Electronic Supplementary Information (ESI)

Scalable preparation of flexible heterogeneous graphene oxide structures for high performance wet power generation

Yu Gao [a,d], Xiaoming Cai [b,c], Yuzhou Zhao [a], Wentian Huang [a,c], Jian Lv [c], Jie Wang [c], Huiming Liang [c], Zhenliang Hao [a,c], Hongling Tan [a], Jinming Cai *[a,c,d]

a. Faculty of Materials Science and Engineering, Kunming University of Science and Technology, Kunming, Yunnan 650000, P. R. China
b. Faculty of Mechanical and Electrical Engineering, Kunming University of Science and Technology, Kunming, Yunnan 650500, P. R. China
c. Guangdong Morion Nanotechnology Co., Ltd, Dongguan 523069, Guangdong, P. R. China
d. Southwest United Graduate School, Kunming, 650000, P. R. China

Correspondence to: j.cai@kust.edu.cn

Supplementary Figures and Tables

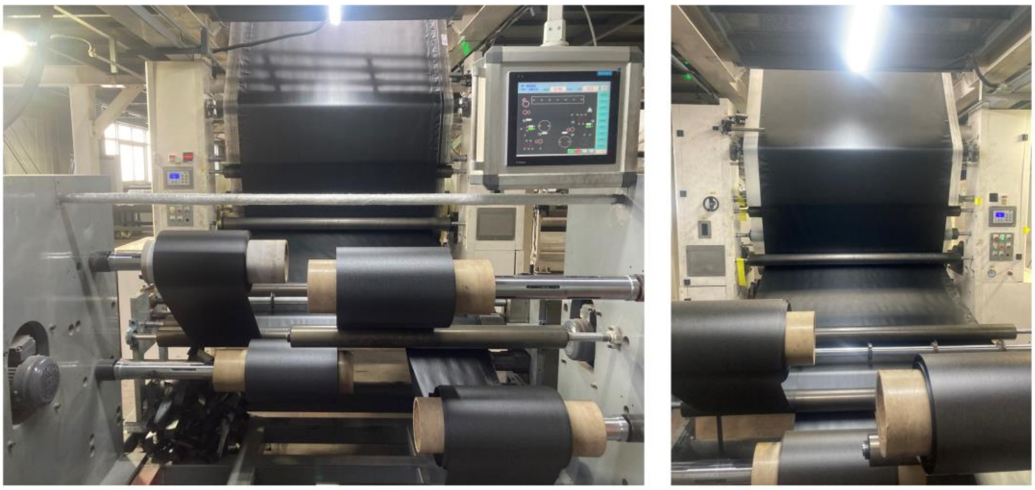

Fig. S1 Digital photos of industrialized coated preparation of graphene oxide films.

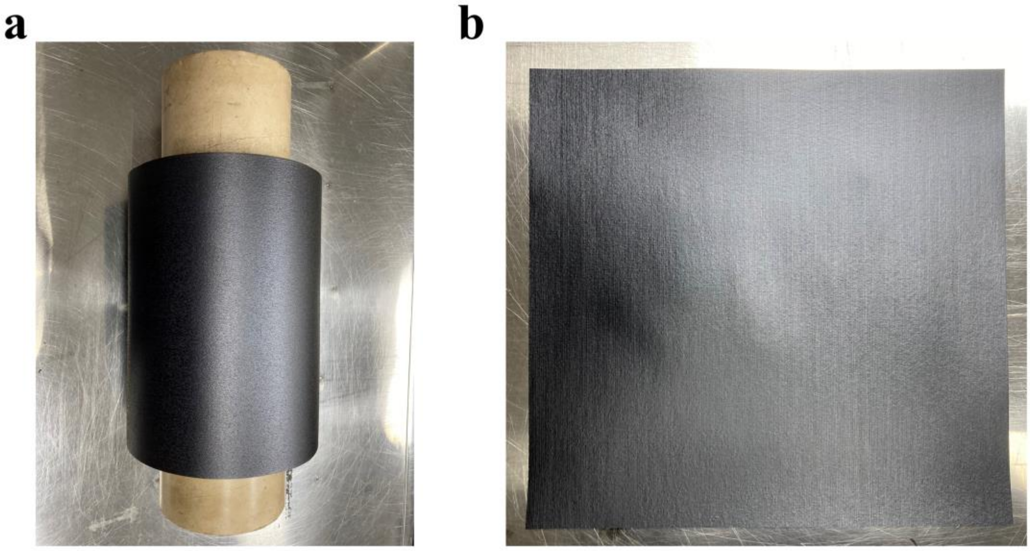

Fig. S2 Photos of (a) graphene oxide roll films. (b) cropped graphene oxide films.

