Supporting Information

Interface Chemical Coupling Enables Janus Elastomer-Hydrogel Composite for Roof-Free Evaporator with Efficient Hydrocooling Condensation

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KEYWORDS: Chemical coupling, janus composites, photothermal hydrogel, roof-free device, water-assisted enhanced condensation

Experimental materials and methods.

Materials

Acrylic acid (AA), acrylamide (AAm), ammonium persulphate (APS), N,N′-methylenebis acrylamide (BIS), 3(Trimethoxysilyl) propyl methacrylate (TMSPMA), photothermal particle graphite were obtained from Aladdin Shanghai Reagent Co. Ltd. Commercial PDMS were obtained from 0.5 mm Rhea Elastomer Membrane Co., LTD.

Chemical Modification of the PDMS film by TMSPMA：

PDMS films were treated by O_2 plasma irradiation (FEMTO plasma cleaner) for 1 min at 100 W and then immersed into the solution ($V_{H2O}/V_{HCl}/V_{H2O2}$, 5:1:1) to produce hydroxyl (-OH) groups. Subsequently, the activated PDMS films were immersed into an ethanol/water solution containing 3% TMSPMA to allow the generation of vinyl functional groups by silanization reaction. Finally, hydroxylated PDMS sheets were rinsed with deionized water.

The fabrication of PDMS/photomental hydrogels composite layers:

PDMS/photomental hydrogels composite layers were prepared by pouring hydrogels precursor (10 mg graphite 10 mg APS+10 mg BIS+0.5 g AAm+0.5 g AA dissolved in 4 g deionized water) was pouring onto TMSPMA/PDMS. Then we obtained the PDMS/functional hydrogels composite layers after polymerization in oven at 70°C for 6h.

The fabrication of PDMS/photomental hydrogels/functional hydrogels composite layers:

The PDMS/functional hydrogels were first immersed in 0.5 mol/L APS solution for 0.5 hour. Next, the precursor of functional hydrogels（10 mg APS+30 mg BIS+1 g DMAPS） dissolved in 4 g deionized water) was poured on the photomental hydrogel side of the bilayer structure. Finally, the whole structure was placed in an oven at 70 °C for 6 hours to obtain the PDMS/photomental hydrogels/ functional hydrogels three-layer structure.

Contrast experiment of photothermal energy management capabilities of SRME and SRFE:

SRME and SRFE were simultaneously exposed to 1 sun($1000w \cdot m^{-2}$) solar irradiation for 60 min. During this process, the 2D infrared thermal imager was used to capture the maximum temperature of its surface and the temperature probe was used to record the real-time temperature of the photothermal hydrogel layer.

COMSOL simulation of SRME and SRFE:

We built a COMSOL model to analyze the temperature distribution of the SRFE and SRME system by COMSOL Multiphysics 5.6. The heat transfer model in the SRFE and SRME system could be calculated by the equation given as following[44]:

$$\rho C_P \frac{\partial T}{\partial t} + \rho C C_P u \cdot \nabla T + \nabla \cdot q = Q \quad (1)$$

$$q = -k\nabla T \quad (2)$$

where T is the temperature, u is the fluid flow speed; ρ is the density, C_p is the special heat capacity and k is the thermal conductivity. In the mode simulation, the final condition of transient analysis was set to t = 30 min. As a result, the temperature distribution in confined heating evaporator slightly varies with thickness.

Calculation of the enthalpy change of evaporation:

$$U_{in} = E_{equ} m_g = E_0 m_0 \quad (3)$$

where m_g is the mass change of SRFE system in the dark environments, E_0 (2350 J·g^{-1}) and m_0 correspond to the evaporation enthalpy and mass of bulk water. As shown in **Figure S4b**, the enthalpy change of evaporation of SRFE system loaded with photothermal hydrogels of different proportions can be calculated as 2203.9 J·g^{-1}, 1837.9 J·g^{-1}, 1607.6 J·g^{-1}, 1702.8 J·g^{-1}, 2234.8 J·g^{-1} by the above formula.

