

Microbubble-based elastic graphene aerogel for efficient solar-driven interfacial evaporation

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ABSTRACT

Solar-driven interfacial evaporation has emerged as one of the most promising solutions to address the shortage of freshwater resources. Graphene aerogels (GAs) exhibit immense potential in solar-driven interfacial evaporation due to their excellent photothermal conversion efficiency. However, the complex fabrication and inherent fragility of conventional GAs limit their application in desalination. To address these challenges, this study incorporates natural latex (NL) into GA to prepare a novel elastic GA/NL. It uses a gas bubbles and ice crystals templating method followed by atmospheric-pressure drying. The prepared GA/NL achieves a compressive strength of 66.9 kPa at 90 % strain. Its three-dimensional interconnected porous network structure contributes to high porosity (~ 97 %) and an average light absorbance of 93.3 %. Experimental determination exhibits an outstanding evaporation rate of about $2.68 \text{ kg m}^{-2} \text{ h}^{-1}$ and a solar-vapor conversion efficiency of 94.54 % under 1 sun irradiation (1 kW m^{-2}). Critically, GA/NL exhibits excellent stability of structure, salt resistance, and high desalination performance after ten evaporation cycles. These results indicate excellent mechanical properties for long-term desalination application, highlighting its exceptional potential as an efficient, durable interfacial solar steam generator.

1. Introduction

The rapid growth of the global population intensifies the demand for sustainable freshwater resources. Freshwater scarcity has escalated into a critical global challenge. Desalination technology provides a feasible solution to alleviate this water scarcity by converting abundant seawater into freshwater [1–4]. Conventional desalination technologies, such as reverse osmosis (RO) [5], vapor compression distillation (VCD) [6], and multi-effect distillation (MED) [7], can alleviate the pressure of freshwater scarcity. However, these technologies still face inevitable challenges, including complex operation, high cost, extensive fossil energy consumption, and environmental pollution. Recently, solar-driven interfacial evaporation technology based on the concept of thermal localization has been widely reported. Solar-driven interfacial evaporation technology utilizes interfacial solar steam generators (ISSGs) that float at the air-water interface to absorb solar radiation energy and convert it into thermal energy, thereby evaporating water to produce freshwater from seawater [8–12]. This technology is considered one of the most promising desalination technologies because of its high efficiency, low energy consumption, and sustainability [13–16].

The interfacial evaporation materials, as the key component of ISSGs, should exhibit high photothermal conversion efficiency and broad spectral absorption within solar irradiation band [17,18]. Photothermal conversion materials serve as the cornerstone of solar-driven interfacial evaporation systems. Currently prevalent photothermal materials encompass metal nanomaterials [19–24], semiconductors [25, 26], carbon-based materials [27–29], and organic polymers [30,31]. Carbon-based materials such as carbon black [32,33], carbon nanotubes [34–37], carbon fibers [38–40], and graphene [41–43] are the most commonly utilized photothermal conversion media due to their high conversion efficiency and stable chemical structures [44]. Notably, the lower-density GAs fabricated based on the graphene enable to self-floating on water, demonstrating significant potential as ISSGs [45, 46]. At present, most GAs are synthesized from graphene oxide (GO) nanosheets via hydrothermal reduction or self-assembly through chemical reduction [47]. For instance, Liu et al. [48] prepared molybdenum sulfide-graphene oxide aerogels (MGA) using a hydrothermal method and freeze-drying approach. The MGA achieved an evaporation rate of $1.72 \text{ kg m}^{-2} \text{ h}^{-1}$ and a photothermal conversion efficiency of 86.5 % under 1 sun (1 kW m^{-2}). During the GA reduction process, the

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decrease in oxygen-containing functional groups facilitates the recovery of the sp^2 structure. This restoration strengthens π - π conjugation between partial reduced graphene oxide (rGO) sheets, resulting in aggregation and stacking of rGO [49–51]. Consequently, this structural instability causes collapse of the GA framework, significantly compromising its practical applications. Furthermore, these conventional fabrication methods heavily depend on complex freeze-drying processes, which bring severe restrictions to large-scale production.

