

Electronic Supplementary Information

**A biomass-derived, all-day-round solar evaporation platform for harvesting clean water from microplastic pollution**

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## **S1. Experimental Section**

### **S1.1 Materials**

Graphite powder (99.95%, 325 meshes) was purchased from XFNANO Tech. Co., Ltd. Hydrogen peroxide ( $\text{H}_2\text{O}_2$ ,  $\approx 30$  wt%) was obtained from the Alfa Aesar. Ammonium molybdate tetrahydrate ( $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$ , 99.9%), thioacetamide (99.9%), polyethylene glycol (PEG-4000) and other chemicals were all gotten from the Sinopharm Chemical Reagent Co., Ltd. The water used in all experiments was filtered through a Millipore filtration system with a resistivity of  $18.2 \text{ M}\Omega\cdot\text{cm}$ . The cotton rolls (product code: C6431) were purchased from Aladdin.

### **S1.2 Fabrication of RGO/Cotton with gradient microstructures**

Graphene oxide (GO) was prepared from the modified Hummer's method.<sup>[1]</sup> The cotton roll was washed with ethanol to remove its surface impurities. After dried at  $90^\circ\text{C}$  for 2 h, the cotton roll was cut into cubic sponge ( $1\text{cm} \times 1\text{cm} \times 1\text{cm}$ ). GO aqueous solution ( $1.2 \text{ mg}\cdot\text{mL}^{-1}$ ) was mixed with L-ascorbic acid (L-AA,  $5 \text{ mg}\cdot\text{mL}^{-1}$ ) by stirring for 15 min at  $280 \text{ rpm}\cdot\text{min}^{-1}$ . Then, the cotton sponges were immersed in GO/L-AA solution (6 mL) for 30 min and heated at  $75^\circ\text{C}$  for 6 h in a sealed bottle. The resultant reduced graphene oxide (RGO)/cotton sponge was further carefully washed by water and ethanol for 3 times. This ethanol-wetted RGO/Cotton sponge was stretched perpendicularly to the cotton layers with a width elongation of 50% and fixed by clamps at two ends for 30 min. The stretched RGO/Cotton was carefully set in an oven and dried for 4 h to induce gradient spacings among cotton layers. The RGO/Cotton sponges without gradient structures were obtained from the same process without stretching. The GO contents discussed in results were mass ratio between the input GO and the cotton sponge in GO/L-AA solution. The real loading mass of RGO within RGO/Cotton sponge after reduction and drying for 1 wt%-GO input was 0.95 wt%, for 7 wt%-GO input was 3.85 wt%, and for 15 wt%-GO input was 5.91 wt%.

### **S1.3 Fabrication of $\text{MoS}_2$ /RGO/Cotton**

The specific preparation method of  $\text{MoS}_2$  was as follows: 1.07 g  $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$  and 770 mg thioacetamide was dissolved in water by magnetic stirring for 30 min. The mixture was

hydrothermally reacted at 200 °C for 24 h in 25 mL Teflon kettle. The MoS<sub>2</sub> products were washed and centrifugated (7000 rpm·min<sup>-1</sup>) with water and ethanol for three times, following by re-dispersing in ethanol with concentration of 1mg·mL<sup>-1</sup>. Then, the RGO/Cotton sponges without stretching were immersed in MoS<sub>2</sub> solution for 30 min and heated at 100 °C for 2 h to obtain the RGO/Cotton loaded with MoS<sub>2</sub>. The resultant MoS<sub>2</sub>/RGO/cotton sponge was further carefully washed by water and ethanol for 3 times, following by stretching and drying for 4 h. The sponge was stretched as the method mentioned before.

#### **S1.4 Fabrication of Polyethylene glycol (PEG)/RGO/Cotton**

PEG-4000 solution with a weight content of 60% was prepared by solving PEG in water under 75 °C in an oven and stirring by glass bar. The RGO/Cotton sponge without stretching was immersed in the as-prepared PEG and placed at 75 °C for 1 h for coating PEG onto the RGO/Cotton sponge. The loading mass of PEG within the composite sponge was measured as 7 wt%. Then, the sponge was carefully cooled at 25 °C for 1h, to obtain the solidified PEG/RGO/Cotton sponge. The PEG/RGO/Cotton sponge was stretched as the method mentioned before to induce the gradient microchannels. Finally, PEG/RGO/Cotton sponge was cooled at 25 °C for 1 h.

#### **S1.5 Simulations of the temperature distributions of RGO/Cotton and PEG/RGO/Cotton sponges**

Results were simulated by the Heat Transfer port, Steady-State interface in COMSOL Multiphysics 5.5. The size (100 μm × 200 μm × 100 μm), density, enthalpy change, and related parameters were measured from the tests, including SEM characterization, differential scanning calorimetry (DSC) test, thermal conductivity test, and so on. The source of heat in models was selected as the solar irradiation, which used the External Radiation Source feature (2 kW·m<sup>-2</sup>). The convective heat flux boundary condition used a bulk heat-transfer-coefficient of 20 W·m<sup>-2</sup>·K<sup>-1</sup> for all exposed surfaces. In the simulation of phase change, solar irradiation was set at first to investigate the temperature-increment under the competition process between photothermal conversion of RGO and the endothermal process of PEG. Then, the solar irradiation interface was

removed to explore the temperature-variation induced by the exothermic process of PEG and the natural convection heat dissipation.

