Supplementary information

Full dimensional water evaporation on macroporous vertically-aligned graphene pillar array under sun

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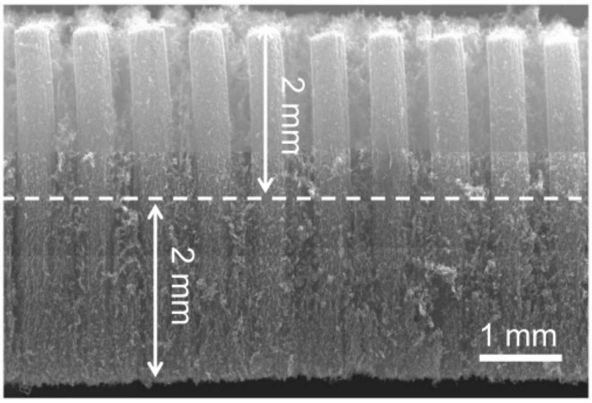

Fig. S1 SEM image of HOPGF (D = 400 μm, S = 300 μm). The thickness of HOPGF is 4 mm, and the height of vertically-aligned pillar array is 2mm.

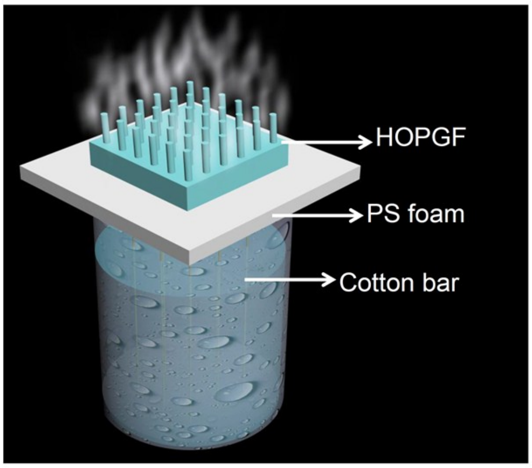

Fig. S2 Schematic illustration of the SSG device. The SSG device includes a thermally insulating layer made of polystyrene (PS) foam and hydrophilic cotton bars wicking water. HOPGF is placed on the commercial PS, and the entire structure is allowed to put on the beaker with only the carton bars in direct contact with bulk water.

Fig. S3 SEM images of HOPGF with different D and S. (a) Top and cross-sectional SEM images of HOPGF (S = 100) with the D of 300, 400, 500, 600 and 800 μm. (b) Top and cross-sectional SEM images of HOPGF (S = 200) with the D of 300, 400, 500, 600 and 800 μm. (c) Top and cross-sectional SEM images of HOPGF (S = 300) with the D of 300, 400, 500, 600 and 800 μm. (d) Top and cross-sectional SEM images of HOPGF (S = 400) with the D of 300, 400, 500, 600 and 800 μm.

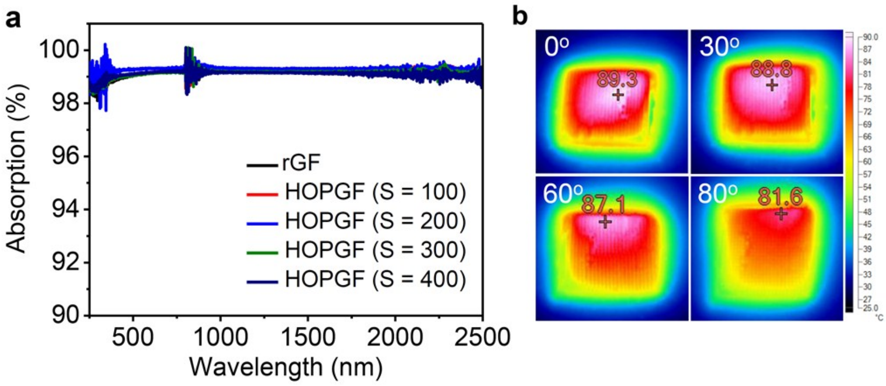

Fig. S4 Sunlight absorption property of HOPGF in dry state with different D and S. (a) Absorption spectra of rGF, and HOPGF (D = 400 μm) with the S of 100, 200, 300 and 400 μm in the dry state, respectively. (b) IR image of HOPGF (D = 400 μm, S = 300 μm) in the dry state at varied incident angles (0^o, 30^o, 60^o and 80^o) after reaching temperature stable state under solar illumination of 1 sun.