As a porous material, the performance of aerogel is highly dependent on a stable pore structure. The mechanical properties of aerogels are found to be closely related to interactions between rGO sheets. Thus, various cross-linking agents have been incorporated into GAs to strengthen their interactions [52]. Guo et al. [53] prepared a graphene/carbon nanotube hybrid aerogel (rGCA) through chemical reduction. The enhanced mechanical properties and elasticity were attributed to the incorporation of crosslinked natural rubber (NR) chains into the aerogel structure. The rGCA exhibited superior strength, enabling it to load a 500 g weight without structural failure. In addition to adding cross-linking agents, constructing ordered porous structures is another effective method to improve the mechanical properties of GAs. For example, Ding et al. [54] fabricated a clay/graphene aerogel (CGA) by using commercial foams as sacrificial backbones. The CGA possessed super-elasticity under compressive strain of 95.0 % and compressive stress of 0.09–0.23 MPa. It exhibited an extremely high evaporation rate of $4.11 \text{ kg m}^{-2} \text{ h}^{-1}$ under 1 sun. Conventional sacrificial template methods typically involve template removal through dissolution or pyrolysis. This method requires complex procedures and carries risks of residual template contamination. In contrast, the bubble templating method is an emerging technology to construct aerogels by directly engineering pore networks using gas bubbles, thereby eliminating energy-intensive processes like supercritical drying or complex chemical crosslinking.

It can be physically eliminated without requiring chemical treatments, thereby mitigating potential structural damage. Kong et al. [55] synthesized an ultralight and porous corn stover/graphene aerogel via a bubble templating method. It exhibited excellent mechanical properties (compressive strength of up to 11.4 kPa at 70 % strain), a high evaporation rate of $2.71 \text{ kg m}^{-2} \text{ h}^{-1}$, and an evaporation efficiency of 95.0 %. The pore structure and size inside aerogels are key parameters for solar steam generation performance [56]. Pore size directly affects capillary forces. Lower nano-scale pores generate stronger capillary forces, driving water to rise rapidly along the vertical direction. Larger micrometer-scale pores facilitate the smooth escape of steam by reducing flow resistance and preventing pore blockage. Additionally, higher porosity provides a larger water/steam interface area, which forms the physical foundation for efficient evaporation [57,58].

Based on the above analysis, current GAs face critical challenges including poor mechanical properties and complex fabrication processes, when utilized as interfacial solar steam generators. To address issues, this work fabricates graphene/natural latex aerogel (GA/NL) with a three-dimensional interconnected network structure via ambient-pressure drying, using mechanically stirred bubbles as templates. Incorporating NL into the graphene matrix theoretically enhances the mechanical properties of composites, which is beneficial for long-term desalination. The porous GA/NL exhibits excellent light absorption properties. Adjusting the pore size appropriately can achieve efficient water transport through a strong capillary effect within the interconnected macropores. The novel GA/NL maintains stable evaporation performance and robust mechanical properties over multiple long-term cycles, significantly advancing the development of solar-driven interfacial evaporation.

2. Experimental

2.1. Materials

Graphene oxide (GO, monolayer content 99 %, monolayer sheet size 0.2–10 μm) was purchased from Shenzhen Suiheng Science and Technology Co. Ltd. and natural latex (Natural Latex, H0101, dry gel content 60.2 %, ammonia content 0.72 %, PH = 10–11) was purchased from Shenzhen Jitian Chemical Co., Ltd. Deionized water, sodium dodecyl sulphate (SDS, AR, 92.5–100.5 %) and L-ascorbic acid (L-AA, $\text{C}_6\text{H}_8\text{O}_6$, AR) were purchased from Chengdu Chron Chemical Reagent Co., Ltd.