Evaporation efficiency calculation：

The resulted solar-driven purification devices were set under the irradiation of Solar Simulator (HM-Xe500W) equipped with an AM 1.5 G filter for water purification.

$$\eta = \frac{mE_{equ}}{qC_{opt}} \quad (4)$$

where m is the mass flux of SRFE system, C_{opt} is the optical concentration on the evaporation surface, and q is the nominal solar intensity (1 kW m^{-2}). E_{equ} refers to the equivalent evaporation enthalpy of the water in the SRFE system, which can be calculated by evaporation water under the dark condition assuming the same energy input (U_{in}). All purification measurements were conducted under an ambient temperature of 26 ~ 29 ºC with humidity of 65~75 %.

Condensation efficiency calculation：

The condensing efficiency is calculated as shown in the following equation:

$$\eta = \frac{m_1}{m_2} \quad (5)$$

where m_1 and m_2 are the mass of water evaporation and mass of water collection, respectively.

Condensate water collection experiments in laboratory environment SRME and SRFE：

A container containing about 1 L of simulated seawater obtained from Zhejiang blue Starfish Salt Product Co., LTD. was placed on a temperature-controlled aluminium sheet. Temperature-controlled probe was placed in the simulated seawater to maintain a stable ambient seawater temperature. The temperature ranges of the simulated seawater were set to 9.5 °C -10.5 °C; 14.5 °C-15.5 °C; 19.5 °C-20.5 °C; 24.5 °C-25.5 °C; 29.5 °C-30.5 °C. The evaporation mass and collection mass of water from the SRFE unit at these five ambient seawater temperatures (10°C, 15°C, 20°C, 25°C, 30°C) was recorded under the irradiation of a solar simulator (HM-Xe500W) equipped with an AM 1.5G filter.

For the SRME, the photothermal hydrogel layer was floated directly on the surface of the simulated seawater. And the electronic scale was used to record the change in mass of water in the SRME during evaporation in the same solar irradiation environment.

Condensate water collection experiments in outdoor environment：

SRFE can be successfully floated on the water surface after being loaded onto PS foam. An array consisting of nine SRFE units with a photothermal area of 3.5*3.5 cm was prepared. As a comparison SRME used a conventional transparent roof structure as a condensation water collection device. The whole evaporation and water collection devices were exposed under the true sunlight irradiation to collect water for 6 hours.

Characterization：

A field emission scanning electron microscope (FE-SEM) with a FE scanning electron microanalyzer (HitachiS4800, 8kV) was used to observe the microstructures of functional hydrogels and PDMS. The Raman spectra for detecting the model molecule of PDMS and modified PDMS were collected using the 785 nm laser and accumulated 15 times for 1 s (1.2 mW). (Raman Systems, Inc., R-3,000 series, Britain). The infrared spectra of PDMS and functional hydrogels were obtained by an attenuated total reflection Fourier transformed Infrared Spectrometer (ATR-FTIR). Ultraviolet-visible-near infrared (UV–vis–NIR)

spectrophotometer (PerkinElmer, Lambda 950, USA) was used to test the transmittance and reflection of the JEHCs. The IR camera (FLIR E8, Germany) was employed to realize a real-time recording of the surface temperature and IR images of the JEHCs samples. The analytical scale (JJ224BC, China) was used to read the SRFE and SRME weight and water mass loss. The temperature and humidity were simultaneously recorded by a Temperature & Humidity recorder (Cos-03, China).