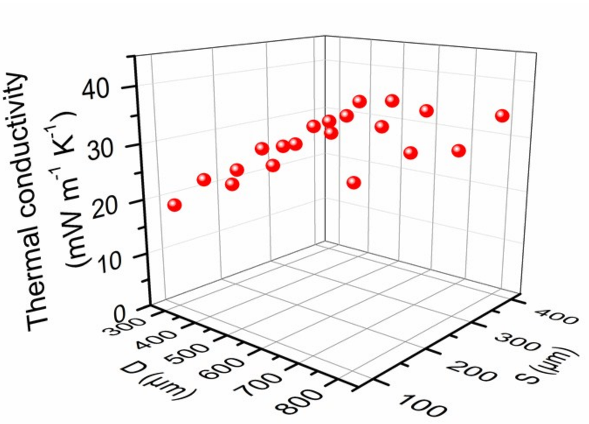

Fig. S5 Thermal conductivity of HOPGF with different D and S. The air filling between the pillars serves as the thermal barriers, resulting in HOPGF with low thermal conductivity (18–35 mW m^{-1} K^{-1}), which can effectively reduce heat loss toward buck water and ambient environment.

The thermal conductivity (κ) is calculated by $\kappa=\rho*\alpha*c$, where ρ is the density, α is the thermal diffusivity, which could be measured by a Netzsch LFA 467 Nanoflash, and c is the specific heat capacity.

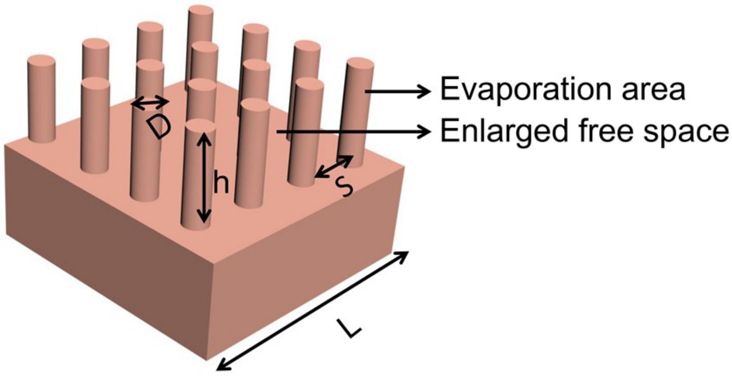

Fig. S6 Schematic illustration of HOPGF, where *L* is the length of HOPGF, *D* is the diameter of graphene pillars, *S* is the spacing between pillars, and *h* is the height of vertically-aligned pillar array.

Estimation the evaporation area (S_{area}) and enlarged free space ($\triangle F$) of HOPGF with different D and S:

The solar absorbers used in the SSG experiments are cut into square shapes with the same size (20 mm × 20 mm × 4 mm) by high precision laser, and the height of vertically-aligned pillar array is 2 mm.

The number (n) of graphene pillar is calculated by the formula:

$$n=[L/(D+S)]^2 \quad \text{(S1)}$$

The evaporation area (S_{area}) is expressed by the formula:

$$\begin{aligned} S_{area} &= L^2+n^2\times \pi Dh \\ &= L^2+\pi Dh[L/(D+S)]^2 \quad \text{(S2)} \end{aligned}$$

The enlarged free space ($\triangle F$) is calculated by the formula:

$$\begin{aligned} \triangle F &= hL^2-n^2\times \pi D^2h/4 \\ &= hL^2-\pi hD^2L^2/4(D+S)^2 \quad \text{(S3)} \end{aligned}$$

Table S1 The evaporation area (S_{area}) and enlarged free space ($\triangle F$) calculation of HOPGF (L = 20 mm, h = 2 mm) with different D and S.