### S1.6 Estimation of evaporation performance of sponges

The sponges with the thickness of 1.5 cm were fixed on a frame for further ensuring the gradient structure and floated on the water surface contained by a flat mouth beaker (Fig. S26). The relative humidity was 86–91% and the surrounding temperature was 21.2–25.4 °C. The evaporation was conducted under the irradiation ( $2 \text{ kW}\cdot\text{m}^{-2}$ ) of a Xenon lamp (CEL-HXF300, Beijing Aulight Co., Ltd.) for 1h. The light-density was measured by an optical power meter (S314C, THORLABS). The water-mass change was measured by the electronic microbalance (XP26, Mettler Toledo) during the evaporation time. Evaporation rate was calculated by subtracting the dark evaporation rate from the total evaporation rate.<sup>[2, 3]</sup> Solar-to-vapor conversion efficiency ( $\eta$ ) was calculated from the following equation S1:

$$\eta = \frac{m \cdot h_{LV}}{\rho L} \quad (\text{S1})$$

where  $m$  is the water evaporation rate ( $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ),  $h_{LV}$  is the total enthalpy of water-vapor phase conversion, including sensible heat and phase conversion enthalpy ( $\text{J}\cdot\text{kg}^{-1}$ ), and  $\rho_L$  is the light power density ( $\text{W}\cdot\text{m}^{-2}$ ).

Commercial polyethylene (PE) fibers with the diameter of 750  $\mu\text{m}$  and length of 3 mm were used as microplastic pollutions in our experiments, which were purchased from Shenzhen Teli Chemical Fiber Co., Ltd. The PE fibers with input density of 2.26 numbers per square centimeter were floated on water under the photothermal sponge. After evaporation ( $2 \text{ kW}\cdot\text{m}^{-2}$ , 1h), the microplastics were collected by filtration through a cellulose membrane (pore size: 0.8  $\mu\text{m}$ ), then washed with ethanol/ultra-pure water several times and dried for further characterizations (*i.e.*, weighting, FTIR test). The contact surface between PE and  $\text{MoS}_2/\text{RGO}/\text{Cotton}$  surface after a short evaporation time (20 min), as well as other collected PE microfibers were observed by an optical microscope (BX53M, Olympus). The  $\text{CO}_2$  test was conducted under aeration of  $\text{O}_2$  for better

venting gas during evaporation. The precipitate in Ba(OH)<sub>2</sub> after CO<sub>2</sub> test was collected by centrifugation (8000 rpm·min<sup>-1</sup>) for 15 min.

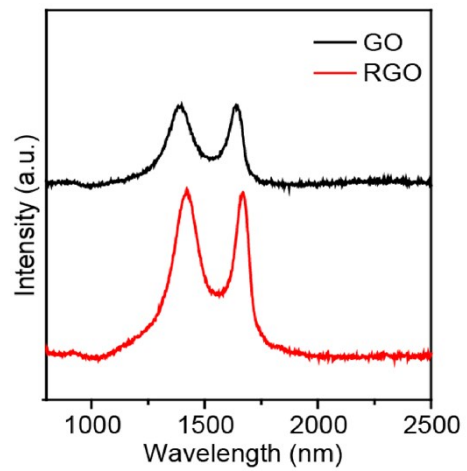
The day-night-cycle water evaporation performance was tested by applying or removing the light-irradiation on top surface of sponges. The evaporation rate normalized to the day time (*rate*) was estimated by the following equation S2:

$$rate = \frac{rate_L \times time_L + rate_D \times time_D}{time_L + time_D} \quad (S2)$$

where *rate<sub>L</sub>* is the evaporation rate under light irradiation (one sun, 1 h), the rate is 1.83 kg·m<sup>-2</sup>·h<sup>-1</sup> for PEG/RGO/Cotton and 1.80 kg·m<sup>-2</sup>·h<sup>-1</sup> for RGO/Cotton counterpart; *rate<sub>D</sub>* is the evaporation rate under dark (zero sun, 1 h) after pre-irradiation (one sun, 1 h), the rate is 1.37 kg·m<sup>-2</sup>·h<sup>-1</sup> for PEG/RGO/Cotton and 0.32 kg·m<sup>-2</sup>·h<sup>-1</sup> for RGO/Cotton counterpart; *time<sub>L</sub>* is the recorded day-time in the Jiangning District, Nanjing Province, China (*i.e.*, 13.53 h, from 5:20 a.m. of April 7<sup>th</sup> to 18:52 p.m. of April 7<sup>th</sup>); *time<sub>D</sub>* is the recorded night-time in Jiangning District, Nanjing Province, China (*i.e.*, 10.45 h, from 18:52 p.m. of April 7<sup>th</sup> to 5:19 a.m. of April 8<sup>th</sup>).

### S1.7 Characterizations

The field-emission scanning electron microscopy (SEM) image was obtained by Inspect F50 (FEI). The Raman spectra were obtained from the Raman microscope (DXR2, Thermo Fisher Scientific) with a laser of 532 nm. The DSC spectra were tested by DSC8000 (PerkinElmer). The contact angle was acquired from the contact angle instrument (OCA 15 plus, Dataphysics). The ζ-potential was tested on ζ-potential Nano ZS90 at room temperature. The FTIR spectra of PE microplastics were collected on the Spotlight 200i FTIR microscopy system by focusing the single PE microfiber or microparticle. The detection of PEG in water samples was conducted on a MALDI-TOF/TOF mass spectrometer with 5 GS·s<sup>-1</sup> 10 bit digitizer (UltrafleXtreme, Bruker). The compressive performance of the sponge was tested by using an electronic universal testing machine (STD-500, Yishite Instruments Co., Ltd.).

