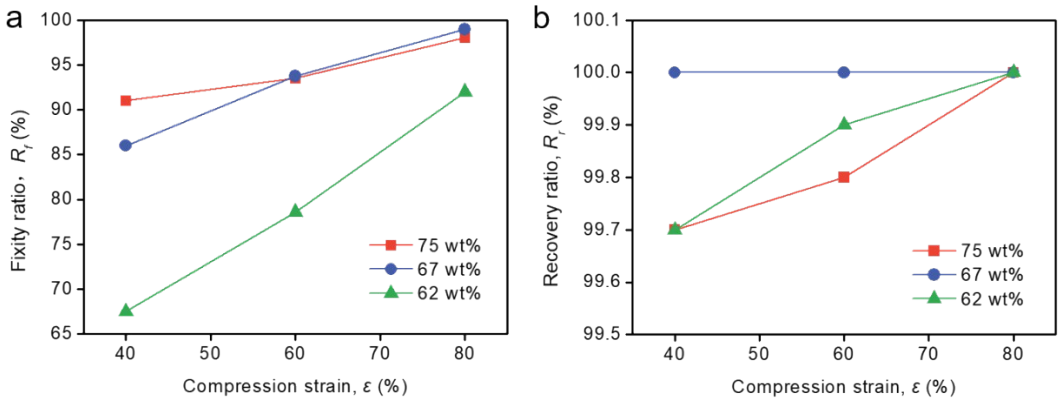

Fig. S5 The shape fixity ratio (a) and shape recovery ratio (b) of SMPU/PU foams with different SMPU contents under compression strain of 40%, 60% and 80%, respectively.

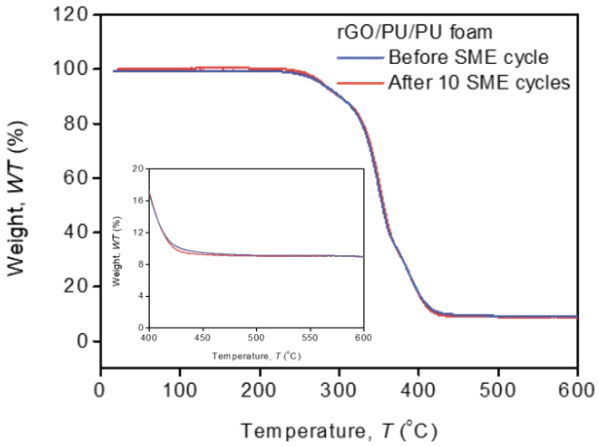

Fig. S6 The TGA curves of rGO/SMPU/PU foam before and after 10 SME cycles.

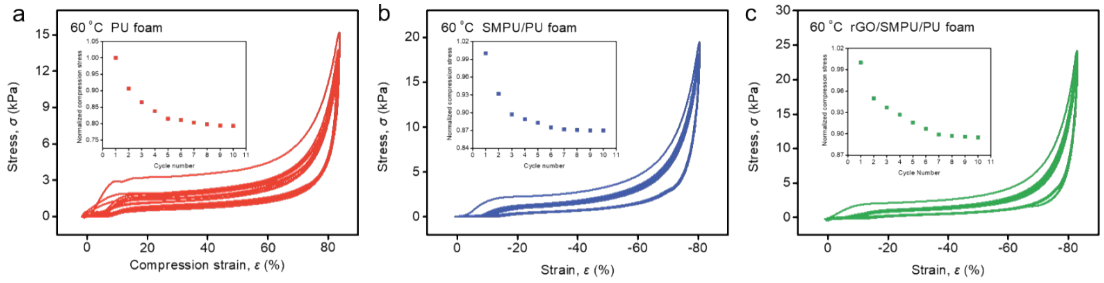

Fig. S7 The cyclic stress-strain curves of 10 cycles of PU foam (a), SMPU/PU foam (b) and rGO/SMPU/PU foam (c) at 60 ºC and 80% strain, respectively. The insets are the normalized compression stress in 10 cycles.