Sample (μm)	Evaporation area (S_{area}, mm^2)	Enlarged free space ($\triangle F$, mm^3)
Original rGF	400	0
HOPGF (D = 300, S = 100)	5110	517.4
HOPGF (D = 300, S = 200)	3414	573.9
HOPGF (D = 300, S = 300)	2492	643.0
HOPGF (D = 300, S = 400)	1936	684.7
HOPGF (D = 400, S = 100)	4419	398.1
HOPGF (D = 400, S = 200)	3192	520.9
HOPGF (D = 400, S = 300)	2452	594.9
HOPGF (D = 400, S = 400)	1970	643
HOPGF (D = 500, S = 100)	3888	364.0
HOPGF (D = 500, S = 200)	2964	479.6
HOPGF (D = 500, S = 300)	2364	554.7
HOPGF (D = 500, S = 400)	1952	606.2
HOPGF (D = 600, S = 100)	3476	338.7
HOPGF (D = 600, S = 200)	2756	446.7
HOPGF (D = 600, S = 300)	2260	520.9
HOPGF (D = 600, S = 400)	1907	573.9
HOPGF (D = 800, S = 100)	2880	306.3
HOPGF (D = 800, S = 200)	2409	400
HOPGF (D = 800, S = 300)	2060	469.5
HOPGF (D = 800, S = 400)	1796	522.1

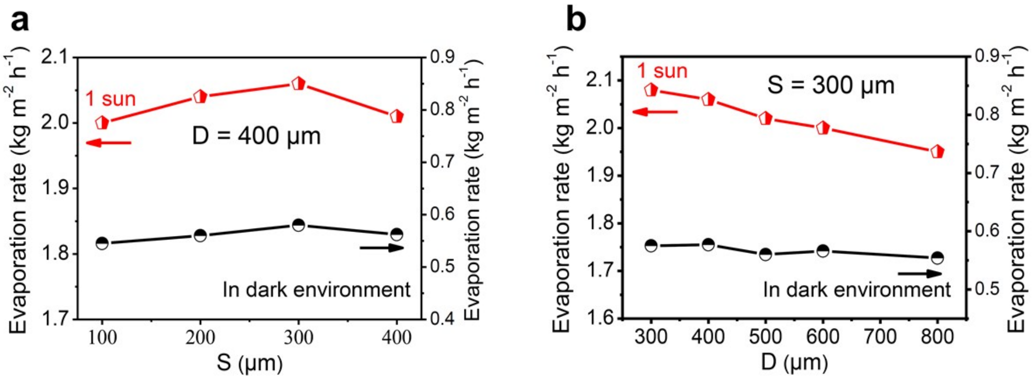

Fig. S7 SSG performance exploration of HOPGF. (a) Water evaporation rate of HOPGF (D = 400 μm) with different S of 100, 200, 300 and 400 μm, respectively. (b) Water evaporation rate of HOPGF (S = 300 μm) with different D of 300, 400, 500, 600 and 800 μm, respectively.

When D is a constant, the increase in S can enlarge area of free space facilitating fast escape of vapor, but will decrease the available evaporation area. Here we take HOPGF (D = 400 μm) with different S as an example to evaluate the SSG performance. With the increase of S, the water evaporation rate increases first and then decreases (Fig. S7a), and the optimized S for HOPGF (D = 400 μm) is 300 μm. Then we choose HOPGF (S = 300 μm) with different D (300, 400, 500, 600 and 800 μm) to measure the water evaporation rate. When D ≥ S, with the increase of D, both the area of evaporation and free space are decreased, resulting in relatively low water evaporation rate (Fig. S7b). As a result, the D and S of HOPGF have the synergistic effect on the performance of SSG.

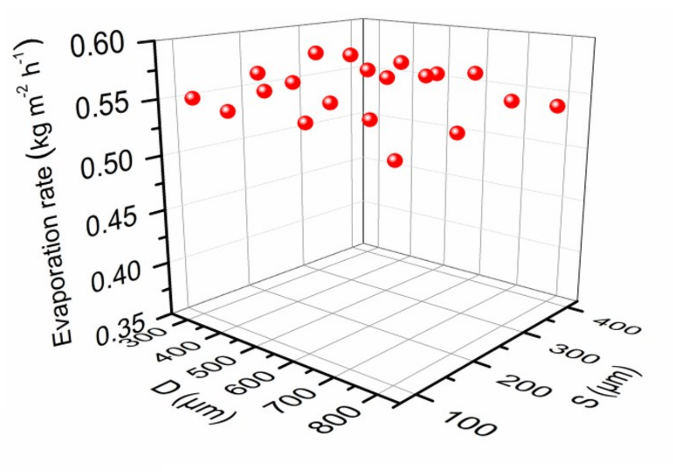

Fig. S8 Water evaporation rates of HOPGF with different D and S in dark environment with the temperature of 25 °C and the humidity of ~20%. The enlarged 3D available evaporation area and free space also promote fast water evaporation in dark environment.