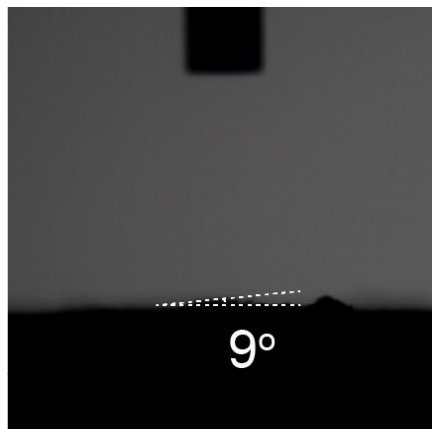


**Fig. S1** The Raman spectra of RGO and pristine GO.

### Supplementary Note

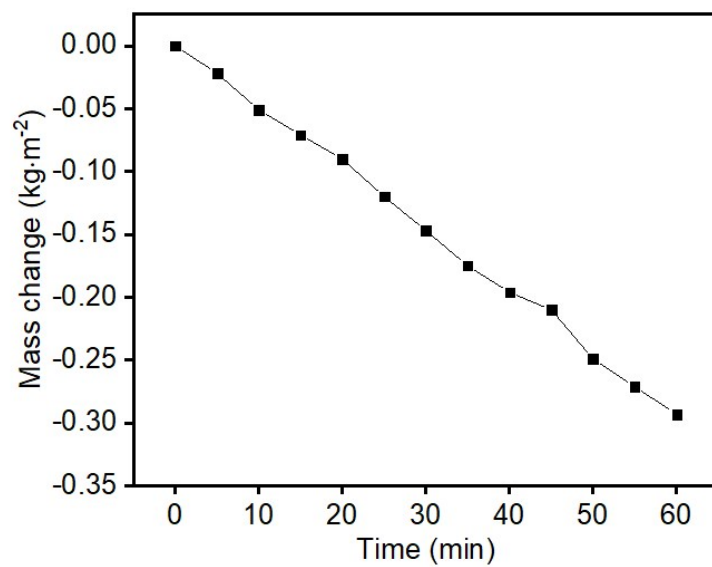
**Note S1.** Comparison of cost between RGO/Cotton and common RGO aerogel with same volume ( $1\text{ cm}^3$ ).

For better comparison, the GO (code: 763713) purchased in Sigma-Aldrich company with a cost of \$ 0.905 was used as the same precursor. The amount of GO input for RGO/Cotton was 7.2 mg ( $1.2\text{ mg}\cdot\text{mL}^{-1}$ ), while the input for RGO aerogel was assumed as 75 mg ( $5\text{ mg}\cdot\text{mL}^{-1}$ ). The cost of degreasing cotton roll ( $1\text{ cm}^3$ ) was \$ 0.058. Hence, the estimated total cost of RGO/Cotton was low as \$ 6.574, while the cost of RGO aerogel was \$ 67.9.

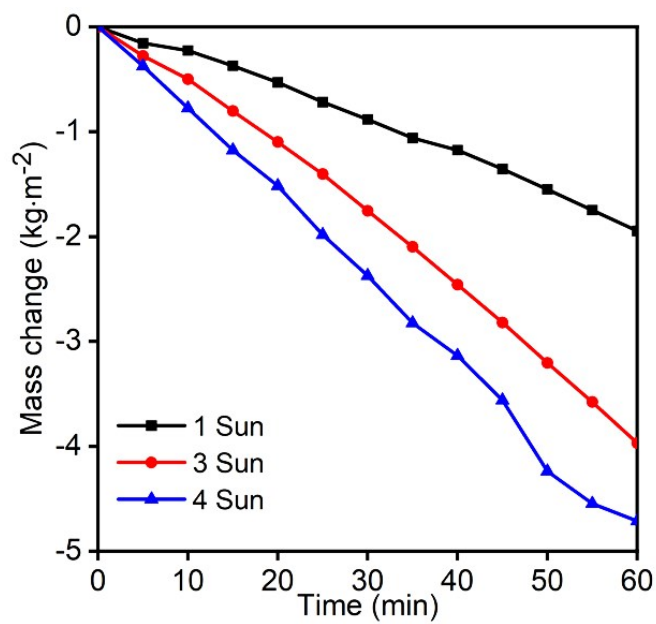


**Fig. S2** Water contact angle on the top surface of initial cotton sponge.





**Fig. S3** The water mass change recording the evaporation performance of 3.85 wt%-RGO/Cotton under dark.

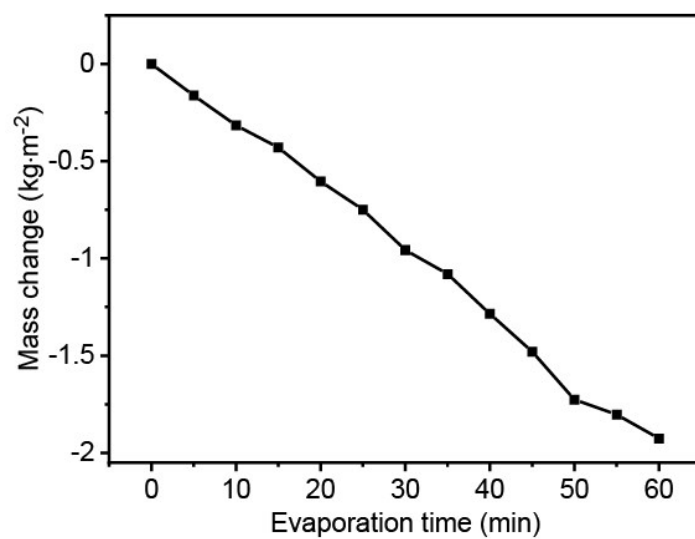


**Fig. S4** The water mass change recording the evaporation performance of 3.85 wt%-RGO/Cotton under different light-intensities.

