Supporting Information

Small Graphene Oxide Sheets/Polyvinylidene Fluoride Bilayer Actuator with Large and Rapid Responses to Multiple Stimuli

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Fig. S1 (a) SEM image of SGO sheets. (b) Size distribution of SGO sheets (the histogram was obtained by counting the largest 100 sheets in the SEM image).



Fig. S2 Water contact angles of (a) SGO, (b) PVDF and (c) ozone plasma treated PVDF films.



Fig. S3 (a) The maximum curvatures of the actuators with 10 μ m-thick PVDF layers and SGO layers with different thicknesses. (b) Plot of the maximum curvature of the actuator with an SGO/PVDF thickness ratio of 0.8 versus its total thickness. The actuations were performed by changing RH from 61% to 75%; size of the actuator = 5 mm × 1 mm.



Fig. S4 Digital image of an actuator-*m* at RH of 11%; scale bar =1 mm.



Fig. S5 (a) C1s XPS spectra of SGO, rSGO-10 and rSGO-60 films. (b) The C/O ratio and curvature of the three kinds films at RH of 68%.



Fig. S6 (a) The curvatures of SGO/PVDF or CNT/PVDF actuator at different temperatures (RH = 43%). (b) Digital photographs of the SGO/PVDF (size = 5 mm × 1 mm) or CNT/PVDF (size = 5 mm × 1 mm) at different temperatures (RH = 43%).



Fig. S7 Tip displacements of a 42 mm× 48 mm actuator-*m* recorded during the process of irradiation with 60 mW cm⁻² IR light.



Fig. S8 C1s XPS spectra of SGO before and after 150 cycle under light with power density of 80 mW cm⁻² on and off.



Fig. S9 Currents generated by SGO/PVDF bilayer actuator with power density of 20, 40, 60 or 80 mW cm⁻².

Table S1	Performance	comparison	of	different	actuators
		1			

Materials	Stimulation	Maximal	Response	Ref.
		curvature (cm ⁻¹)	time (s)	
rGOCNT/PDMS ^a	Light	4.91	3.6	[S1]
SWCNT ^b /PVDF	Light	3.18	1.1	[S2]
SACNT ^c /BOPP ^d	Electric	1.03	4	[S3]
SGO/rGO	Moisture	19.1	6.8	[S4]
PILTf ₂ N ^e	Acetone	13.5	0.4	[S5]
CLCP ^f	Moisture, light	2.3	0.4	[S6]
SGO/PVDF	Moisture, heat, light	22	0.3	This work

^a PDMS: Polydimethylsiloxane;

^b SWCNT: single walled carbon nanotube;

°SACNT: super-aligned carbon nanotube;

^dBOPP: biaxially oriented polypropylene;

^ePILTf₂N:poly(3-cyanomethyl-1-vinylimidazolium bis(trifuoromethanesulfonyl)imide);

^fCLCP: cross-linked liquid crystals polymer containing an azobenzene moiety.

Supplementary References

S1 Y. Hu, G. Wu, T. Lan, J. Zhao, Y. Liu and W. Chen, Adv. Mater., 2015, 27, 7867–7873.

- S2 Y. Tai, G. Lubineau and Z. Yang, Adv. Mater., 2016, 28, 4665–4670.
- S3 L. Chen, M. Weng, Z. Zhou, Y. Zhou, L. Zhang, J. Li, Z. Huang, W. Zhang, C. Liu and S. Fan, ACS Nano, 2015, 9, 12189-12196.
- S4 G. Xu, J. Chen, M. Zhang and G. Shi, Sens. Actuators, B, 2017, 242, 418-422.
- S5 Q. Zhao, J. W. C. Dunlop, X. Qiu, F. Huang, Z. Zhang, J. Heyda, J. Dzubiella, M. Antonietti and J. Yuan, *Nat. Commun.*, 2014, **5**, 4293.
- S6 Y. Liu, B. Xu, S. Sun, J. Wei, L. Wu and Y. Yu, Adv. Mater., 2017, 29, 1604792.