Highly Salt-Resistant and All-Weather Solar-Driven Interfacial Evaporators with Photothermal and Electrothermal Effects Based on Janus Graphene@Silicone Sponges

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Abstract

Solar-driven interfacial evaporators are very promising for obtaining clean water, but suffer from serious performance degradation by salt-fouling, low evaporation rate under weak illumination and low clean water collection rate. Here, we report highly salt-resistant and all-weather evaporators with photothermal and electrothermal effects based on the Janus graphene@silicone sponges with opposing wettability. The evaporators achieve a remarkable high evaporation rate of 6.53 kg m\(^{-2}\) h\(^{-1}\) for 3.5 wt% NaCl solution under 1 sun illumination with a 5 V solar cell as compensation owing to their high solar absorption, low thermal conductivity, unique Janus structure and synergetic photothermal and electrothermal effects. Even in gloomy and dark environments, the evaporators could still generate vapor (1.51 kg m\(^{-2}\) h\(^{-1}\)). Moreover, the evaporators feature long-term excellent salt-resistance, e.g., >10 d continuous evaporation in 10 wt% NaCl solution without performance degradation and salt precipitation, because of ultrafast water supply and salt diffusion in the macroporous superhydrophilic shell. Furthermore, the evaporators show high clean water collection rates of 21.92 kg m\(^{-2}\) d\(^{-1}\) (1 sun-9 h/0 sun-15 h + 5 V, indoor) and 9.65 kg m\(^{-2}\) d\(^{-1}\) (natural sun light + 5 V, outdoor). This study offers a new approach for efficiently obtaining clean water via solar desalination.

Keywords: interfacial evaporation; solar vapor generation; silicone sponge; graphene; desalination
**Introduction**

Clean water scarcity is an enormous challenge to human survival and development.\(^1\) Water purification by efficiently harvesting solar energy has always been an attractive area in academia and industry.\(^{1-4}\) Solar-driven interfacial (SI) evaporation has recently been emerging as a very promising approach to purify water by localizing heat at the water-vapor interface.\(^{4-5}\) High evaporation rate and solar-vapor efficiency have been achieved by developing advanced SI evaporators via the combination of new photothermal materials (e.g., semiconductors,\(^6\) carbonaceous materials\(^7\) and metal oxides\(^8\)), structural optimization and thermal management.\(^3, 9-10\) Now, the main bottleneck hindering SI evaporators from practical solar desalination is serious salt-fouling due to insufficient water supply, strong solar flux and high salt concentration, etc.\(^11\) The precipitated salts on/in solar evaporators will gradually block the pathways for light absorption, water transport and vapor escape, and ultimately deteriorate the evaporation rate and long-term stability. Two types of attempts including surface hydrophobization and multi-level evaporation architectures have been made to inhibit salt precipitation.\(^{12-14}\) Nevertheless, these strategies often result in high heat loss and low evaporation rate.

Furthermore, the existent SI evaporators can only generate vapor under strong illumination. The evaporation performance degrades markedly under weak sunlight such as cloudy, rainy and dark environments, which cannot satisfy practical demands for continuous water purification. This is because the conventional SI evaporators are designed mainly by innovations from the aspect of enhancing photothermal efficiency. As we all known, solar energy can be converted to not only heat, but also electricity. To enhance the evaporation under weak sunlight, only very few studies have been reported by applying the photo-electro-thermal effect but did not take into consideration the salt-resistance and clean water collection performance.\(^{15-17}\)

Therefore, SI generators with excellent salt-resistance and all-weather high evaporation performance are greatly desired for practical solar desalination yet very challenging to develop. Generally, high evaporation performance and salt-resistance are constrained by each other. High evaporation rate results
in salt precipitation more easily, whereas excellent salt-resistance is often at the sacrifice of evaporation rate. Here, we report highly salt-resistant and all-weather SI evaporators with photothermal and electrothermal effects (Fig. 1a). The SI evaporators were prepared by coating the superelastic silicone sponges with graphene followed by activation with O₂-plasma, forming the Janus graphene@silicone (p-GS) sponges with opposing wettability, i.e. superhydrophilic shell and superhydrophobic core. The p-GS sponges achieve a remarkable evaporation rate of 6.53 kg m⁻² h⁻¹ for 3.5 wt% NaCl solution under 1 sun illumination with the aid of a 5 V solar cell, benefiting from high solar absorption (~99%), low thermal conductivity (0.043 W m⁻¹ K⁻¹), unique wettability, and synergetic photothermal and electrothermal effects. Moreover, the p-GS sponges show long-term excellent salt-resistance because of ultrafast water supply and salt diffusion in the macroporous superhydrophilic shell (Fig. 1b). Even under very weak light conditions, the p-GS sponges can still generate vapor (1.51 kg m⁻² h⁻¹) by using electrical energy stored in the solar cell as a supplement (Fig. 1c).