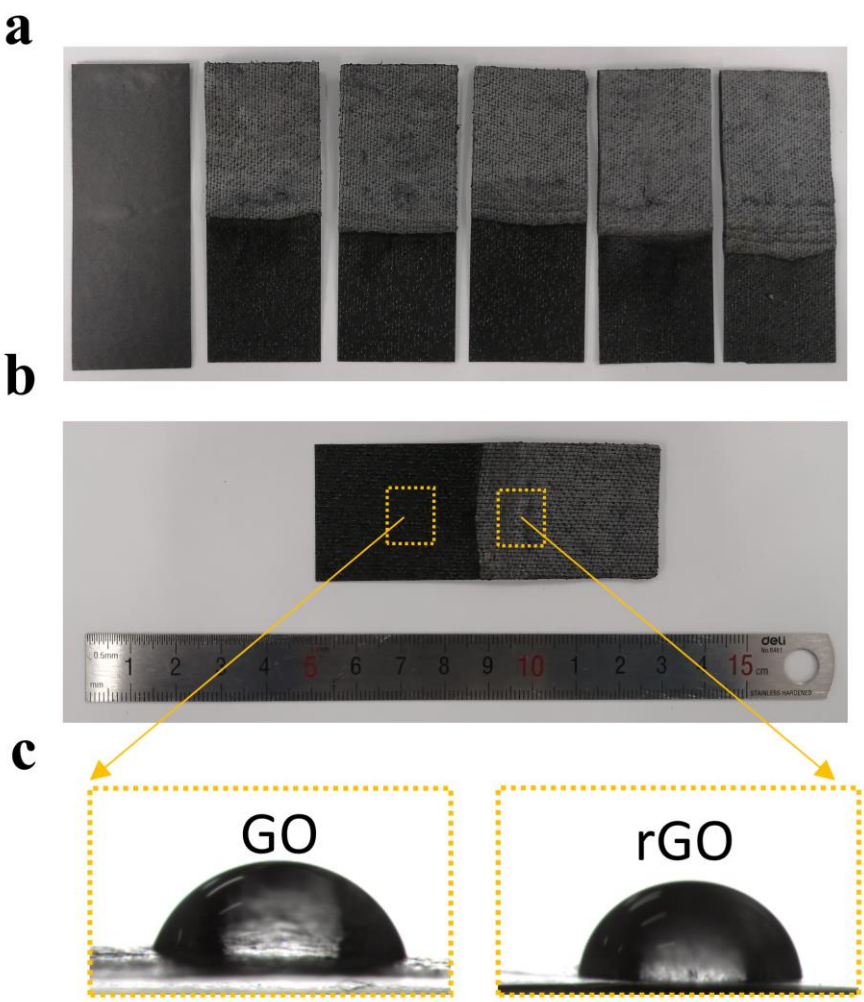

Fig. S3 (a,b) Images of HGOS prepared by thermal reduction. The color of the GO film is black, while the reduced rGO turns light gray. (c) Water contact angle of GO and rGO. The water contact angle increases when GO is reduced to rGO.