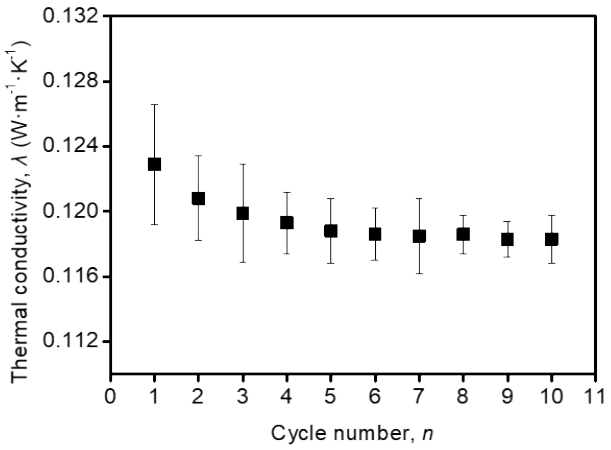

Fig. S8 The thermal conductivity of rGO/SMPU/PU foam during 10 shape memory cycles at 25 ºC.

As shown in **Fig. S8**, the thermal conductivity of rGO/SMPU/PU foam has good stable. It firstly decreases and then remains stable.

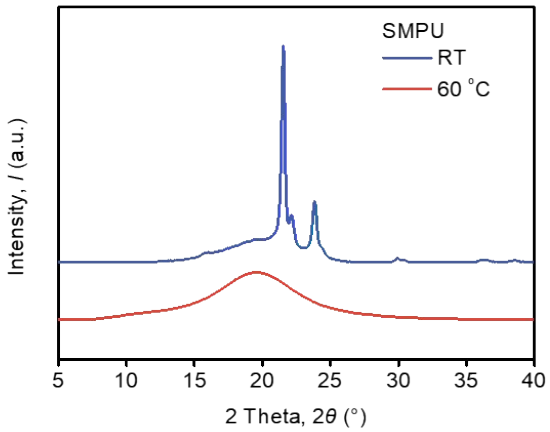

Fig. S9 XRD curves for SMPU at RT and 60 °C.

Fig. S9 illustrates the crystallinity of SMPU at different temperatures. At room temperature (RT), two sharp diffraction peaks are located at 22° and 24°, and they belong to (110) and (200) crystallographic plane, respectively. When the temperature increases to 60 °C, two sharp diffraction peaks disappear, leaving a broad peak at 20°, which is attributed to the amorphous phase. The above results show that the crystals of SMPU are melted at 60 °C.

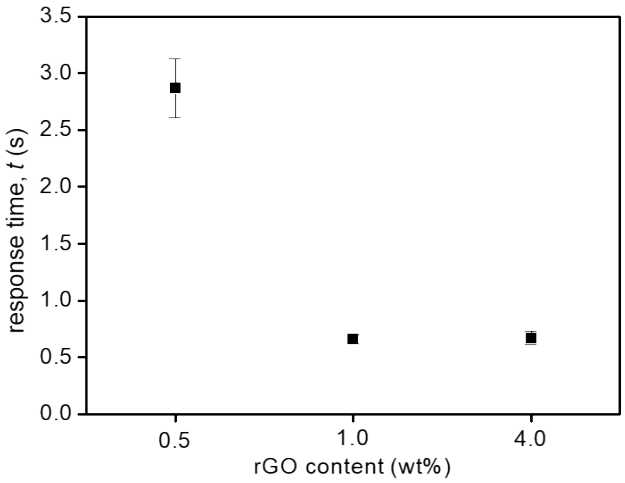

Fig. S10 The effect of rGO content on the response time of rGO/SMPU/PU foam.

As shown in **Fig. S10**, when the rGO content increases from 0.5 wt% to 1.0 wt%, the response time of rGO/SMPU/PU foam decreases rapidly from 2.87 s to 0.66 s. Moreover, when the rGO content further increases to 4.0 wt%, the response time does not decrease any more.