**Fig. 1 Schematic illustration of the p-GS sponge.** a, the p-GS sponge with photothermal and electrothermal effects for efficient SI evaporation, b, the salt-resistance mechanism and (c) the electrothermal effect of the p-GS sponge.
Results and discussion

Preparation and properties of p-GS sponges

The p-GS sponges were prepared by sequential sol-gel, dip-coating, vacuum reduction and O₂-plasma activation processes (Fig. 2a and Supplementary S1a). First, the silicone sponges were fabricated by hydrolytic condensation of methyltrimethoxysilane (MTMS) and dimethyldimethoxysilane (DMDMS) in the presence of traces of HAc, Na₂CO₃ and dihexadecyldimethyl ammonium bromide (DHDAB).¹⁸

Compared with conventional porous materials, the silicone sponge has many unique properties, such as superhydrophobic, thermally insulating, superelastic and highly stable in various harsh conditions.¹⁹ Subsequently, the silicone sponges were coated with graphene oxide (GO, 0.25 mg mL⁻¹ unless otherwise specified) by dip-coating, which were then converted to the gray GS sponges by partial reduction of GO at 180 °C in vacuum.²⁰ Graphene has broadband solar absorption, tunable electrical conductivity and extraordinary photo/chemical stability.²¹-²² GS sponges in large size can be readily prepared (Supplementary Fig. 1b). Finally, the external surface of the GS sponges was activated using O₂-plasma, forming the Janus p-GS sponges with opposing wettability to achieve ultrafast water supply while maintaining the inherent low thermal conductivity.

The p-GS sponges showed 3D macroporous network consisting of highly branched silicone skeleton with a uniform graphene coating on the skeleton surface (Figs. 2b-c and Supplementary Fig. 2). The graphene nanosheets anchored on the silicone skeleton surface by hydrogen bonding between silanols of the silicone sponge and GO nanosheets in the dip-coating step.²³ The graphene coating densified the network according to the cross-sectional elemental maps (Supplementary Fig. 3) and increased surface roughness of the skeleton (Figs. 2b-c and Supplementary Fig. 2). The presence of graphene was also confirmed by the X-ray photoelectron spectroscopy (Supplementary Fig. 4). The C/Si atomic ratio on the surface of the p-GS sponge is 1.87, which is higher than that of the silicone sponge (1.44).¹⁸ This network is conducive to light absorption, rapid liquid transport by capillary action and vapor diffusion via natural convection.¹⁴,²⁴-²⁷
Fig. 2. Fabrication of the p-GS sponges. a, Schematic illustration of fabrication of the p-GS sponges. b, c SEM images of the p-GS sponge. d, Photograph of the GS and wet p-GS sponges with opposing wettability connected to a circuit. The sponges in d are 29 mm in diameter and 5 mm in height.

For practical applications, high mechanical strength, flexibility and stability of SI evaporators are desired but paid little attention. The p-GS sponges are strong, superelastic and flexible owing to inherent excellent mechanical properties of the silicone sponge and reinforcement by the graphene coating (Supplementary Fig. 5). The p-GS sponge could sustain at least 200 g weight without any deformation (Supplementary Fig. 6). The p-GS sponge could almost completely recover its original shape even after 80% compressive strain or 500 cyclic compressive cycles with 50% strain (Supplementary Fig. 7). After 500 cycles, no fracture or collapse of the p-GS sponge occurred and the graphene nanosheets still tightly bonded with the silicone skeleton (Supplementary Fig. 8).