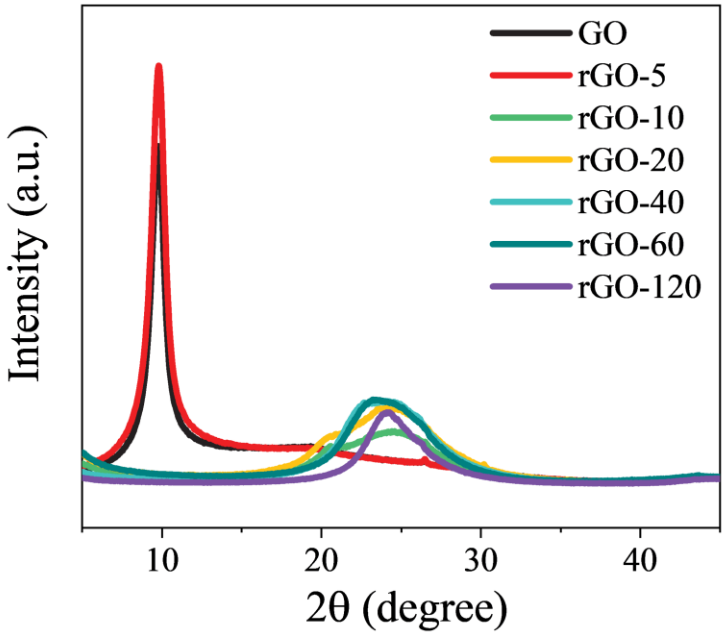

Fig. S4 XRD patterns of GO and rGO prepared with different thermal reduction time.

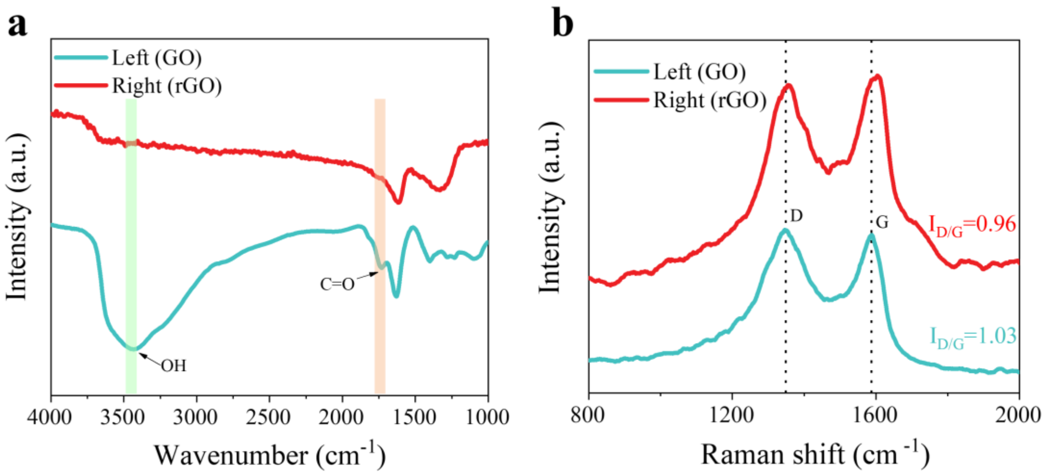

Fig. S5 (a) FTIR spectra of HGOS. (b) Raman spectra of HGOS.

Figure S5a shows the FTIR spectra of HGOS, blue colour corresponds to GO and red colour corresponds to rGO after thermal reduction. As can be seen from the figure, the intensity of the peaks of OH at 3470 cm^{-1} and C=O at 1735 cm^{-1} decreases after thermal reduction, suggesting that GO is reduced by the shedding of these groups during the thermal reduction process. Figure S5b shows the Raman spectra of HGOS, after thermal reduction, the D peak of GO decreases and $I_{D/G}$ decreases from 1.03 to 0.96, indicating the reduction of GO to rGO.