2.2. Preparation of GA

The 0.25 mL of SDS (50 mg mL^{-1}) and 70 mg L-AA were added into 5 mL of GO dispersion (7 mg mL^{-1}). The mixture was stirred at 6000 rpm for 20 min using a high-speed shear emulsifier. Next, the mixture was poured into a 3 cm diameter glass bottle, heated at 70°C for 1 h, and then placed in the refrigerator to freeze for 6 h. After being thawed at room temperature, the mixture was continued to heat for 6 h at 85°C . The GA was dialyzed with ethanol remove residual impurities and dried in a 55°C air-blast oven to obtain the final aerogel.

2.3. Preparation of GA/NL

Fig. 1 describes the synthesis process of GA/NL. Firstly, 0.25 mL of SDS (50 mg mL^{-1}) was added to 5 mL of GO dispersion (7 mg mL^{-1}). The mixture was stirred at 6000 rpm for 10 min using a high-speed shear emulsifier, generating copious bubbles due to the emulsion of SDS. Then the natural latex/SDS solution (containing 0.175 mL natural latex solution (100 mg mL^{-1}) and 0.25 mL SDS solution (50 mg mL^{-1})) and 70 mg L-AA were dissolved into the above GO/SDS solution. Adding SDS to the GO dispersion solution before mixing it with natural latex solution can effectively avoid GO sedimentation caused by the electrostatic effect. The mixture was fully emulsified at 6000 rpm for 10 min. Next, the mixture was poured into 3 cm diameter glass bottle, heated at 70°C for 1 h, and then placed in the refrigerator to freeze for 6 h. The mixture thawed at room temperature was heated for 6 h at 85°C . Finally, the GA/NL aerogel was formed by dialysis with ethanol and drying in a 55°C air-blast oven. The samples obtained were sequentially named GA/NL-x according to the rotational speeds, where x represents the rotational speed. (for instance, GA/NL-6 denotes simply prepared at 6000 rpm).

2.4. Characterization and measurements

The micromorphology of the samples was observed by scanning electron microscopy (SEM, Thermo Fisher FEI Inspect F, USA). The pore size distribution of the aerogels was determined by a mercury intrusion porosimeter (AutoPore IV 9500, Micromeritics, USA).

Elemental analysis of the samples was performed by X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha, USA). The chemical structures were characterized by a Fourier transform infrared spectrometer (FT-IR, Nicolet iS50 Thermo Scientific Co., Ltd., USA) in the wavenumber range of 4000 to 400 cm^{-1} . Raman spectroscopy was recorded using a LabRAM HR spectrometer (HORIBA Scientific, France) at an excitation wavelength of 532 nm. The crystal structures and crystallization behaviors of the samples were investigated using an X-ray diffractometer (XRD, Malvern Panalytical, UK). UV-Vis-NIR spectrophotometer (Shimadzu UV-3600, Japan) was employed to determine the absorbance of samples in the range of 200–2500 nm. Thermal conductivity is measured by the thermal conductivity meter (XIATECH, Xi'an Xiayi Electronic Technology Co., Ltd., China).

Differential scanning calorimetry (DSC, TA Instruments Co., Ltd., USA) was used to calculate the evaporation enthalpy of water in the sample. The sample was placed in an aluminum pan with a pierced lid

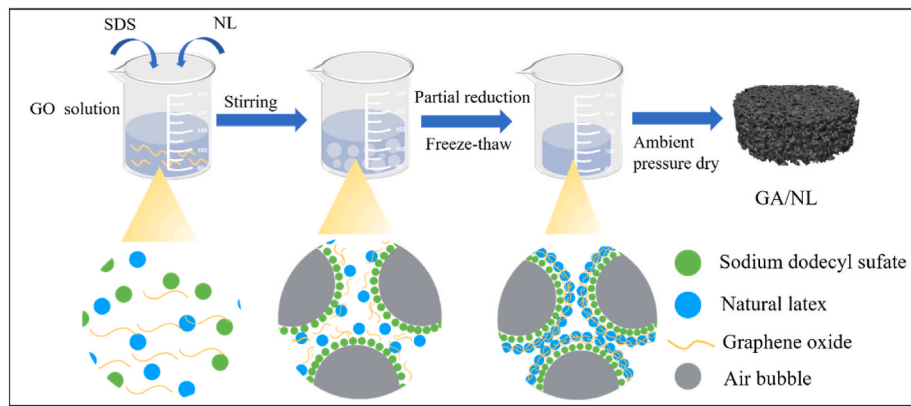


Fig. 1. Schematic diagram of GA/NL preparation.