The p-GS sponge is electrically conductive (~0.018 S m$^{-1}$) because of the continuous graphene coating on the silicone skeleton. A LED lamp could be lightened after being linked to a circuit using the GS and wet p-GS sponges (Fig. 2d). The brightness of the lamp relies on the compressive strain of the p-GS sponge (Supplementary Fig. 9 and Supplementary Video 1). The normalized electrical resistance ($R/R_0$) decreased rapidly to 6.49% with increasing the strain to 60% (Supplementary Fig. 10), as more contact points and areas were formed among the graphene coating. Upon unloading, the sponge sprang back quickly, and thus the electrical resistance recovered completely with very small loop. The sponge
remained conductive even after large stretching, bending or twisting (Supplementary Fig. 11 and Supplementary Video 1). The lamp could still be lightened when two pieces of broken p-GS sponges connected with each other (Supplementary Video 2). Moreover, the sponge showed high stability in extreme conditions, e.g., 30 min in liquid N₂ (-196 °C) or 2 h in 300 °C oven, for the changes of R/R₀ in the compression test were very small compared with the pristine p-GS sponge (Supplementary Fig. 12a). The sponge was still flexible and conductive in liquid N₂ (Supplementary Video 3), and could recover its original shape after squeezing out the liquid N₂. Even after 20 cycles of the tests in extreme conditions, R/R₀ remained steady in the compression test (Supplementary Fig. 12b). These results demonstrate reliable electrical conductivity of the p-GS sponge. This is owing to high robustness of the silicone sponge and stable bonding of the graphene coating.

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**SI evaporation performance**

Hydrophilic materials are often used as substrates of conventional SI evaporators for convenient water transport but at the cost of serious heat loss due to high thermal conductivity of water (0.6 W m$^{-1}$ K$^{-1}$).\textsuperscript{10} To guarantee sufficient water supply and to reduce heat loss simultaneously, the Janus p-GS sponge with opposing wettability was designed using the superhydrophobic GS sponge. The superhydrophilic shell ~1 mm in thickness serves as the pathway for rapid water transport driven by capillary force (Fig. 3a and Supplementary Fig. 13). The superhydrophobic core retains the intrinsic low thermal conductivity (0.043 W m$^{-1}$ K$^{-1}$, Fig. 3b) due to the macroporous network with air-filled interspaces,\textsuperscript{31} which is an order of magnitude lower than most of evaporators.\textsuperscript{32-34} The thermal conductivity of the wet p-GS sponge is still very low (0.16 W m$^{-1}$ K$^{-1}$). The wet p-GS sponge shows broadband light absorption of 95–99% across a wide wavelength of 200-2500 nm (Fig. 3c), covering the most vigorous part of the solar spectrum. This is due to the combination of the macroporous network, the graphene coating and the wet shell, which increased the optical path and decreased the reflection via multiple scattering of the incident light.\textsuperscript{28,35}

The top surface temperature of the dry p-GS sponge increased to 107 °C in 20 min under 1 sun (Supplementary Fig. 14), demonstrating excellent photothermal effect. During SI evaporation, the top surface of the p-GS sponge increased to ~39.5 °C in 5 min and then maintained at a steady state due to adequate water supply and evaporation. The water evaporation rate of the p-GS sponge is 1.61 kg m$^{-2}$ h$^{-1}$ under 1 sun (Fig. 3d), much higher than that of pure water (0.36 kg m$^{-2}$ h$^{-1}$) and silicone sponge (0.71 kg m$^{-2}$ h$^{-1}$). The evaporation rate and top surface temperature increased with GO concentration (Fig. 3d and Supplementary Fig. 15), but the effects were not obvious at concentrations above 0.25 mg mL$^{-1}$. The evaporation rate increased to 4.5 kg m$^{-2}$ h$^{-1}$ under 3 sun due to excellent light absorption and photothermal effect of the p-GS sponge (Fig. 3e). However, it is hard to have so strong illumination in natural environment and thus alternative approaches must be developed.
Fig. 3 Properties of the p-GS sponges. a, Cross-sectional photograph of the wet p-GS sponge and CAwater of its shell and core. b, Thermal conductivities of the p-GS sponge. c, UV-Vis-NIR absorption spectra of the p-GS sponge. d, Water mass changes with GO concentration. e, water mass changes with illumination intensity. The inset in e is photograph of steam generated under 3 sun. The sponge in a is 29 mm in diameter and 5 mm in height.