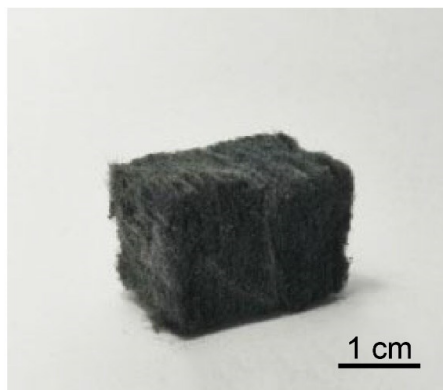
**Table S1.** Comparison of the evaporation rate and photothermal conversion efficiency normalized to the mass of photothermal materials.

Photothermal materials	Evaporation rate ( $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ )	Mass of the photothermal material (mg)	Light density ( $\text{kW}\cdot\text{m}^{-2}$ )	Reference
RGO	1.97	2.93	1	This work
RGO	2.49	2.93	2	This work
RGO	2.25	8.65	1	[4]
RGO	2.33	17.9	1	[5]
RGO	1.25	36.8	2	[6]
RGO	1.48	150	1	[7]
RGO	0.83	23.7	1	[8]

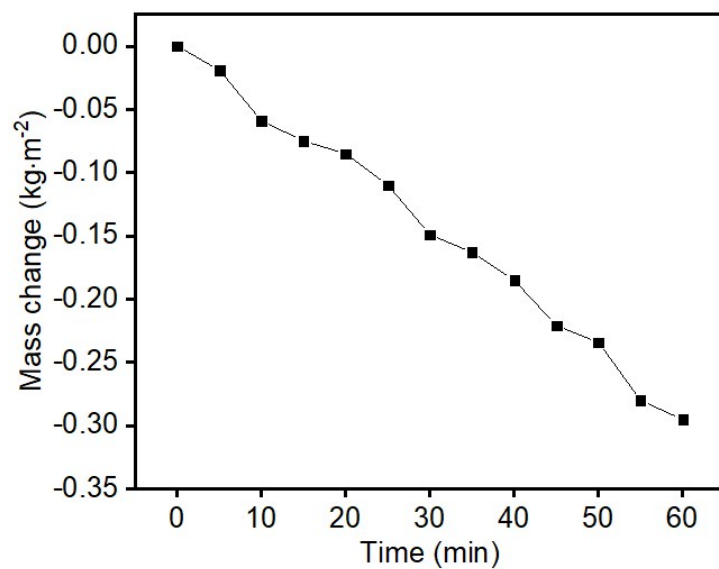
Data were given or calculated from results in the respective references.



**Fig. S5** The water mass change recording the evaporation performance of 5.91 wt%-RGO/Cotton.



**Fig. S6** Digital photo of 3D MoS<sub>2</sub>/RGO/Cotton sponge.



**Fig. S7** The water mass change recording the evaporation performance of MoS<sub>2</sub>/RGO/Cotton under dark.

## Supplementary Note

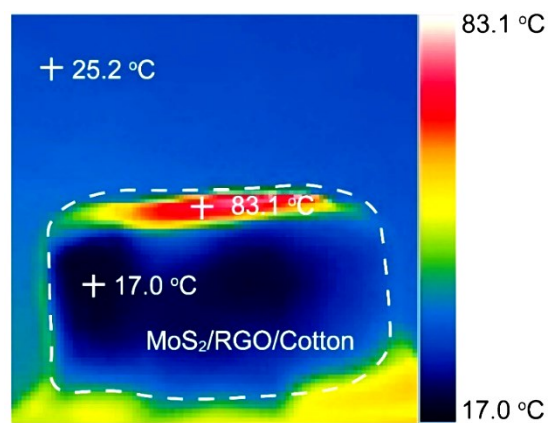
**Note S2.** The calculation of net energy gained from environment and the energy balance of 3D MoS<sub>2</sub>/RGO/Cotton sponge during photothermal evaporation.

The net energy ( $E_s$ ) gained from environment can be estimated by the following equation S3:<sup>[9, 10]</sup>

$$E_s = -A_{\text{top}} \cdot \varepsilon \cdot \sigma \cdot (T_{\text{top}}^4 - T_s^4) - A_{\text{side}} \cdot \varepsilon \cdot \sigma \cdot (T_{\text{side}}^4 - T_s^4) - A_{\text{top}} \cdot h \cdot (T_{\text{top}} - T_s) - A_{\text{side}} \cdot h \cdot (T_{\text{side}} - T_s) \quad (3)$$