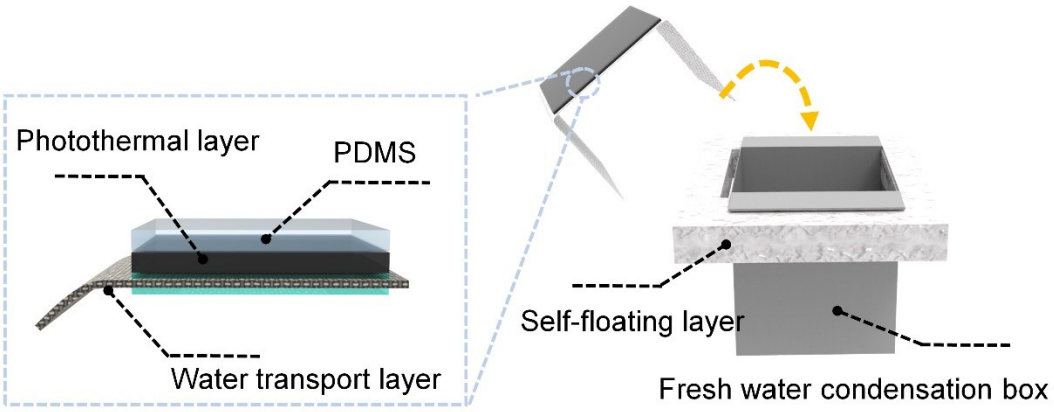

Fig. S1 Schematic diagram of the components of a single asymmetric inverted water condensation device

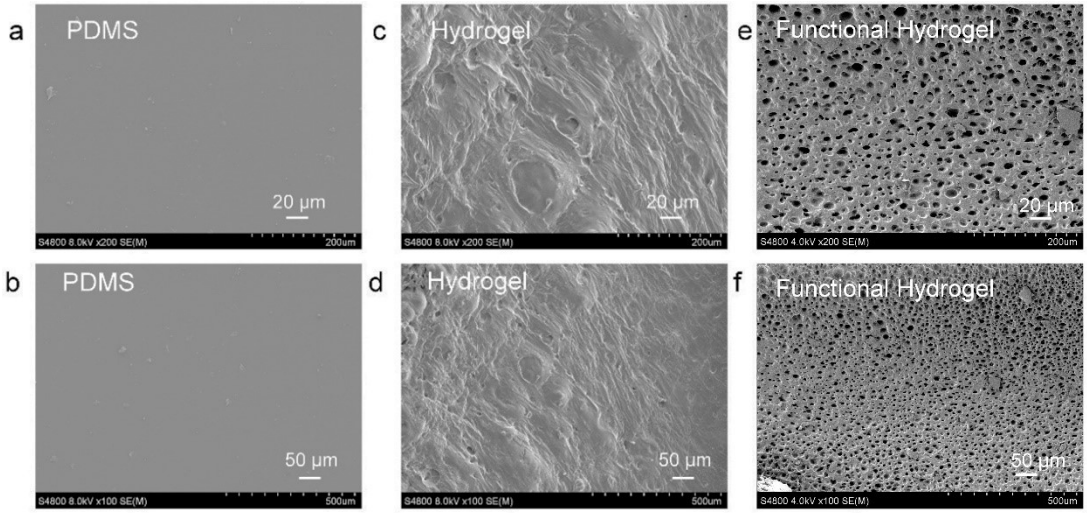

Fig. S2 (a)(b) SEM images of PDMS at different resolutions. (c)(d) SEM images of PDMS at different resolutions. (e) (f) SEM images of interface of PDMS and hydrogel. The thickness of the commercial PDMS is about 0.5 mm.

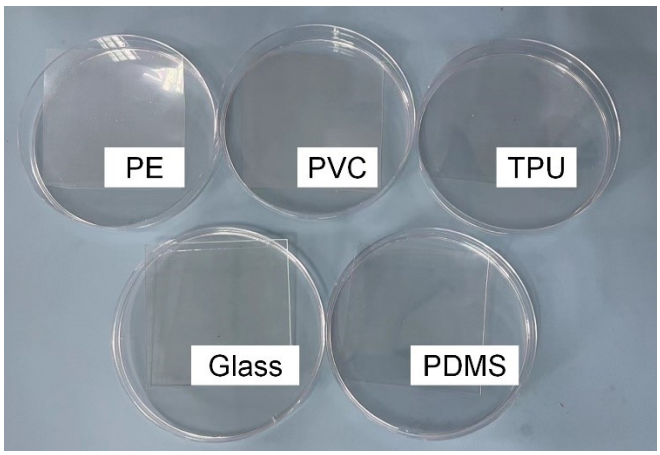

Fig. S3 Photographs of PE film, PVC film, TPU film, glass and PDMS film.