and heated up from 20 °C to 140 °C with a linear heating rate of 5 °C min⁻¹. In addition, the thermal stability of the sample was determined on a TGA/DSC3+ (METTLER TOLEDO Co., Ltd., Switzerland) in the temperature range of 30–600 °C at heating rate of 10 °C min⁻¹ under a

nitrogen atmosphere. The hydrophilicity of the aerogel was determined using an automatic goniometer (SDC-100H, Dongguan SINDIN Precision Instrument Co., Ltd) with 25 μ L of water droplets. The compressive performance of aerogels was measured using a universal testing machine

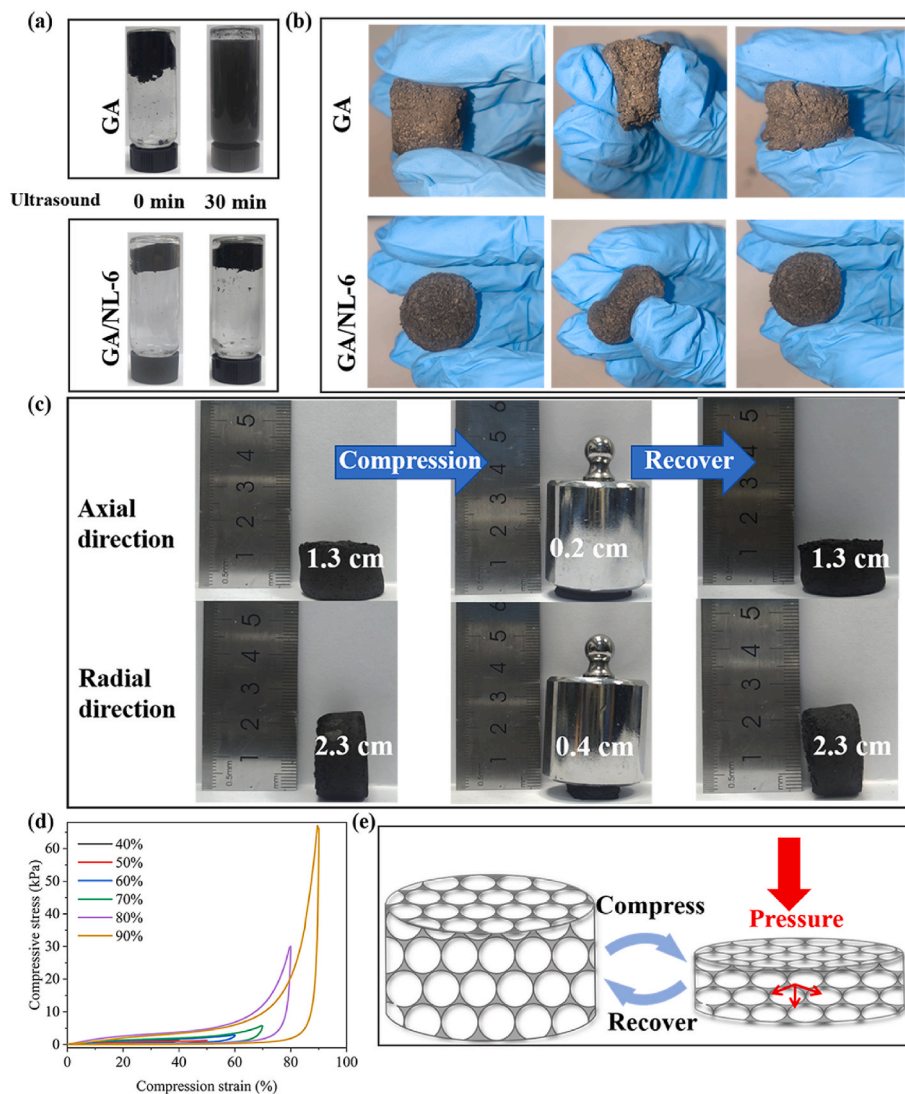


Fig. 2. (a) Images of GA and GA/NL-6 after 30 min of ultrasound treatment. (b) Schematic illustration of the bending of GA and GA/NL-6. (c) Schematic of axial and radial compression of GA/NL-6. (d) Stress-strain curves of GA/NL-6 under different strain levels. (e) Schematic showing the changes in the pore structure of GA/NL during compression.

(Instron 5967, USA). The absorbance of the dye wastewater before and after purification was characterized by UV–Vis spectrophotometer (Shimadzu UV-2600, Japan) where the wavelength range was 300–800 nm. Ion concentration in water was measured by inductively coupled plasma optical emission spectroscopy (ICP-OES, Agilent 5110, USA).

2.5. Steam generation experiments

The steam generation experiments employed a xenon lamp (CEL-S500, CEAULIGHT, China) to simulate sunlight. The light intensity was calibrated by a photo radiometer (VLP-2000, China), and it was maintained at 1 kW m^{-2} . The samples were placed in a beaker containing 200 mL of test liquid (pure water, brine, or seawater), and expanded polystyrene (EPS) was used to fill the remaining exposed water to prevent natural evaporation caused by light exposure. The real steam generation experimental set-up is shown in Fig. S1. Mass change was recorded in real-time via a precision electronic balance (Lucky YT10003C, precision: 0.001 g) connected to a computer. Surface temperature distribution was monitored using an infrared camera (Testo 890, Germany).

3. Results and discussion

3.1. Mechanical properties

