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Supplementary Materials for

Bioinspired shape-memory graphene film with tunable wettability

Jie Wang, Lingyu Sun, Minhan Zou, Wei Gao, Cihui Liu, Luoran Shang, Zhongze Gu, Yuanjin Zhao

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Other Supplementary Material for this manuscript includes the following: (available at advances.sciencemag.org/cgi/content/full/3/6/e1700004/DC1)

• movie S1 (.avi format). Compressive tests of the graphene sponge for 10 cycles.

- movie S2 (.avi format). Compressive tests of the graphene/TPI hybrid sponge for 10 cycles at 100°C.
- movie S3 (.avi format). Dynamic wettability of the graphene/TPI hybrid film by monitoring the liquid droplets on the slippery surface shown in Fig. 5.



fig. S1. Raman spectra of GO and reduced GO of the sponge.



fig. S2. SEM images of pure TPI, and the photos of the graphene sponge and the graphene/TPI hybrid sponge. (A, B) SEM images of the crystallized morphology of pure TPI. The appearance of (C) the graphene sponge and (D) the graphene/TPI hybrid sponge. The scale bars are 50 μm in (A), 5 μm in (B), and 5 mm in (C, D).



fig. S3. DSC thermograms of TPI, the hybrid sponge without TPI, and the hybrid sponge with different concentrations of TPI. The measured T_c and T_m of the hybrid sponge with 20% (w/v) TPI were about 56.6 °C and 18.9 °C, respectively.



fig. S4. SEM images of the graphene sponge before and after 10 cycles' compression. SEM images of the graphene sponge (A, B) before and (C, D) after 10 cycles' compression. (B, D) are magnified images of (A, C), respectively. The scale bars are 100 μ m in (A, C) and 50 μ m in (B, D).



fig. S5. The mechanical properties of graphene/TPI hybrid sponges. (**A**) The height variation of the graphene/TPI hybrid sponge as a function of compression numbers, the critical number of compression showing the degradation of structural reversibility (decreased to 99%) is about 700 times. (**B**) The height variation of hybrid sponges with different concentrations of GO and AAm after 2000 cycles of compression.



fig. S6. The photos of graphene/TPI hybrid sponge before, under, and after compression at room temperature. The appearances of graphene/TPI hybrid sponge (A) before, (B) under and (C) after compression under room temperature. Scale bars are 5 mm.



fig. S7. The resistance and temperature of a cylindrical graphene/TPI hybrid sponge as a function of time.



fig. S8. The wettability of the graphene/TPI hybrid film with and without perfluorinated oil. The water contact angles of the graphene/TPI hybrid film (A) with and (C) without perfluorinated oil. The oil contact angles of the graphene/TPI hybrid film (B) with and (D) without perfluorinated oil. Both of the water and oil droplets are $2 \mu L$.



fig. S9. The dynamic control of droplet mobility on a tilted graphene sponge film. (A) The progress of a water droplet sliding down the surface of a graphene sponge film filled with perfluorinated oil. (B) The progress of the water droplet pinned on the graphene sponge film surface without perfluorinated oil. The time differences that the images taken from the first images in (A, B) are shown in corresponding images. Scale bars are 1 cm.



fig. S10. The schematic diagram and the photo of the hybrid graphene film array with four units. (**A**) The schematic diagram and (**B**) the optical image of the hybrid graphene film array with four units. The scale bar is 1 cm in (**B**).

Sample	GO	AAm	MBAA	KPS	Ascorbic acid	H ₂ O
	/mg	/mg	/mg	/mg	/mg	/mg
G ₁ P ₀	75	0	0	0	150	10
G1P0.5	75	37.5	1.6	0.3	150	10
G1P5	75	375	16.5	3	150	10
G1/3P1	25	75	3.3	0.6	50	10
$G_{2/3}P_1$	50	75	3.3	0.6	100	10
G ₁ P ₁	75	75	3.3	0.6	150	10
G4/3P1	100	75	3.3	0.6	200	10

table S1. Compositions of initial aqueous solutions used for preparing hybrid sponges with different concentrations of GO (G) and AAm (P).

movie S1. Compressive tests of the graphene sponge for 10 cycles.

movie S2. Compressive tests of the graphene/TPI hybrid sponge for 10 cycles at 100°C.

movie S3. Dynamic wettability of the graphene/TPI hybrid film by monitoring the liquid droplets on the slippery surface shown in Fig. 5.