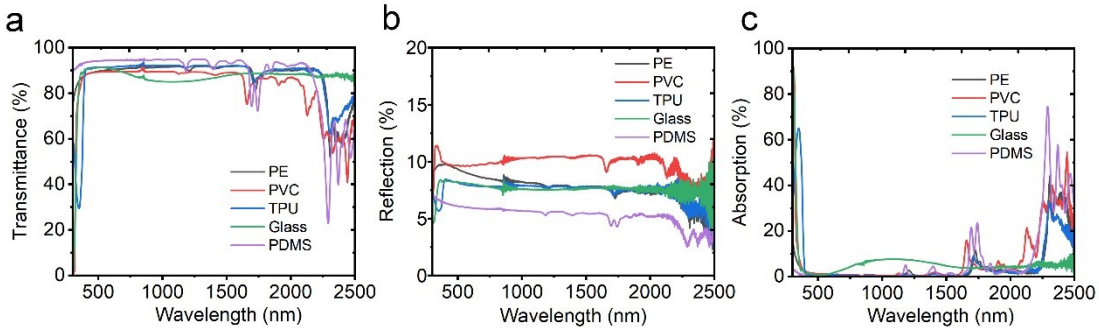

Fig. S4 (a) UV−vis−NIR transmittance spectra of PE, PVC, TPU, glass and PDMS. (b) UV−vis−NIR reflection spectra of PE, PVC, TPU, glass and PDMS.. (c) UV−vis−NIR absorption spectra of PE, PVC, TPU, glass and PDMS.

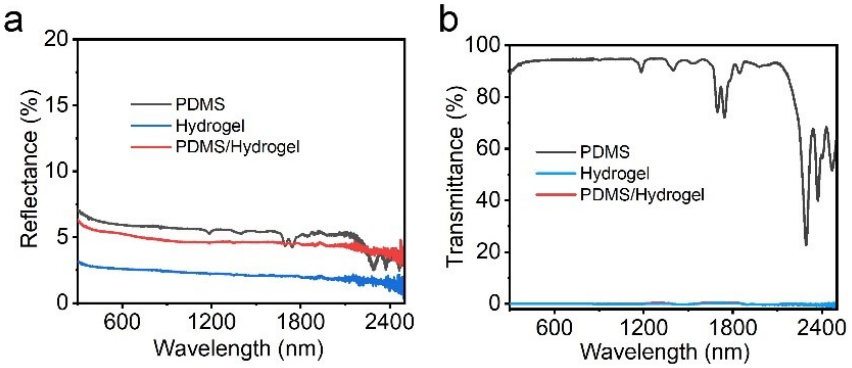

Fig. S5 (a) UV−vis−NIR reflectance spectra of PDMS, hydrogel and PDMS/functional hydrogels composite layers. (b) UV−vis−NIR transmittance spectra of PDMS, hydrogel and PDMS/functional hydrogels composite layers.

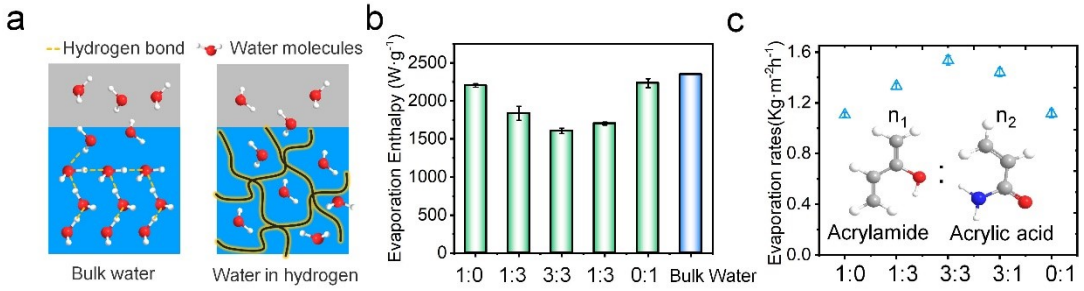

Fig. S6 (a) Schematic illustration of water molecules in pure water and the hydratable polymer network of evaporator. (b) Evaporation Enthalpy of Hydrogels in different ratios. (c) Evaporation rates of hydrogels with different ratios.