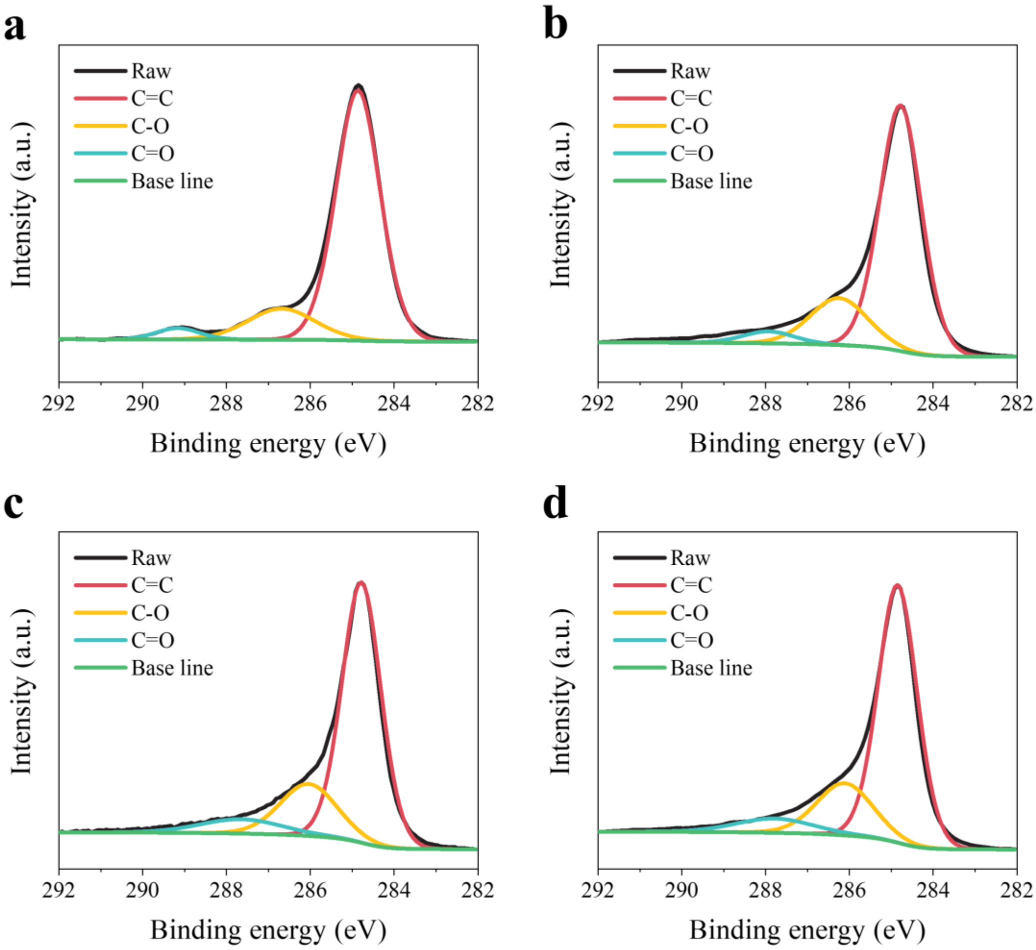

Fig. S6 C1s XPS spectra of (a) rGO-5, (b) rGO-10, (c) rGO-40, and (d) rGO-120.

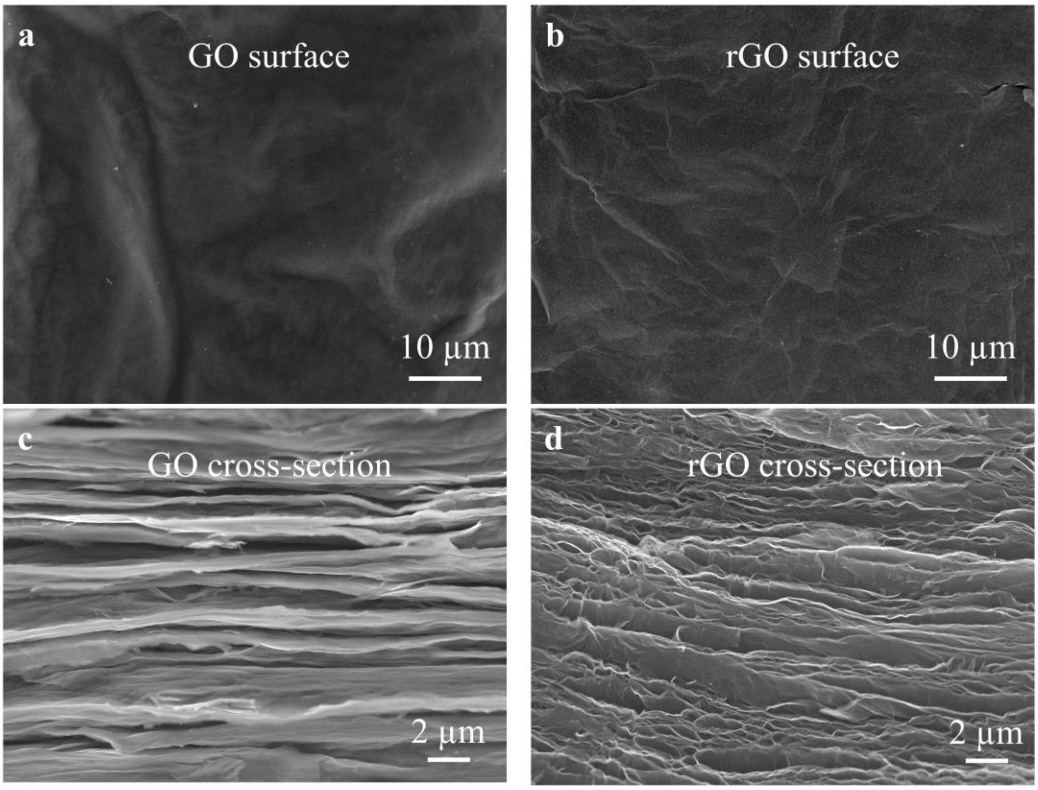

Fig. S7 SEM images of surface (a,b) and cross-section (c,d) of HGOS.