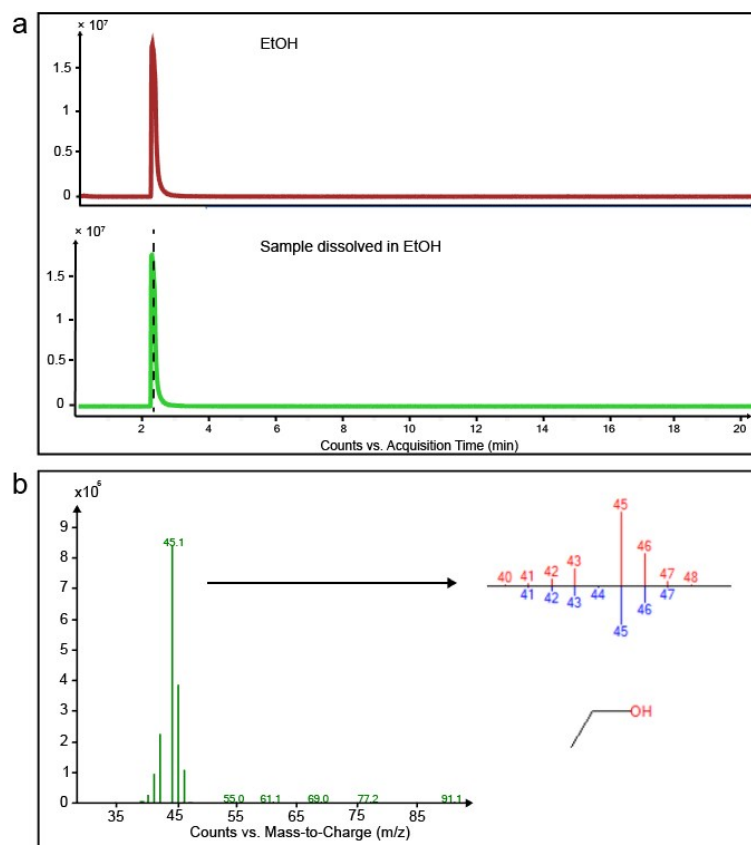
where  $A_{\text{top}}$  is the top surface area,  $A_{\text{side}}$  is the side wall surface area;  $T_{\text{top}}$  is the average temperature of top surface,  $T_{\text{side}}$  is the average temperature of side surface,  $T_s$  is the surrounding temperature, which are obtained from the infrared image in Fig. S8;  $\varepsilon$  is emissivity of the graphene absorber,  $\sigma$  is the Stefan–Boltzmann constant, and  $h$  is the average convection heat transfer coefficient.

As illustrated in Fig. S8, the exposed side surface has a lower temperature than that of surrounding, and thus can gain energy from the surrounding.<sup>[9, 10]</sup> According to the energy balance of our evaporation system in equation S3, the radiation loss from the top surface of MoS<sub>2</sub>/RGO/Cotton is 0.036 W, while the convection loss from the top surface of RGO/Cotton is 0.051 W. The radiation energy gain of the MoS<sub>2</sub>/RGO/Cotton's side surface from surrounding environment is 0.221 W, while the convection energy gain of the MoS<sub>2</sub>/RGO/Cotton's side surface from surrounding environment is 0.425 W. Therefore, the net energy of MoS<sub>2</sub>/RGO/Cotton evaporator gained from environment is 176 mW, which is 14.6% of the input light energy (1200 mW).

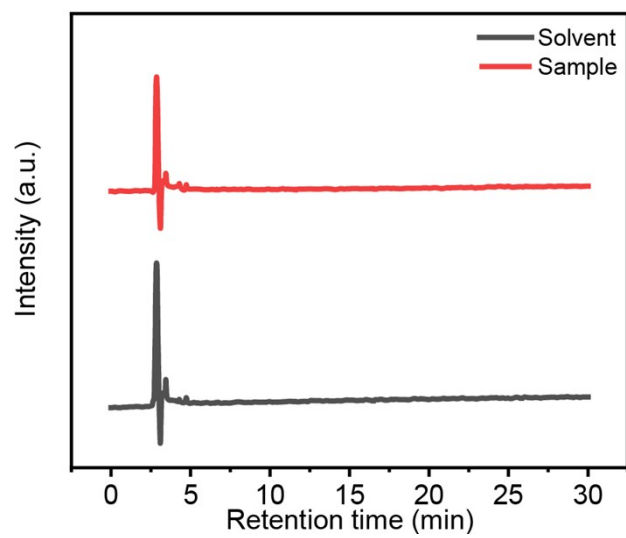


**Fig. S8** The infrared image of MoS<sub>2</sub>/RGO/Cotton sponge during water evaporation. The white lines highlight the boundary of MoS<sub>2</sub>/RGO/Cotton.