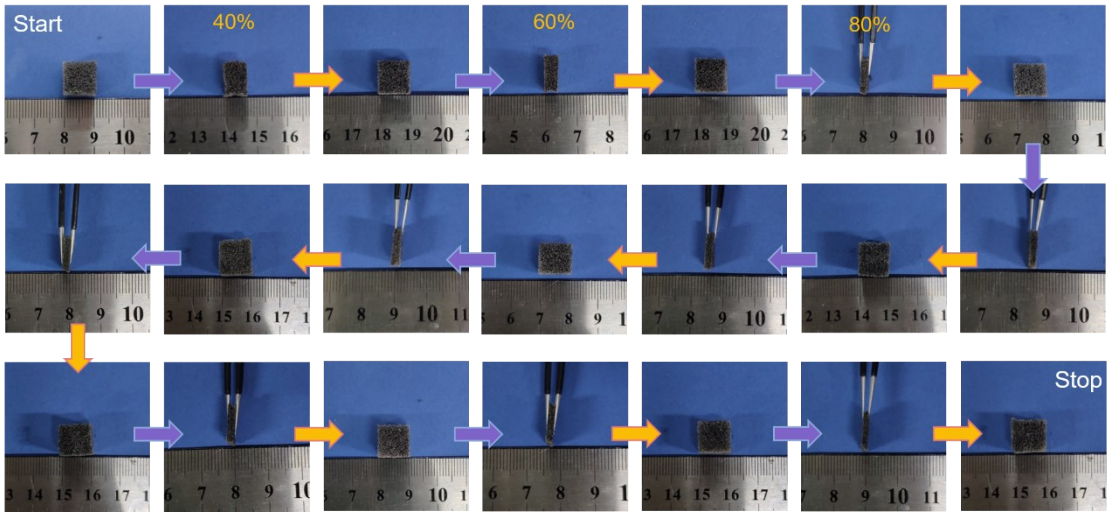

Fig. S11 The photographs of rGO/SMPU/PU foam with 10 shape memory cycles.

As shown in **Fig. S11**, the rGO/SMPU/PU foam exhibit good shape memory performance. The shape fixity ratio of the rGO/SMPU/PU foam under 80 % compression strain is about 98.95%±0.03% and the shape recovery ratio is always 100% during 10 shape memory cycles.