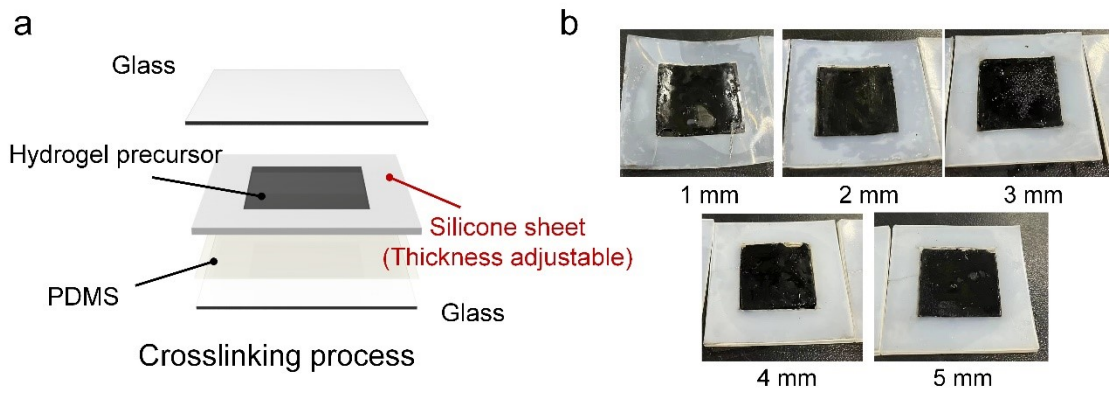

Fig. S7 (a) Schematic diagram of the method for preparation of photothermal hydrogel layers. (b) Photographs of different thicknesses of photothermal hydrogel layers.

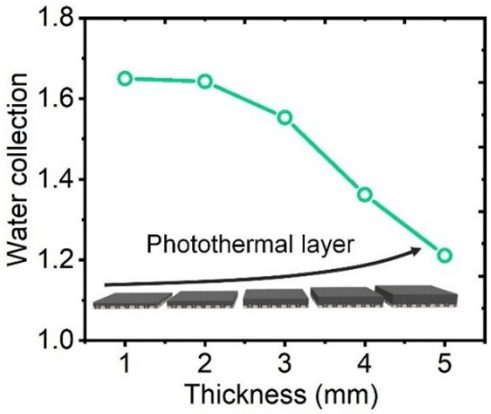

Fig S8 Effect of different thicknesses of photothermal hydrogel layers on water collection performance.

