## **Supplementary Information**

A dual-crosslinked macroporous aerogel with enhanced mechanical durability for efficient solar-driven desalination of seawater and wastewater

**Pictures of supporting materials** 

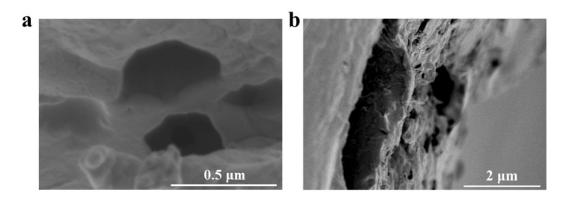


Fig. S1 (a-b) The pore structure of several microns of gelatin-CNF-CNT aerogel.

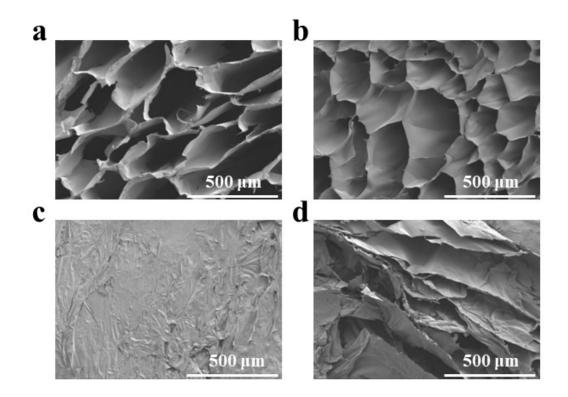


Fig. S2 SEM images of (a) cross section and (b) longitudinal section of pure gelatin aerogel. SEM images of (c) cross section and (d) longitudinal section of pure CNF

aerogel.

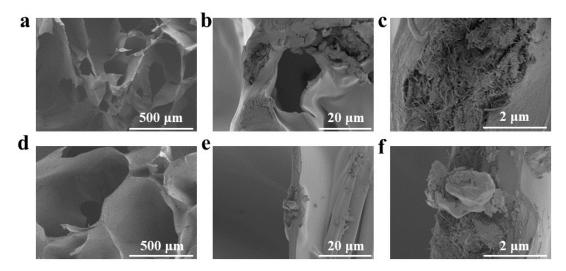


Fig. S3 SEM images of (a-c) cross section and (d-f) longitudinal section of gelatin-CNT aerogel.

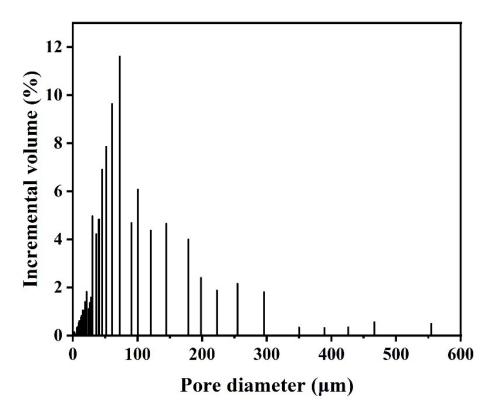


Fig. S4 Pore size and distribution of pure gelatin aerogel (2.5 wt%).

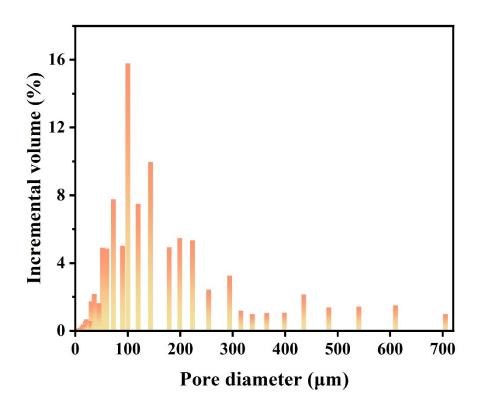


Fig. S5 Pore size and distribution of gelatin-CNT aerogel.

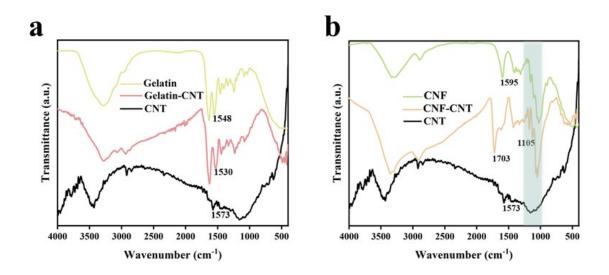


Fig. S6 FT-IR curves of (a) gelatin aerogel, gelatin-CNT aerogel and pure CNT (b) CNF aerogel, CNF-CNT aerogel and pure CNT.