Fig. S9 The photograph of the real device for sewage treatment and desalination.

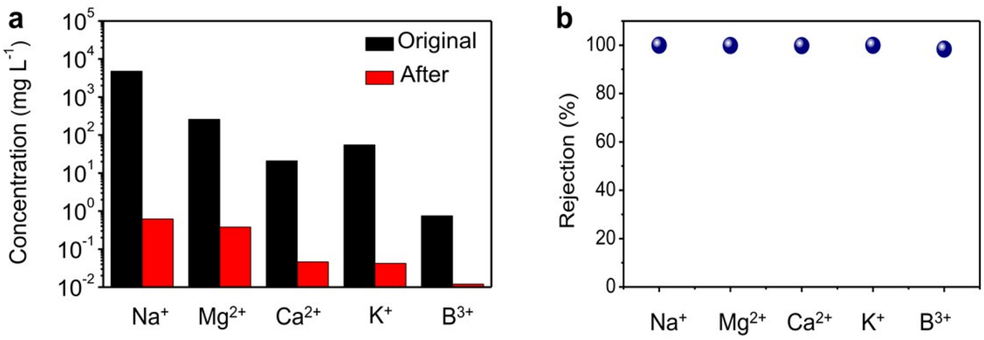

Fig. S10 Purification of seawater. (a) The concentrations of five primary ions in an actual seawater sample (Nanhai Sea) before (original) and after solar purification. (b) The ion rejection of seawater sample undergoing the solar thermal purification.

Steady-state energy balance analysis.

The analysis of energy loss:

The reflection and transmission energy loss of HOPGF in wet state is 1.5–2%.

The analysis of heat loss:

(1) Radiation:

The radiation loss was calculated by the Stefan-Boltzmann equation.

$$\Phi=\varepsilon A\sigma(T_1^4-T_2^4) \tag{S4}$$

Where Φ represents heat flux, ε is the emissivity, and emissivity in this equation is supposed has a maximum emissivity of 1.

A is the surface area, σ is the Stefan-Boltzmann constant, T_1 is the average surface temperature (~38.5 °C) of HOPGF at a steady state condition, and T_2 is the ambient temperature (~32 °C) under the solar illumination of 1 sun. The laser-processed pillar array structure of HOPGF increases the available evaporation area. However, the converted exploitable thermal energy is highly located on HOPGF, and the vertically-aligned pillar arrays within HOPGF show the similar temperatures (~38.5 °C). Therefore, the radiation and convection losses from the enlarged evaporation area can be ignored. The radiation and convection losses mainly come from the surface of HOPGF. According to the equation (1), the radiation heat loss is calculated ~1.2%.

(2) Convection:

The convective heat loss is defined by Newton' law of cooling.

$$Q= hA\Delta T \qquad \text{(S5)}$$

Where Q represents the heat energy, h is the convection heat transfer coefficient, which is about 5 W m^{-2} K as reported, and ΔT is different value between the average surface temperature of HOPGF and the ambient temperature upward the absorber. According to the equation (2), the connection heat loss is measured ~0.9%.

(3) Conduction:

$$Q=Cm\Delta T \qquad \text{(S6)}$$

Where Q is the heat energy, C is the specific heat capacity of water (4.2 kJ $^{o}C^{-1}$ kg^{-1}), m (40 g) is the weight of pure water used in this experiment. ΔT (0.2 ^{o}C) is the average temperature difference of pure water after and before solar illumination under 1 sun after 1 h. The conduction loss is calculated ~0.9%.