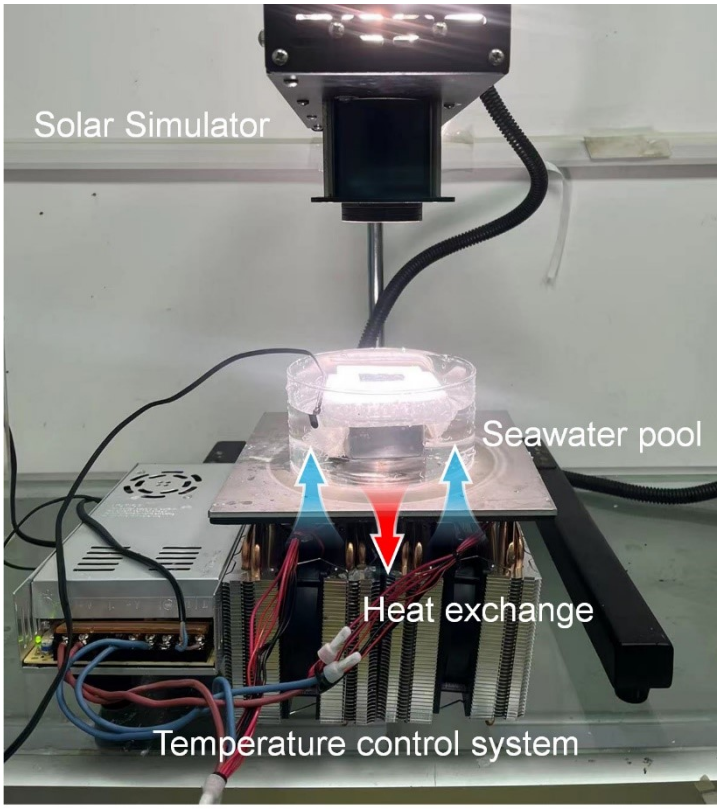

Fig. S9 Photograph of the solar-driven water collection capacity of the solar-driven roof-free evaporator (SRFE). Thermoelectric coolers and temperature control systems are used to precisely control the temperature of the simulated sea water.

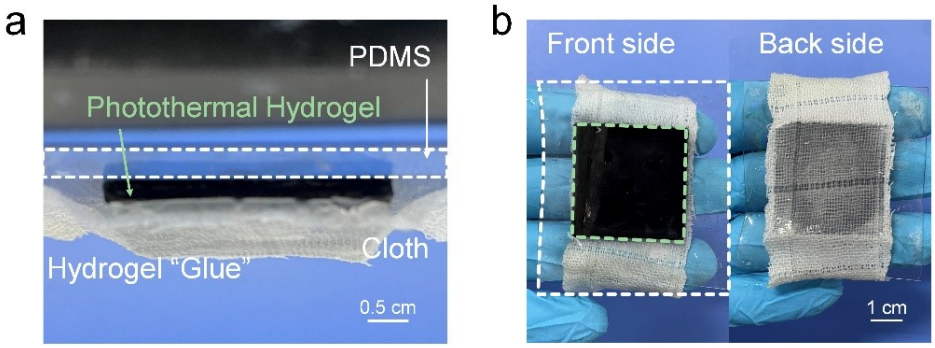

Fig. S10 Photographs of JEHCs cross-link the hydrophilic fabric.

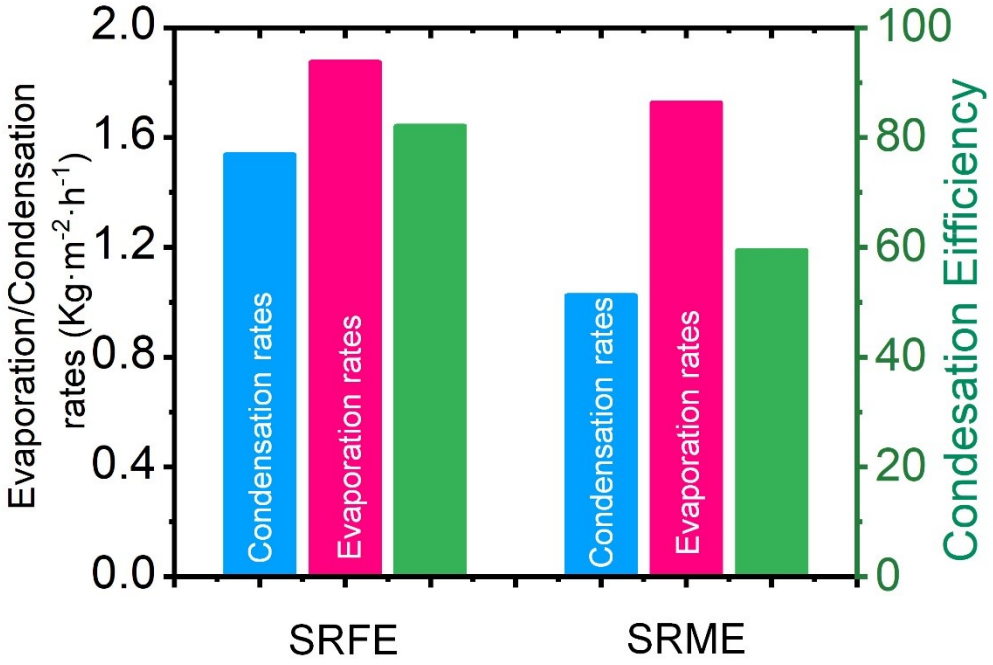

Figure S11 Comparison of evaporation rate and condensation efficiency between SRFE and SRME.