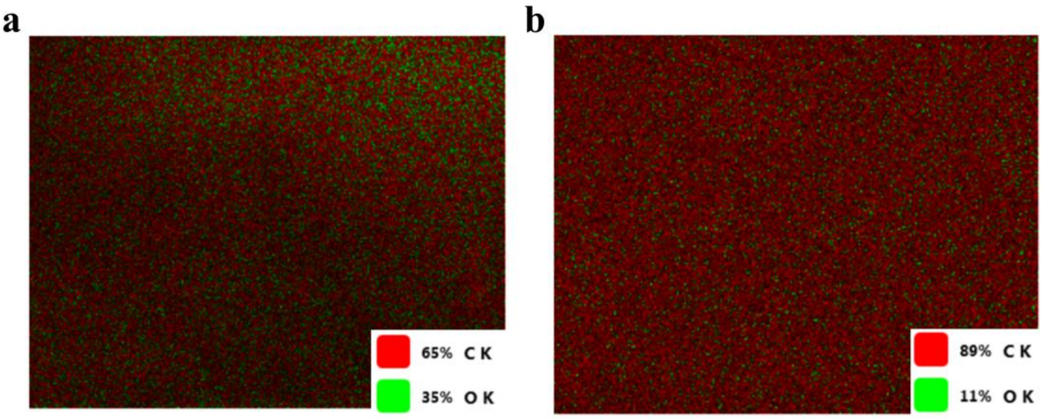

Fig. S8 EDS energy spectrum of the (a) left side (GO), and (b) right side (rGO) of HGOS.

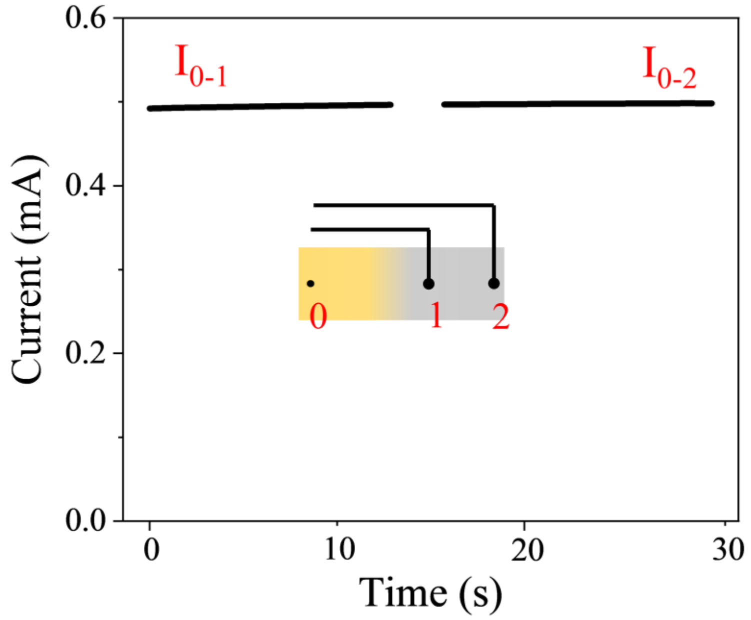

Fig. S9 Short circuit current between different electrodes on rGO.