Salt-resistance

The salt-resistance of the p-GS sponge under long-time illumination and at high salt concentration was studied. In 3.5 wt% NaCl solution (average salinity of the world ocean), the evaporation rate of the p-GS sponge is ~1.58 kg m\(^{-2}\) h\(^{-1}\) under 1 sun. During the 20 d SI evaporation in 3.5 wt% NaCl solution with daily 6 h continuous 1 sun illumination, the evaporation rate slightly decreased to 1.51 kg m\(^{-2}\) h\(^{-1}\) in 6 h owing to falling of water level (Supplementary Fig. 16), but could completely recover upon water replenishment. The sponge maintained a stable average evaporation rate of ~1.55 kg m\(^{-2}\) h\(^{-1}\) throughout the 20 d consecutive test (Fig. 4a). Stable SI evaporation exceeding 12 h is challenging, for the salt precipitation rate usually exceeds its dissolution and diffusion rates.\(^{41}\) When the daily continuous illumination time was increased to 12 h, the water mass changed linearly with time and the evaporation
rate gradually reduced to 1.46 kg m$^{-2}$ h$^{-1}$ (Supplementary Fig. 17 and Fig. 4b). Throughout the 10 d consecutive test, the evaporation rate was very stable and no salt precipitated on the top surface of the sponge (Supplementary Fig. 18). After being dried without rinsing at atmospheric pressure, there was only a little salt sediment on the top surface. Also, the macroporous network of the sponge was maintained very well (Supplementary Fig. 19).

The evaporation rate decreased gradually to 1.17 kg m$^{-2}$ h$^{-1}$ under 1 sun with increasing the NaCl concentration to 20 wt% (nearly saturated, Fig. 4c), for the surface tension increased with NaCl concentration.\(^4\) No salt deposited on the top surface of the sponge during 10 d SI evaporation in 10 wt% NaCl solution with daily 12 h continuous 1 sun illumination (Supplementary Fig. 20 and 21). However, salt sediment appeared after 1 h under 1 sun for the 20 wt% NaCl solution and grew continuously as time evolved (Supplementary Fig. 22). Notably, when the light was turned off, the salt sediment gradually redissolved and re-diffused back to bulk liquid in 12 h.

![Fig. 4 Salt-resistance of the p-GS sponges. a, Evaporation rate of the p-GS sponge during the 20 d SI evaporation in 3.5 wt% NaCl solution with daily 6 h continuous 1 sun illumination. b, Evaporation rate of the p-GS sponge during the 10 d SI evaporation in 3.5 wt% NaCl solution in simulated natural environment with daily 12 h continuous 1 sun illumination (daytime) and 12 h of light off (nighttime). c,](image-url)}
Water mass changes with the NaCl concentration under 1 sun. d, Evaporation rate of the p-GS sponge in 3.5 wt% NaCl solution compared to recently reported SI evaporators.2a, 9c, 16c, 19, 20a, 22 e, Dynamic wetting behavior of shell and core of the p-GS sponge by 10 μL water droplets.

The above results demonstrate high evaporation rate and excellent salt-resistance of the p-GS sponge compared to previous studies (Fig. 4d and Supplementary Table 1). The superior salt-resistance of the p-GS sponge is attributed to ultrafast water transport in its superhydrophilic shell, originating from the macroporous network and abundant hydrophilic groups.43-44 Although the core is perfectly superhydrophobic, a 10 μL water droplet can instantaneously wet and permeate into the shell within 7 ms (Fig. 4e and Supplementary Video 4), which is the shortest record among all the reported SI evaporators (≥10 ms, Supplementary Table 2). The 10 wt% CuCl₂ solution can climb to the top of the O₂-plasma activated silicone sponge 60 mm in height in 90 s via the shell. This is greatly quicker than the conventional materials for water transport (Supplementary Fig. 23 and Supplementary Video 5). Due to ultrafast water transport and replenishment in the superhydrophilic shell, spontaneous salt diffusion is sufficient and salt concentration gradient diminishes during SI evaporation (Fig. 1b and Supplementary Fig. 24, and Supplementary Video 6). Consequently, salt precipitation is efficiently hindered and the temporarily deposited small amount of salt can redissolve and re-diffuse back to bulk liquid in time, even under long-lasting strong illumination and at high salt concentration. In addition, a thin water film is maintained on the top surface of the p-GS sponge during SI evaporation, which also contributes to the superior salt-resistance.45
Synergetic photothermal and electrothermal effects