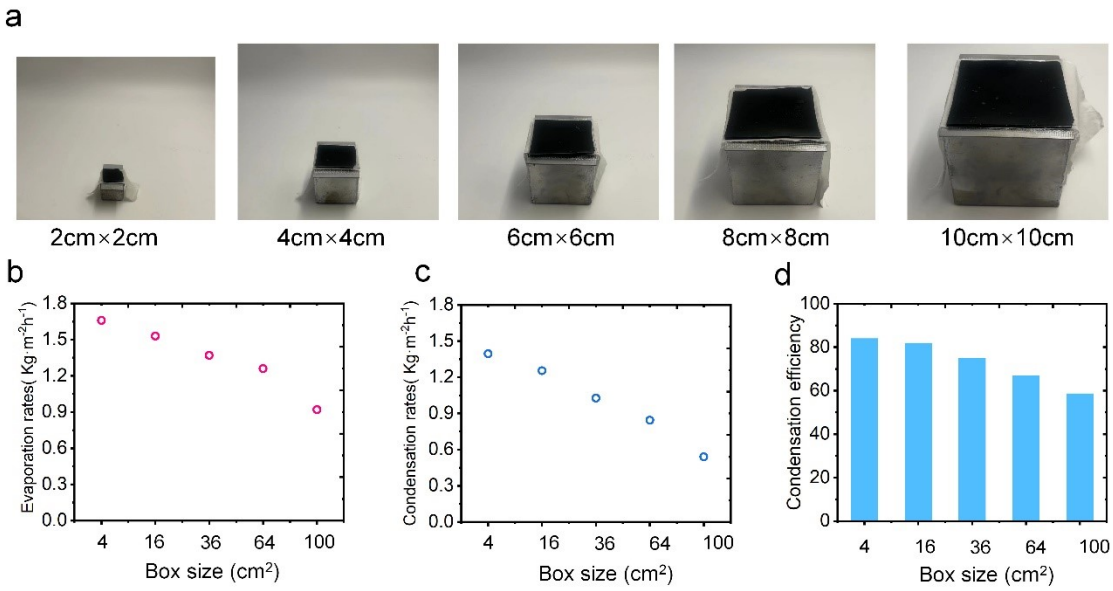

Fig. S12 (a) Photographs of SRFM device in different sizes (2cm×2cm, 4cm×4cm, 6cm×6cm, 8cm×8cm, 10cm×10cm). (b) Evaporation rate of SRFM using different metal box sizes. (c) Condensation rates of SRFM using different metal box sizes. (d) Calculated condensation efficiency of SRFM using different metal box sizes.

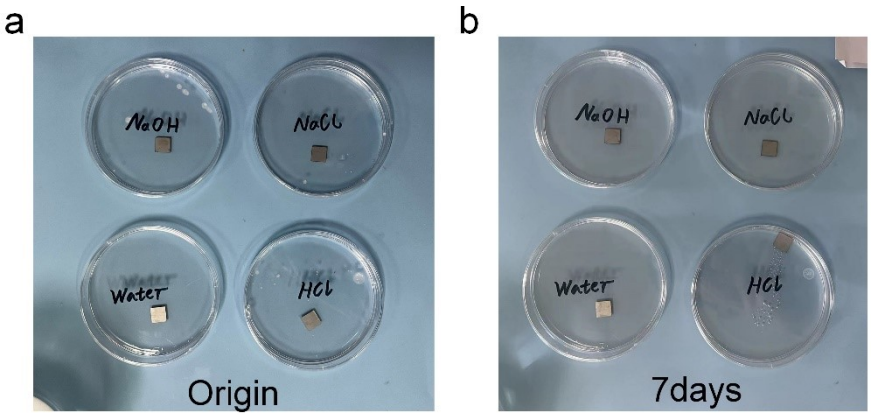

Fig. S13 Photograph of the 304 metal sheet (used in the metal box) in NaOH, HCl, NaCl solution and pure water for 7 days.