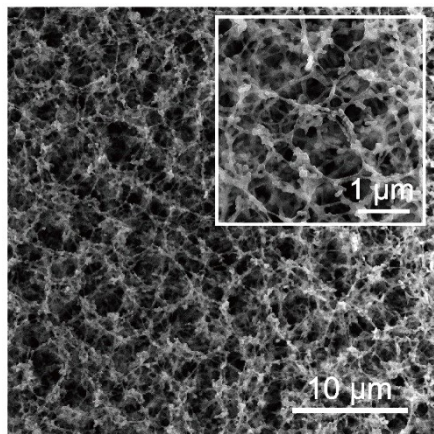




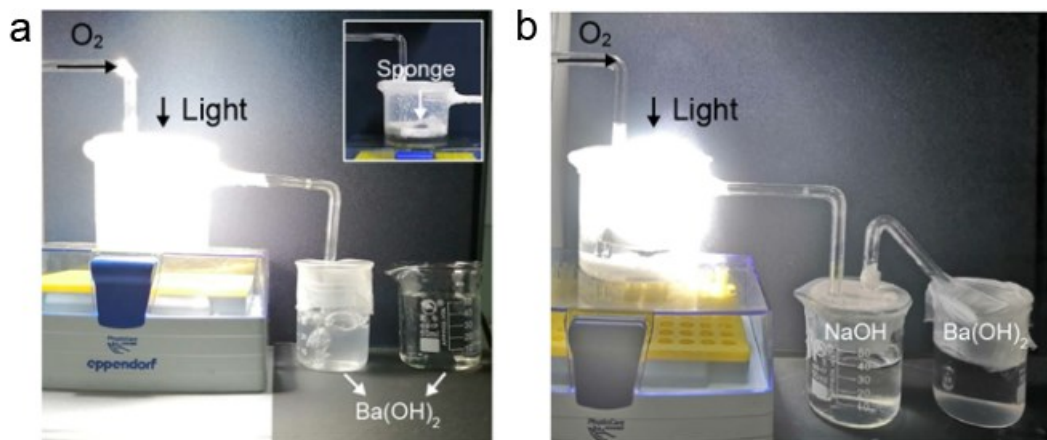
**Fig. S9** GC-MS spectra of the water sample evaporated from MoS<sub>2</sub>/RGO/ Cotton sponge after the microplastic degradation for 1 h.



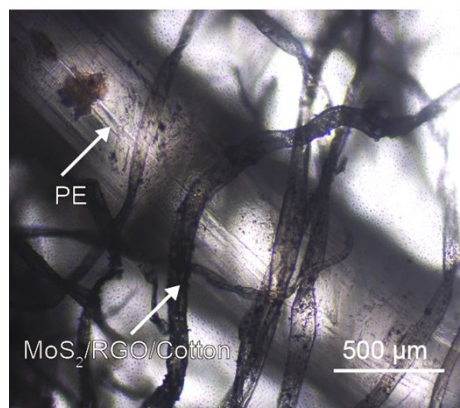
**Fig. S10** HPLC spectra of water sample underlying RGO/MoS<sub>2</sub>/Cotton sponge after the microplastic degradation for 1 h.



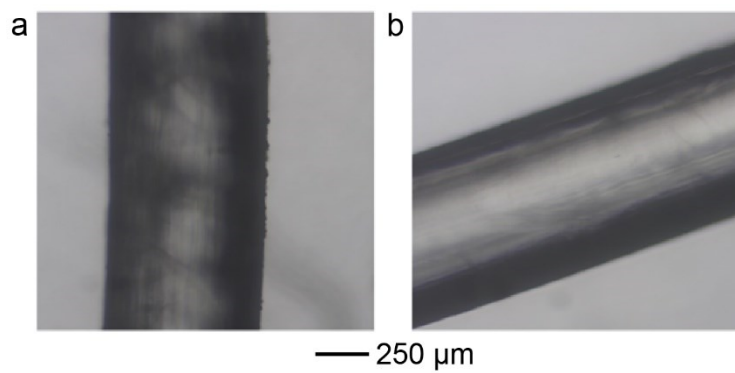
**Fig. S11** SEM image of the cellulose membrane after filtering the evaporation water which harvested by RGO/Cotton from the microplastic-polluted water.



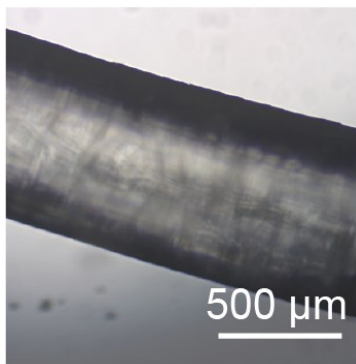
**Fig. S12** (a) Optical image of  $Ba(OH)_2$  showing the existence of  $CO_2$  which released from the reaction system during the microplastic degradation. The inset showing the sponge in the setup device. (b) Optical image shows the successful absorption of  $CO_2$  by NaOH with zero  $CO_2$  emission.



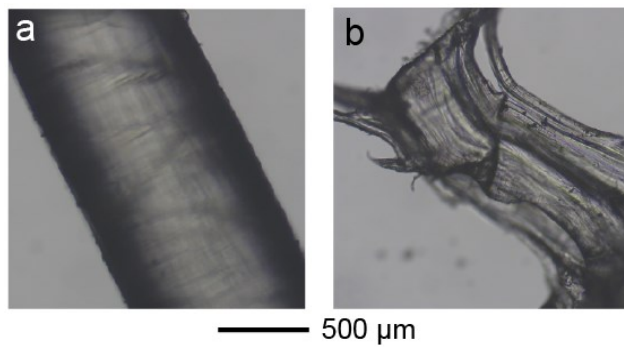
**Fig. S13** PE microplastics intertwined by MoS<sub>2</sub>/RGO/Cotton microfibers.



**Fig. S14** PE microplastic degraded by RGO/MoS<sub>2</sub>/Cotton after quenching of (a) OH $\cdot$  and (b) O<sub>2</sub> $\cdot^-$ .