Solar steam generation involves the filtration and treatment of large quantities of seawater, and structural damage to the aerogel will adversely affect its evaporation performance. To evaluate the stability of the aerogel under harsh conditions, GA and GA/NL-6 were subjected to sonication in an aqueous solution. As shown in Fig. 2a, the GA exhibits graphene sheet delamination after 30-min sonication, whereas GA/NL-6

maintains its original morphology under identical conditions. This remarkable mechanical stability is attributed to the hydrogen bonding interaction between the remaining oxygen functional groups of rGO sheets and protein molecules on the NL surface.

To investigate the effect of natural latex on aerogel flexibility, bending tests were conducted on GA and GA/NL-6 (Fig. 2b). The GA developed obvious surface cracks after bending. This indicates inherent structural fragility. In contrast, GA/NL-6 maintained an intact surface structure. This flexibility arises from NL particles cross-linking rGO layers. As shown in Fig. 2c, GA/NL-6 (initial height: axial 1.3 cm, radial 2.4 cm) is compressed to 0.2 cm ($\sim 85\%$ axial strain) and 0.4 cm ($\sim 83\%$ radial strain) under a 200 g weight. Crucially, upon load removal, GA/NL-6 exhibits full and rapid recovery to its original dimensions. This superior elasticity is consistent across other GA/NL (Fig. S2). Further mechanical testing used GA/NL-6 as an example. Fig. 2d shows the stress-strain curves of the GA/NL-6 under 40–90 % compressive strain. All the curves could return to the origin, similar to the curves of previously reported elasticity porous materials [59,60]. The GA/NL-6 can withstand up to 90 % of strain without cracking and demonstrates a maximum stress of 66.9 kPa. This exceptional performance is attributed to two primary factors: the elastic bonding between NL particles and rGO building block layers, which forms a resilient network, and the unique 3D porous structure of the GA/NL [61–64]. This structure disperses pressure and prevents stress concentration, resulting in more uniform stress distribution (Fig. 2e).