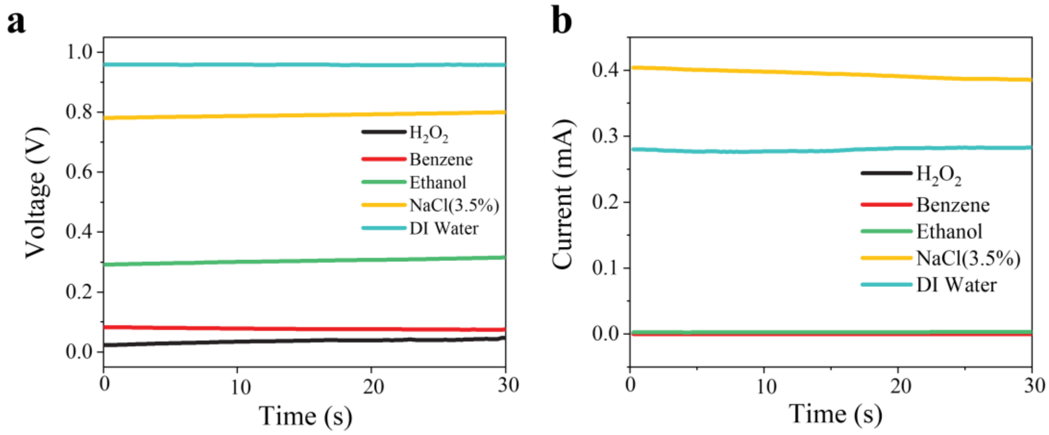

Fig. S10 (a)Voltage and (b)current of HGOS under different electrolytes.

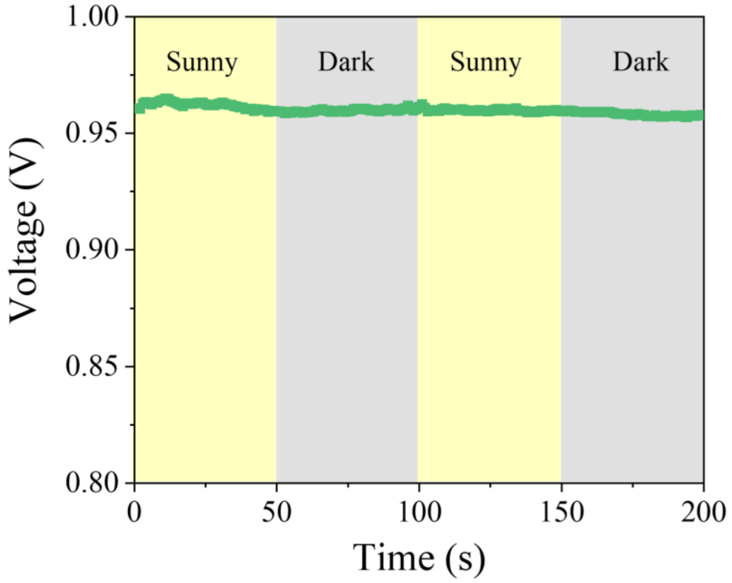

Fig. S11 Open-circuit voltage (Voc) of HGOS devices under sunny and dark ambient conditions with a relative humidity (RH) of about 75%.

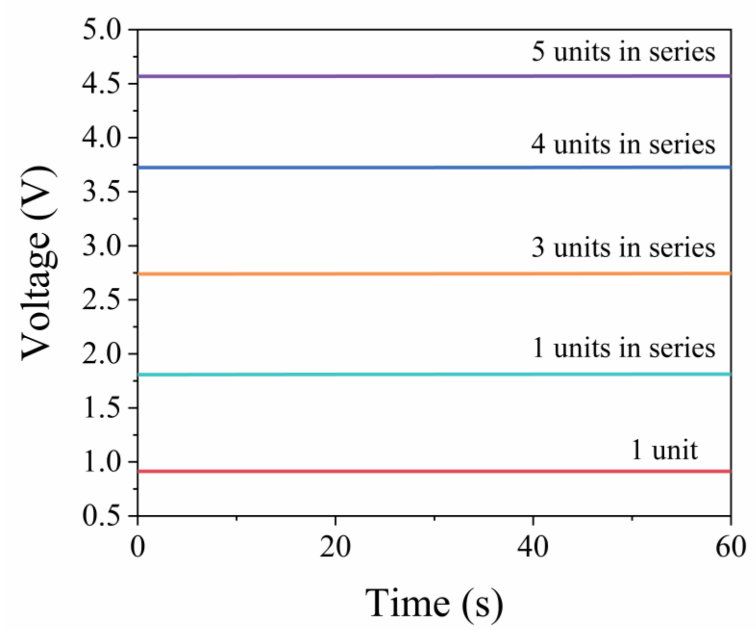

Fig. S12 The open circuit voltage of a series device.