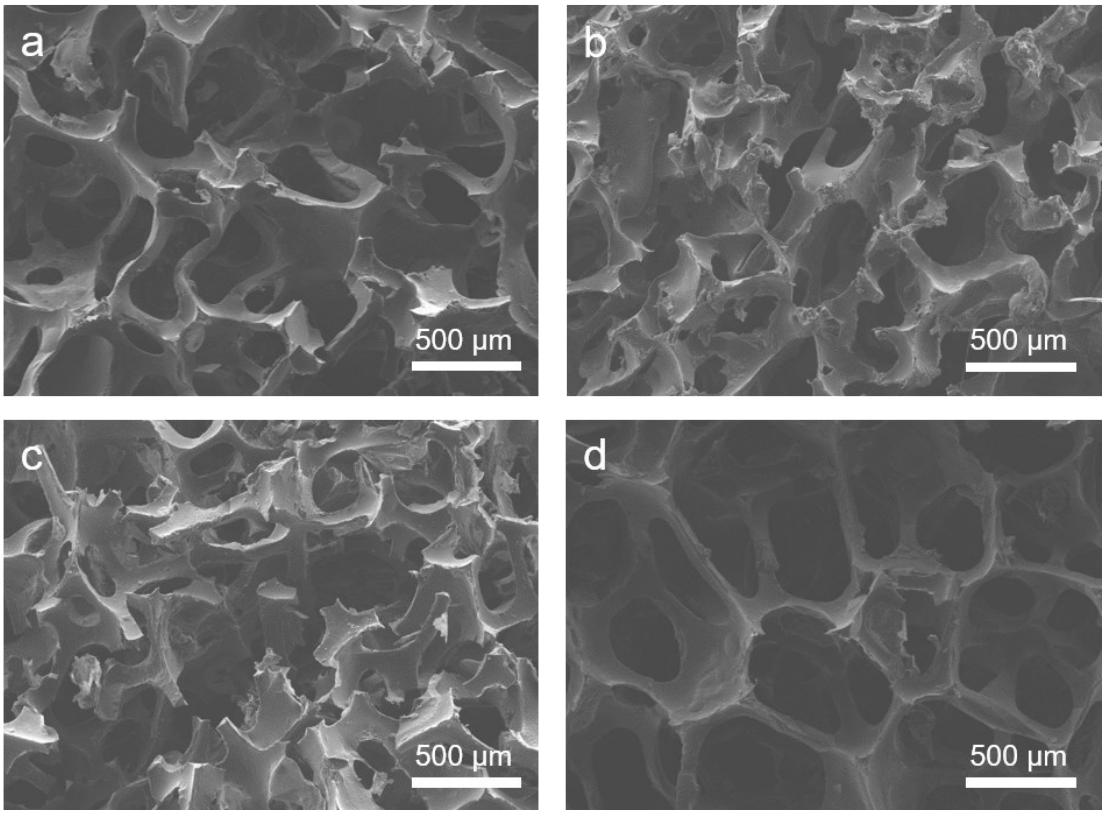

Fig. S12 The cross-sectional SEM images of rGO/SMPU/PU foams with 40% strain (a), 60% strain (b), 80% strain (c) and shape recovery (d).

Table S1 Shape memory performance of various SMPs.

Materials	Stimuli	Recovery time (s)	Recovery ratio (%)	Reference
PGEC composite	electricity	150	96	38
NIPAM hydrogels	solvent	10	98	8
TPI-graphene-PAAm composites	electricity	7	99	28

PNIPAM/PPY hydrogels	solvent	10	99	7
CB@CNC/NR@PU	solvent	3-12	100	39
CNT/SMP	electricity	10	98	32
Graphene-CNT/PU	heat	23-40	79-90	27
CNT/EVA	heat	20	90	25
AgNWs/MF/Epoxy	electricity	25	99	16
PLA-PEG	light	7200	58-80	5
PVA hydrogels	heat	600	80	4
PECU/CNC	solvent	1800	85	40
PEG-PCL-CNC	solvent	300	90	41
CB/PU	light	125-350	78-100	20
dPTB-mfGO	light	25	96	20
dPTB-mfGO	heat	342	96	20
dPTD-mfGO	light	28	93	20
dPTD-mfGO	heat	390	93	20
dPTD-GO	light	65	69	20
dPTD-GO	heat	485	69	20
SMPU/PU foam	heat	＜5s	100	This work
rGO/SMPU/PU foam	heat	＜1s	100	This work

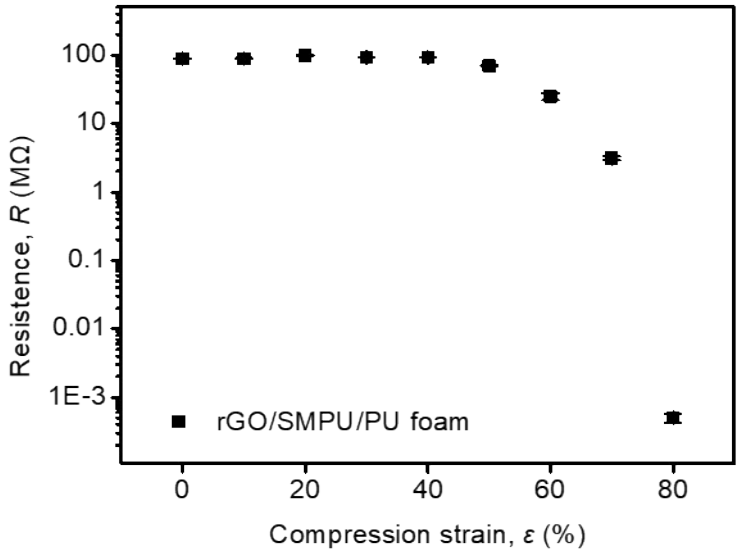

Fig. S13 Variation of resistance change concerning compression strain for rGO/SMPU/PU foam.