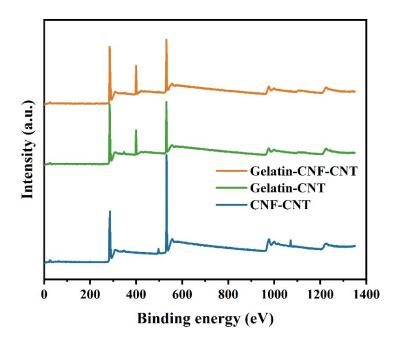


Fig. S7 XPS curves of gelatin-CNF-CNT, gelatin-CNT and CNF-CNT aerogel.

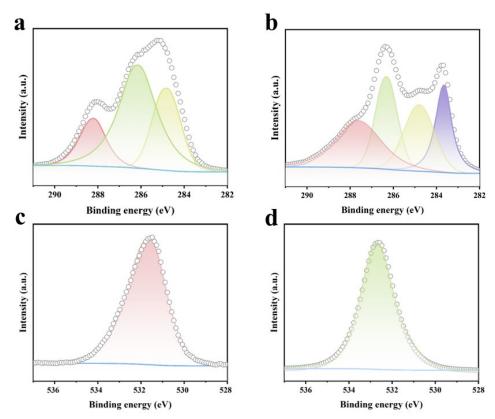


Fig. S8 High-resolution C 1s spectrum of (a) gelatin-CNT aerogel and (b) CNF-CNT aerogel. High-resolution of O 1s spectrum of (c) gelatin-CNT aerogel (d) CNF-CNT aerogel.

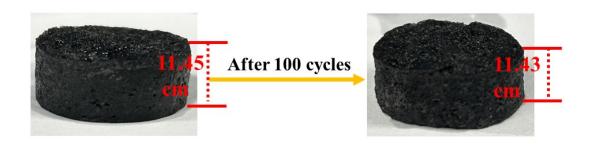


Fig. S9 Height change of gelatin-CNF-CNT aerogel after 100 compression cycles

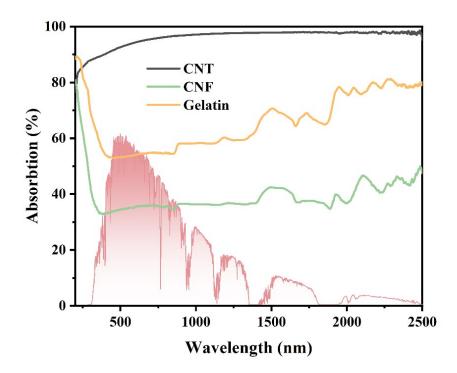
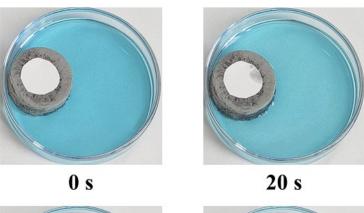


Fig. S10 Absorption spectra of pure CNT, CNF aerogel and gelatin aerogel in the range of full-spectrum.



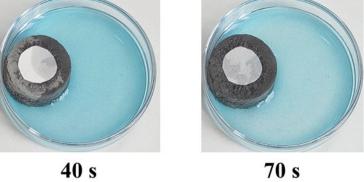


Fig. S11 The ability of gelatin-CNF-CNT aerogel to transport water.

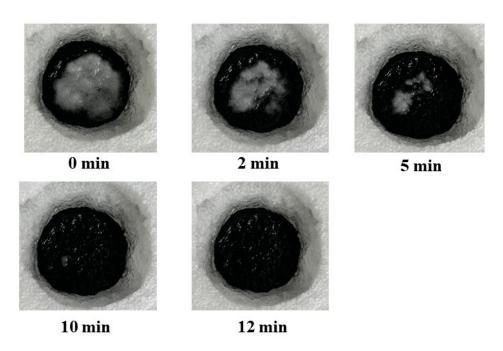


Fig. S12 The 1.5g salt crystals on the surface of the aerogel in 10 wt% saline were completely dissolved within 12 minutes.



Fig. S13 The 1.5g salt crystals on the surface of the aerogel in 3.5 wt% saline were completely dissolved within 50 minutes in dark environment.