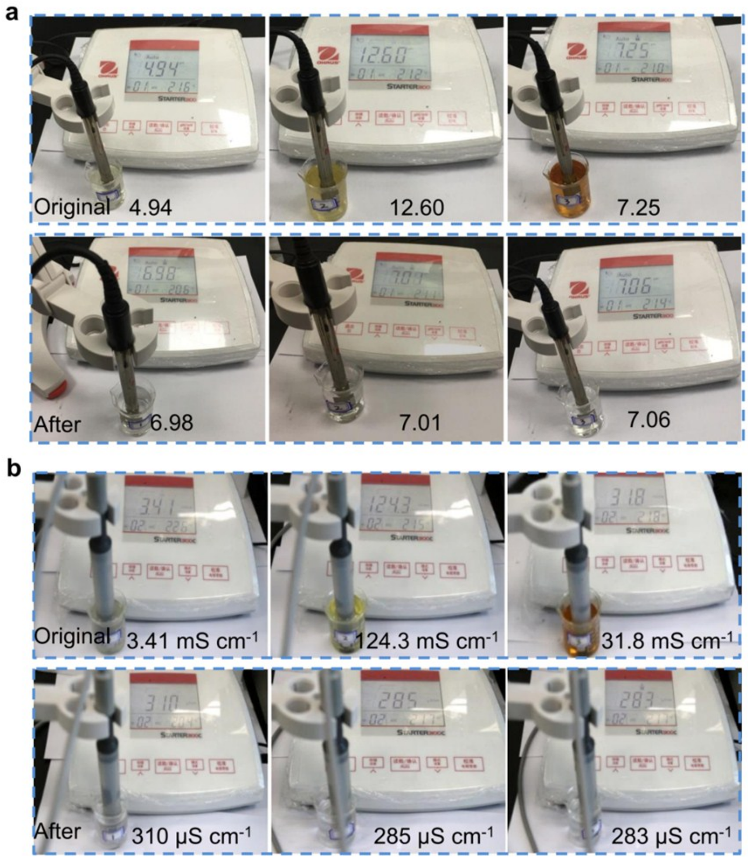

Fig. S11 pH and conductivity measurements. (a) The pH value of three wastewater samples before and after purification (1 sun), respectively. (b) The conductivity measurement of three wastewater samples before and after purification (1 sun), respectively.

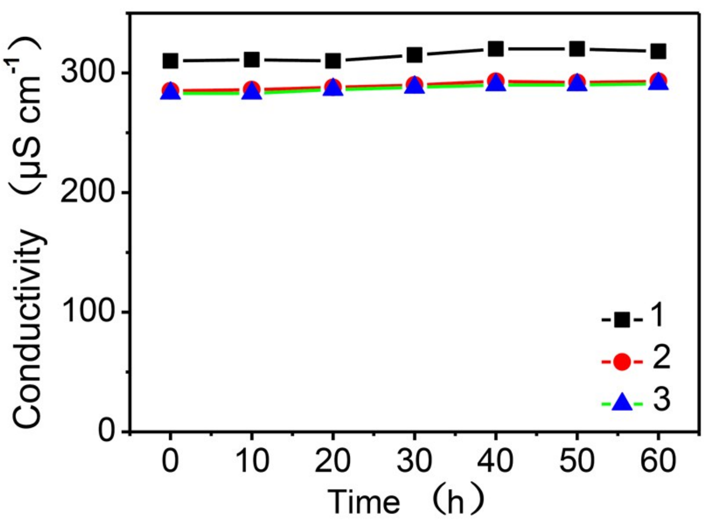

Fig. S12 The duration test of the HOPGF based on a continuous solar purification of three wastewater samples for 60 h (1 sun).

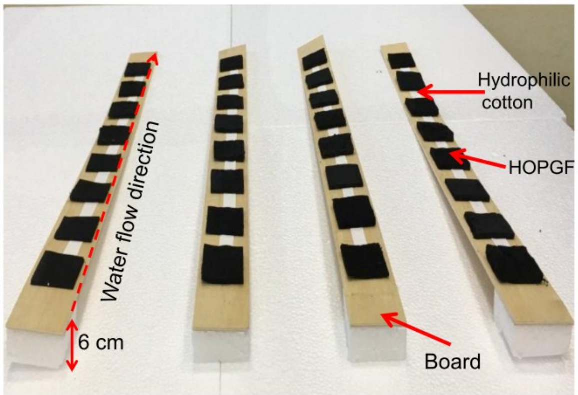

Fig. S13 Photograph of HOPGF array. The total area of HOPGF arrays (4 × 4 cm^2 for each unit) is 512 cm^2.