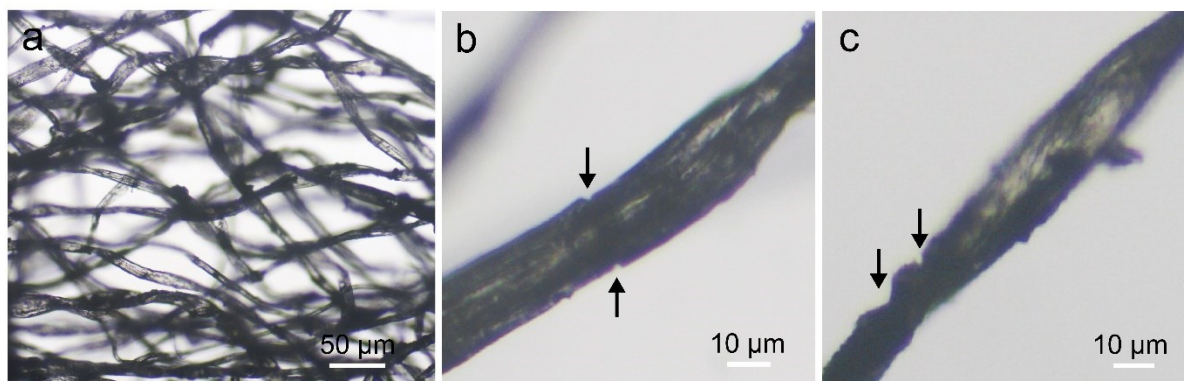


**Fig. S15** PE microplastics immersed in 95 °C water for 1 h, without light irradiation. The observed PE products were carefully collected by cellulose filter paper (pore size: 0.8 μm) and observed by optical microscope.

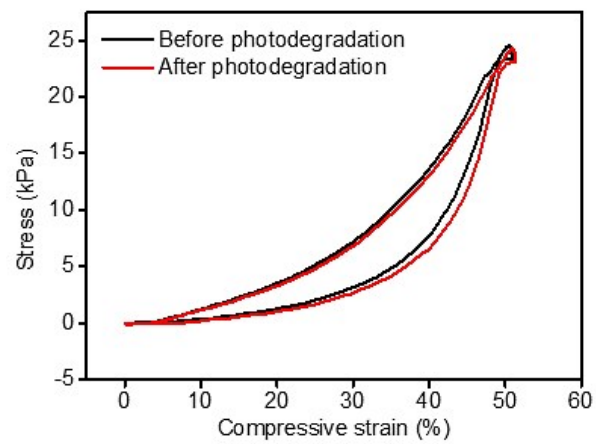


**Fig. S16** (a) PE microplastics before and (b) after degraded by MoS<sub>2</sub>/RGO/Cotton in 95 °C water, without light irradiation.

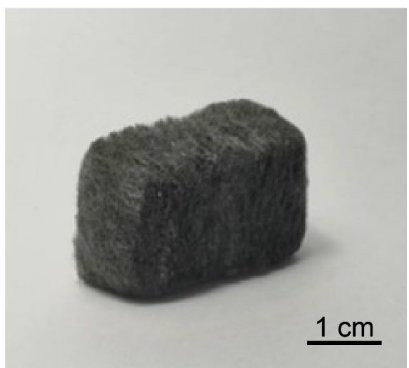




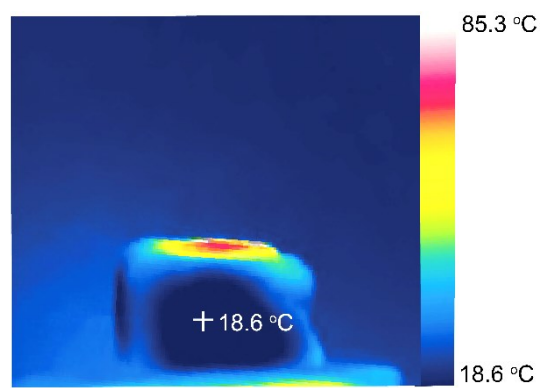
**Fig. S17** (a) Well-maintained cotton-based framework and (b, c) the etched cotton-based microfibers after PE degradation under light irradiation (1 h).



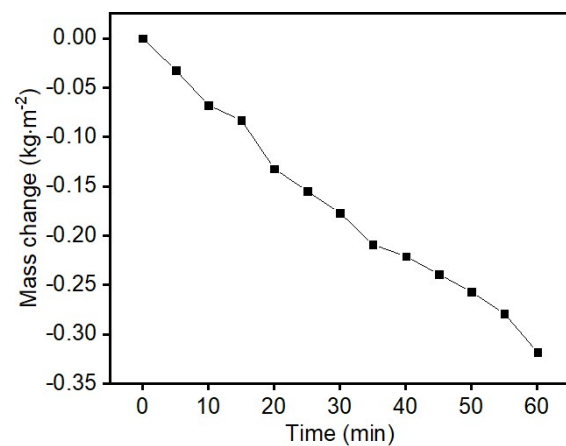
**Fig. S18** Mechanical properties of MoS<sub>2</sub>/RGO/Cotton sponge before and after the photodegradation of PE under light irradiation (1 h).



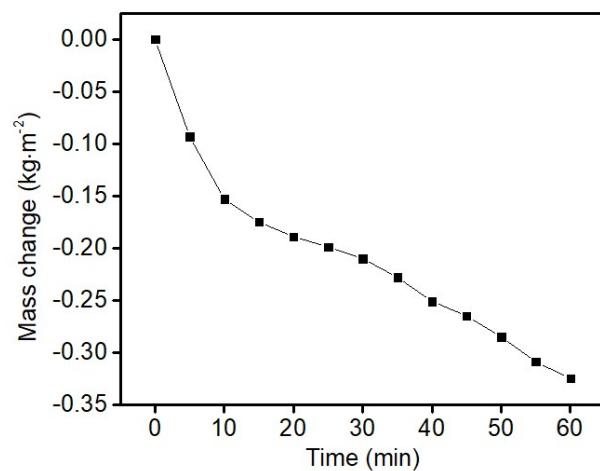
**Fig. S19** Digital photo of 3D PEG/RGO/Cotton sponge.