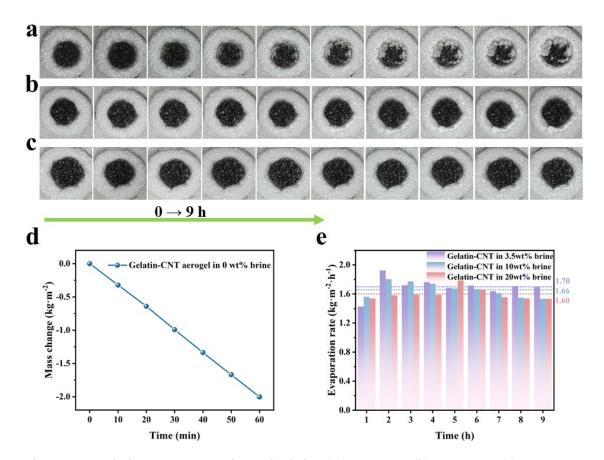


Fig. S14 Gelatin-CNT aerogel worked in (a) 20wt%, (b) 10wt%, (c) 3.5wt% concentration brine for 9 hours, and the salt output of the photothermal surface in each hour. (d) Evaporation rate of gelatin-CNT aerogel in pure water. (e) The evaporation rates of gelatin-CNT aerogel per hour within 9 hours of working in 3.5wt%, 10wt%, 20wt% saline.



Fig. S15 Apparatus for the collection and purification of dye-containing wastewater.

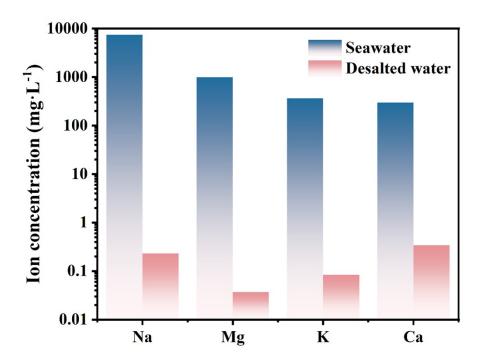


Fig. S16 Concentrations of ions before and after the solar-driven purification.

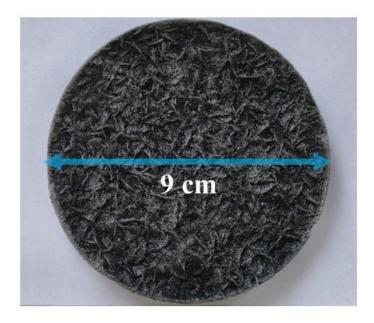


Fig. S17 Appearance of large-diameter gelatin-CNF-CNT aerogel

## **Calculation formula**

1.Evaporation rate

$$v = \frac{m}{St} \# (Eq. S1)$$

where m is the mass of the evaporated water, S is the surface area of the sample, and t is the evaporation time.

## 2. Energy conversion efficiency

$$\eta = v H_{eau} / Q \# (Eq. S2)$$

where v is the net evaporation rate,  $H_{equ}$  is the equivalent water evaporation enthalpy of gelatin-CNF-CNT aerogel at the operating temperature under one-sun illumination, and Q is the solar power density simulated.

## 3.Energy balance analysis

The input light intensity during the whole experiment was 1 kW·m<sup>-2</sup>, and the main uses of the energy were divided into (1) energy for evaporating brine to produce clean

water, which accounted for most of the input light intensity; (2) reflected energy; (3) Heat loss, which includes heat conduction, heat convection and heat radiation loss. After testing, the light absorption rate of aerogel is 95.1%, so the energy loss reflected is 4.9%. The calculation of the three types of heat loss is as follows:

(1) Conduction loss

$$q = k \bigtriangleup \frac{T}{L} \#(Eq. S3)$$

Where k is the thermal conductivity of the water (0.59 W·m<sup>-1</sup>·K<sup>-1</sup>), and  $\Delta T/L$  is the temperature gradient of the water at the bottom of the evaporator. The measured temperature gradient in 3.5 wt% brine is 53.96 K·m<sup>-1</sup>. Thus, q is 31.8 W·m<sup>-2</sup>.

(2) Convection loss

$$p = hS(T_1 - T_e) \#(Eq.S4)$$

Where *h* denotes the convection heat transfer coefficient (5 W·m<sup>-2</sup>·K<sup>-1</sup>). *S* represents the surface area of sample.  $T_l$  is the average surface temperature of the aerogel after the end of illumination (316.15 K), and  $T_e$  is the ambient temperature (308.15 K). Thus, *p* is 40.0 W·m<sup>-2</sup>.

(3) Radiation loss

$$\varphi = S \left( T_1^4 - T_e^4 \right) \# (Eq.S5)_{\mathcal{E}} \sigma$$

 $\varepsilon$  denotes emissive rate (assumed to be 0.8), *S* is evaporation surface area,  $\sigma$  represents the Stefan-Boltzmann constant (5.67×10<sup>-8</sup> W·m<sup>-2</sup>·K<sup>-4</sup>) *T*<sub>1</sub> is the average surface temperature of the aerogel after the end of illumination (316.15 K), and *T*<sub>e</sub> is the ambient temperature (308.15 K). Thus,  $\varphi$  is 44.1 W·m<sup>-2</sup>.