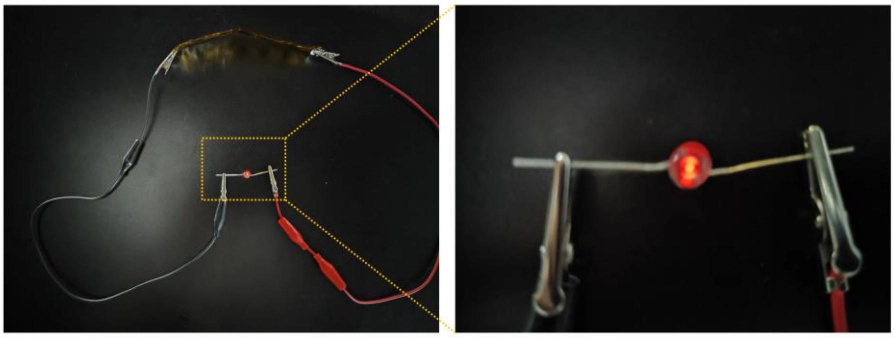

Fig. S13 Soft-packaged HGOS devices can power red LEDs.

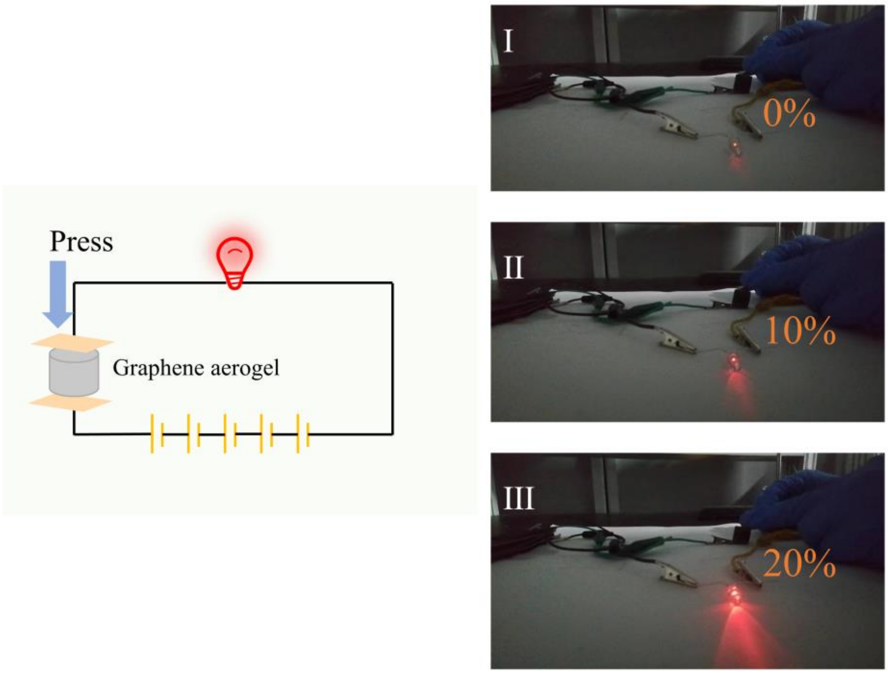

Fig. S14 The integrated HGOS device is connected in series with a graphene aerogel to control the brightness of the red LED by squeezing the aerogel to produce a change in resistance.

Table S1 Effect of thermal reduction time on oxygen content

Samples	**heat treatment temperature (℃)**	**Heat treatment time (s)**	**O Atomic %**
rGO-5	450	5	35.99
rGO-10	450	10	21.97
rGO-40	450	40	13.25
rGO-120	450	120	12.47

Table S2 2θ and d values of rGO prepared with different thermal reduction times. According to the Bragg equation $2d\sin\theta = n\lambda$ (λ = 0.15406 nm, n = 1).

Samples	**2θ (°)**	**d (nm)**
GO	9.78	0.90
rGO-5	9.80	0.90
rGO-60	23.64	0.38
rGO-120	24.17	0.37

Table S3 Summary of reported water-energy harvestings and performance comparisons