Fig.5 Synergetic photothermal and electrothermal effects of the p-GS sponges. 

a, Schematic illustration of the experimental setup for SI evaporation via the synergetic photothermal and electrothermal effects of the p-GS sponge. 
b, Mass changes of 3.5 wt% NaCl solution over time under different conditions and c, Mass changes of 3.5 wt% NaCl solution over time under 1 sun + 1-5 V. 
d, Evaporation rate of the p-GS sponge in 3.5 wt% NaCl solution under 1 sun + 1-5 V. 
e, Evaporation rate of the p-GS sponge in 3.5 wt% NaCl solution during 21 h SI evaporation by turning on and off the 5 V solar cell every 1 h under 1 sun. 
f, Photographs of the p-GS sponge during 10 d SI evaporation in 3.5 wt% NaCl solution with daily 12 h continuous evaporation under 1 sun + 5 V. 
g, Solar flux and outdoor temperature, and h, the corresponding mass changes of 3.5 wt% NaCl solution and top surface
temperature of the p-GS sponge during outdoor SI evaporation under different weather conditions (gloomy, cloudy and sunny) at Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences. Mass changes of 3.5 wt% NaCl solution and top surface temperature of the p-GS sponge during outdoor SI evaporation on Jan. 15, 2020. The sponge in f is 29 mm in diameter and 5 mm in height.

To enhance the SI evaporation performance by taking full advantage of solar energy, the p-GS sponge was combined with a solar cell (Fig. 5a and Supplementary Fig. 25). Thus, the p-GS sponge could not only directly convert solar energy into heat via the photothermal effect, but also convert electricity stored in the solar cell to additional heat via the electrothermal effect. Compared to 1 sun illumination, the evaporation rate of 3.5 wt% NaCl solution increased significantly to 2.34 kg m\(^{-2}\) h\(^{-1}\) under 1 sun + 1 V (Fig. 5b). Also, the top surface temperature of the sponge increased quickly to 40.3 °C in 2 min under 1 sun + 1 V, which is much faster than that under 1 sun (12 min, Supplementary Fig. 26). The evaporation rate even reached 6.53 kg m\(^{-2}\) h\(^{-1}\) under 1 sun + 5 V (Fig. 5c), which is higher than all the reported salt-resistant SI evaporators (Supplementary Table S1). In addition, in the repeated voltage up/down and solar cell on/off processes, alternate evaporation rate up/down was highly stable and controllable (Fig. 5d-e). Furthermore, the evaporation rate was still as high as 1.5 kg m\(^{-2}\) h\(^{-1}\) under weak indoor light conditions (~0.079 sun, 18 °C) with a 5 V solar cell as compensation. These results manifest that the synergetic photothermal and electrothermal effects can largely enhance the SI evaporation performance even under weak light conditions. During the above tests and the 10 d SI evaporation test in 3.5 wt% NaCl solution with daily 12 h continuous evaporation under 1 sun + 5 V, no salt deposited on the top surface of the sponge (Fig. 5f), further demonstrating excellent salt-resistance while maintaining high evaporation rate (~6.41 kg m\(^{-2}\) h\(^{-1}\)).
The outdoor SI evaporation performance of the p-GS sponge under different weather conditions (24 h) at Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences is shown in Figs. 5g-h. As the solar flux and outdoor temperature changed, the evaporation rate of 3.5 wt% NaCl solution and the top surface temperature of the p-GS sponge changed correspondingly. On a sunny day in early spring (Mar. 05, 2020), the evaporation rate reached the maximum of only 0.73 kg m\(^{-2}\) h\(^{-1}\) at 13:00 under 0.43 sun illumination. On a gloomy day in winter (Jan. 15, 2020) with very low solar flux (<0.1 sun) and outdoor temperature (-3~1 °C, Supplementary Fig. 27), the average evaporation rate was merely 0.02 kg m\(^{-2}\) h\(^{-1}\) and the top surface temperature of the p-GS sponge was only -3~0 °C in the daytime (9:00-18:00). Particularly, the SI evaporation almost stopped from 18:00 at night. However, the average evaporation rate sharply increased to ~1.3 kg m\(^{-2}\) h\(^{-1}\) under the same conditions with a 5 V solar cell as compensation (Fig. 5i). Also, the average evaporation rate was still as high as ~1.2 kg m\(^{-2}\) h\(^{-1}\) in the nighttime (18:00-09:00) with the aid of a 5 V solar cell. Thus, efficient outdoor evaporation of saline water was achieved in gloomy weather and even nighttime owing to electrothermal effect of the p-GS sponge.