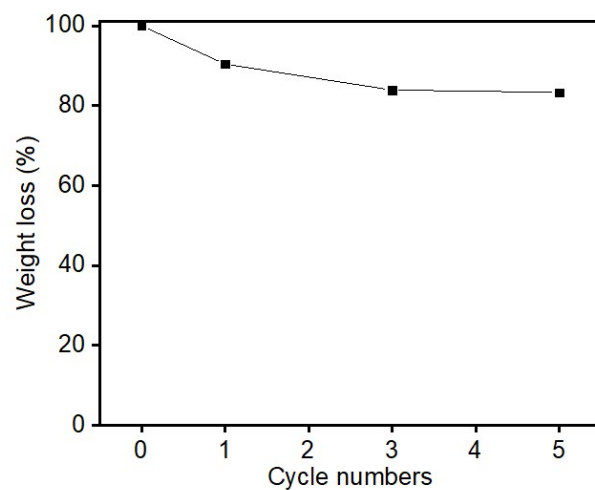
**Fig. S20** The infrared image of PEG/RGO/Cotton sponge during water evaporation.



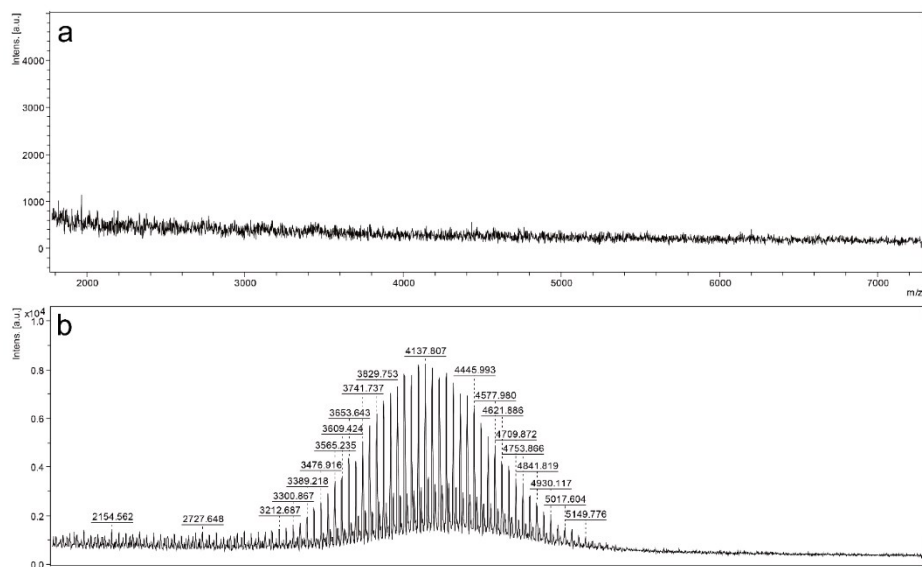
**Fig. S21** The water mass change recording the evaporation performance of PEG/RGO/Cotton under dark without pre-irradiation.



**Fig. S22** The water mass change recording the evaporation performance of RGO/Cotton under dark with pre-irradiation.

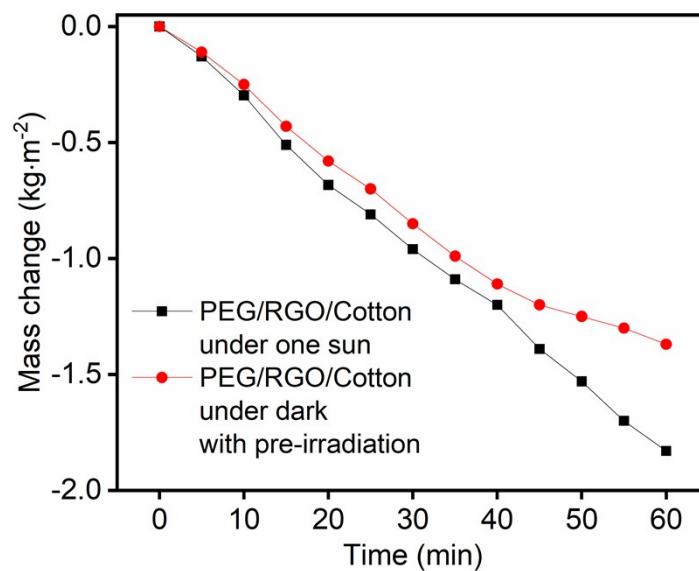


**Fig. S23** The weight loss of PEG loading after cycling evaporation. The weight loss here is the percentage relative to the original loading of PEG on RGO/Cotton sponge (7%).

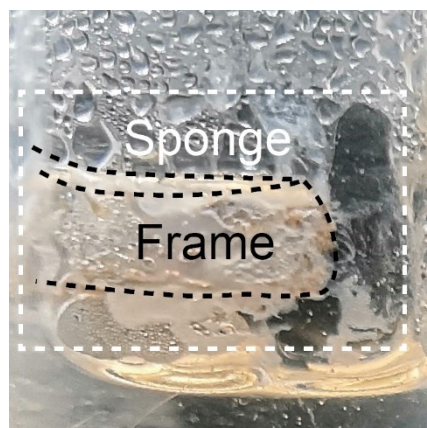


**Fig. S24** (a) The MALDI-TOF mass spectrometer spectra of the collected evaporation water and (b) the underlying water after cycling evaporation of PEG/RGO/Cotton.





**Fig. S25** The water mass change recording the evaporation performance of PEG/RGO/Cotton under light irradiation (one sun), and under dark with pre-irradiation.



**Fig. S26** The magnified digital photo of floating device.

## References

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