Material	V_{oc}	I_{sc}	Water form	Solution	Ref.
CB sheet	~1 V	~150 nA	Water evaporation	DI water	Nat Nanotechnol.,2017[1]
Porous GO sponges	~0.63 V	~27.4 μA	Water evaporation	DI water	Carbon,2019[2]
rGO film	~1 V	~10 μA	Water evaporation	NaCl	ACS Appl. Mater. Interfaces,2021[3]
3D wood	~0.3 V	~10 μA	Water evaporation	DI water	ACS Appl. Mater. Interfaces,2020[4]
TiO2 nanoparticle film	~0.2 V	~120 nA	Water evaporation	DI water	Journal of Materials Science & Technology, 2021[5]
GO film	~20 mV	~5 μA cm^{-2}	Moisture	ΔRH~30 %	Adv Mater, 2015[6]
GO framework	~0.26 V	~3.2 mA cm^{-2}	Moisture	ΔRH~75 %	Energy & Environmental Science, 2016[7]
Flexible GO film	~0.2 V	~1.2 μA	Moisture	ΔRH~80 %	Adv Mater, 2019[8]
GO fiber	~0.35 V	~1.06 mA cm^{-2}	Moisture	ΔRH~65 %	Nano Energy, 2017[9]
PSS/PVA film	~0.6 V	~13.2 μA cm^{-2}	Moisture	ΔRH~85 %	Nano Energy, 2020[10]
Graphene/PET	<1 mV	<1 μA	Water drolet	NaCl	Nat Nanotechnol.,2014[11]
Graphene/PET	~0.5 V	~25 μA	Water drolet	NaCl	J Am Chem Soc, 2018[12]
PTFE	~4 V	~0.5 μA	Water drolet	Water	Nat commun, 2013[13]
PTFE	~150 V	~250 μA	Water drolet	Water	Nature, 2020[14]
GO-rGO	**~1 V**	**~490 μA**	**Water contact**	**DI Water**	**This work**

Reference

S1. G. Xue, Y. Xu, T. Ding, J. Li, J. Yin, W. Fei, Y. Cao, J. Yu, L. Yuan, L. Gong, J. Chen, S. Deng, J. Zhou and W. Guo, *Nat Nanotechnol*, 2017, **12**, 317-321.

S2. G. Zhang, Z. Duan, X. Qi, Y. Xu, L. Li, W. Ma, H. Zhang, C. Liu and W. Yao, *Carbon*, 2019, **148**, 1-8.

S3. M. Wu, M. Peng, Z. Liang, Y. Liu, B. Zhao, D. Li, Y. Wang, J. Zhang, Y. Sun and L. Jiang, *ACS Appl Mater Interfaces*, 2021, **13**, 26989-26997.

S4. X. Zhou, W. Zhang, C. Zhang, Y. Tan, J. Guo, Z. Sun and X. Deng, *ACS Appl Mater Interfaces*, 2020, **12**, 11232-11239.

S5. T. Zhong, H. Li, T. Zhao, H. Guan, L. Xing and X. Xue, *Journal of Materials Science & Technology*, 2021, **76**, 33-40.

S6. F. Zhao, H. Cheng, Z. Zhang, L. Jiang and L. Qu, *Adv Mater*, 2015, **27**, 4351-4357.

S7. F. Zhao, Y. Liang, H. Cheng, L. Jiang and L. Qu, *Energy & Environmental Science*, 2016, **9**, 912-916.

S8. C. Yang, Y. Huang, H. Cheng, L. Jiang and L. Qu, *Adv Mater*, 2019, **31**, e1805705.

S9. Y. Liang, F. Zhao, Z. Cheng, Q. Zhou, H. Shao, L. Jiang and L. Qu, *Nano Energy*, 2017, **32**, 329-335.

S10. H. Wang, H. Cheng, Y. Huang, C. Yang, D. Wang, C. Li and L. Qu, *Nano Energy*, 2020, **67**.

S11. J. Yin, X. Li, J. Yu, Z. Zhang, J. Zhou and W. Guo, *Nat Nanotechnol*, 2014, **9**, 378-383.

S12. S. Yang, Y. Su, Y. Xu, Q. Wu, Y. Zhang, M. B. Raschke, M. Ren, Y. Chen, J. Wang, W. Guo, Y. Ron Shen and C. Tian, *J Am Chem Soc*, 2018, **140**, 13746-13752.

S13. J. K. Moon, J. Jeong, D. Lee and H. K. Pak, *Nat Commun*, 2013, **4**, 1487.

S14. W. Xu, H. Zheng, Y. Liu, X. Zhou, C. Zhang, Y. Song, X. Deng, M. Leung, Z. Yang, R. X. Xu, Z. L. Wang, X. C. Zeng and Z. Wang, *Nature*, 2020, **578**, 392-396.