**Clean water collection**

An all-in-one closed device was constructed using the p-GS sponge in order to collect clean water by SI evaporation of seawater (Fig. 6a). Under 1 sun + 5 V, vapor mist appeared quickly in 30 min and condensed droplets were formed on the inner wall of the cone-shaped glass cover (Supplementary Fig. 28). The cover was mist- and droplet-free within the first 2 h, and then condensed with more and more water droplets over time. The condensed water dropped to the bottom of the device and was collected in a beaker. The average evaporation rate of 3.5 wt% NaCl solution was 2.08 kg m\(^{-2}\) h\(^{-1}\) in the closed device, which is much lower than that under open atmosphere (6.53 kg m\(^{-2}\) h\(^{-1}\)). This is owing to (i) reduction of light intensity by the glass cover, vapor mist and condensed water droplets on the cover, and (ii)
inhibition of water evaporation by the high humidity in the closed device. Within 9 h consecutive operation, 17.77 kg m\(^{-2}\) condensed water was collected, corresponding to a water collection rate of 1.97 kg m\(^{-2}\) h\(^{-1}\) and a vapor collection ratio of \(~95\%\), which are higher than the reported results (Supplementary Table 3).

**Fig. 6 Collecting clean water.** a, Schematic illustration and b, photograph of the all-in-one closed device for collecting clean water by SI evaporation of seawater via photothermal and electrothermal effects of the p-GS sponge. c, Summary of energy flow and SI evaporation performance (outdoor solar flux <0.5 sun) of the p-GS sponge. d, Concentrations of four primary ions in simulated seawater and collected water via solar desalination using the closed device in b under 1 sun + 5 V.
The outdoor clean water collection performance of the closed device with the aid of a 5 V solar cell was also studied (Fig. 6b). In the daytime (09:00-18:00, Apr. 24, 2020, solar flux = 0.04-0.49 sun, outdoor temperature = 10-17 °C, Supplementary Fig. 29), ~6.43 kg m⁻² clean water was collected in 9 h. In the nighttime (18:00-09:00, Apr. 24-25, 2020, solar flux = 0-0.39 sun, outdoor temperature = 8-12 °C), ~3.22 kg m⁻² clean water was still collected in 15 h. Thus, the device achieved high clean water collection of 9.65 kg m⁻² in 24 h owing to the synergetic photothermal and electrothermal effects. When fully charged by absorption of solar energy in 40 h, the solar cell can last for around 20 h with an output voltage of 5 V. So, with two of the small solar cells (180 cm²), clean water can be continuously collected at such a high rate. The overall energy flow of the integrated SI evaporation system is summarized in Fig. 6c. It is capable of efficiently converting solar energy to thermal energy via photothermal and electrothermal effects of the p-GS sponge as well as photoelectric effect of the solar cell. Consequently, the p-GS sponge shows a high evaporation rate of 6.53 kg m⁻² h⁻¹ (1 sun + 5 V, open atmosphere), excellent salt-resistance (>10 d, 10 wt% NaCl solution) and a high clean water collection rate of 9.65 kg m⁻² d⁻¹ (natural sunlight + 5 V, outdoor closed device).

After solar desalination of the simulated seawater using the closed device, the concentrations of several typical ions in the collected water were measured using the inductively coupled plasma optical emission spectrometer. The Na⁺, Mg²⁺, K⁺ and Ca²⁺ concentrations in simulated seawater were sharply reduced from 11521, 1200, 395 and 400 mg L⁻¹ to 3.85, 1.25, 0.6 and 0.7 mg L⁻¹, respectively (Fig. 6d). The concentrations of the ions in the collected water are greatly lower than the drinking water standards defined by the World Health Organization (WHO) as indicated by the dash lines.
Conclusions

In summary, highly salt-resistant and all-weather SI evaporators with photothermal and electrothermal effects were developed by coating the superelastic silicone sponges with graphene followed by activation with O\textsubscript{2}-plasma. The evaporators show high solar absorption, low thermal conductivity, excellent mechanical properties, reliable electrical conductivity, unique Janus structure (superhydrophilic shell and superhydrophobic core) as well as synergetic photothermal and electrothermal effects owing to the combined merits of the silicone sponge and the graphene coating. Consequently, the evaporators feature remarkable high evaporation rates of 6.53 kg m\textsuperscript{-2} h\textsuperscript{-1} under 1 sun + 5 V and even 1.51 kg m\textsuperscript{-2} h\textsuperscript{-1} in dark environments for 3.5 wt% NaCl solution. Also, the evaporators show excellent salt-resistance during long-term continuous evaporation of high concentration saline water without performance degradation and salt precipitation. Due to ultrafast water supply in the macroporous superhydrophilic shell, spontaneous salt diffusion is sufficient and salt concentration gradient diminishes during SI evaporation. Moreover, clean water can be continuously collected at a high rate of 9.65 kg m\textsuperscript{-2} d\textsuperscript{-1} under weak sunlight (0-0.49 sun) using the evaporators with a small solar cell as the supplement. This study offers a new strategy to construct high-performance SI evaporators, which opens up new opportunities for efficient solar desalination.

Methods

Materials. Graphite was supplied by Asbury Graphite Mills, Inc., United States. GO was prepared by a modified Hummers method.\textsuperscript{46} MTMS and DMDMS were purchased from Gelest. DHDAB, HAc, Na\textsubscript{2}CO\textsubscript{3}, NaCl, MgSO\textsubscript{4}, CaCl\textsubscript{2}, HCl and NaOH were purchased from China National Medicines Co., Ltd. All chemicals were used as received without further purification.

Preparation of silicone sponges. The silicone sponges were synthesized by hydrolytic condensation of MTMS
and DMDMS in a weak acidic buffer solution. Typically, 1.93 mg of DHDAB in 1.0 mL of water and 2 mg of Na₂CO₃ in 2.0 mL of water were added into a glass vial containing 4.7 mL of HAc solution (55 mM). Then, 1.43 g of MTMS and 0.84 g of DMDMS were sequentially added into the solution. The mixture was vibrated at 200 rpm for 1 h at 30 °C in a thermostatic incubator shaker. Due to hydrolysis of the precursors and the presence of DHDAB, a homogeneous and transparent sol was formed. Subsequently, the sol was transferred into a sealed reactor, which was placed in an oven at 95 °C for 24 h without any shake to form the silicone hydrogel. The silicone hydrogel was washed with water and dried in an oven at 60 °C under ambient pressure to obtain the silicone sponge.

**Preparation of p-GS sponges.** A piece of the as-prepared silicone sponge (~29 mm in diameter and 5 mm in height) was immersed in 100 mL of a GO solution (0-1 mg mL⁻¹) in ethanol at room temperature for 2 h. The silicone sponge was repeatedly squeezed to refresh the GO solution inside the sponge to facilitate uniform absorption of GO. Subsequently, the silicone sponge with absorbed GO was taken out of the solution and dried at 180 °C for 6 h in a vacuum oven to form the GS sponge. Finally, the external surface of the GS sponge was activated using O₂-plasma for 5 min, forming the Janus p-GS sponge with opposing wettability.

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Author contributions
L. X. L. and J. P. Z. conceived and designed research, L. X. L. performed research and data analysis; L. X. L. and J. P. Z. wrote the manuscripts with contributions from all authors; J. P. Z. supervised the work. All authors discussed the results and approved the final version of the manuscript.

Competing interests
The authors declare no conflict of interest.