Based on these tests, GA/NL demonstrates superior structural stability, flexibility, compressibility, and elastic recovery. Compared to the GA, the GA/NL displays greater suitability for interfacial solar steam generators.

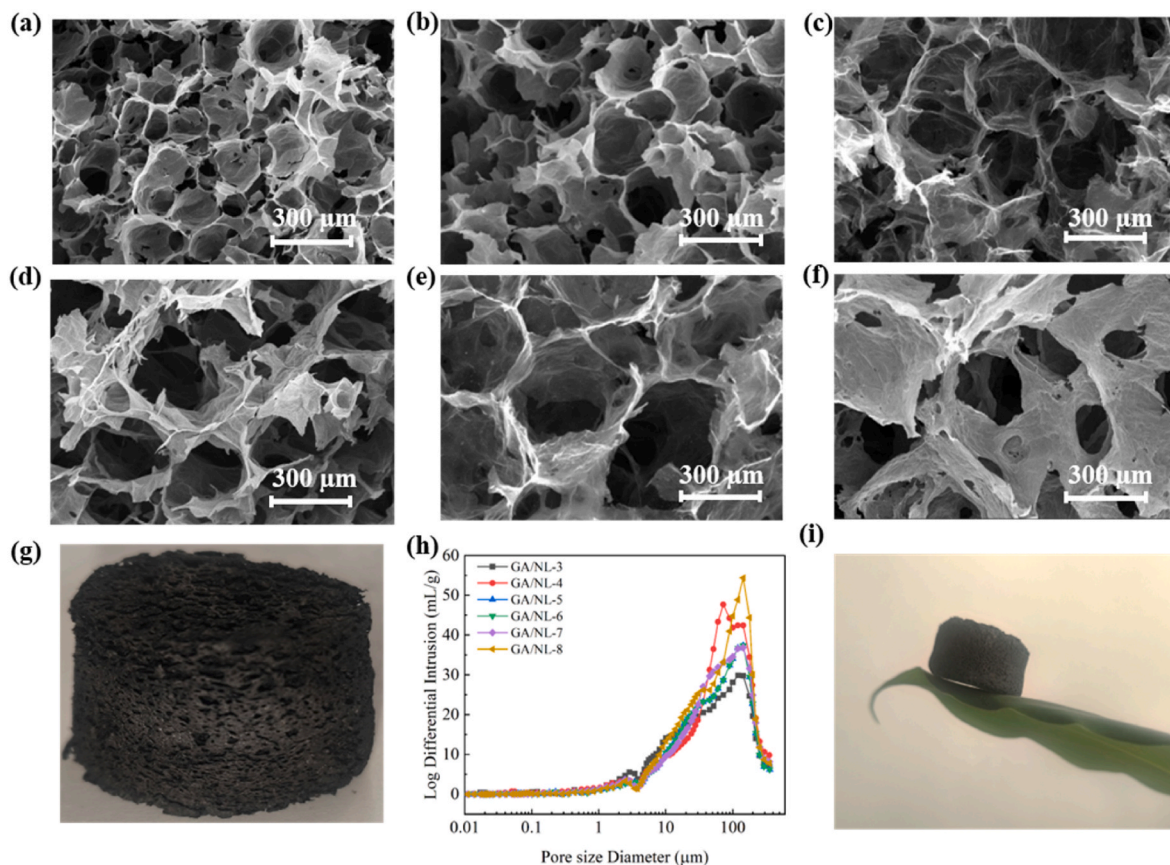


Fig. 3. SEM images of the (a) GA/NL-3, (b) GA/NL-4, (c) GA/NL-5, (d) GA/NL-6, (e) GA/NL-7, and (f) GA/NL-8. (g) Appearance of GA/NL-6. (h) Pore size distribution curves of GA/NL aerogels prepared at different rotational speeds. (i) Digital images showing ultralight nature.

3.2. Morphologies properties

The microscopic morphologies of the synthesized GA/NL were observed by SEM. As shown in Fig. 3(a–f), it is evident that the aerogel possesses a rich pore structure accompanied by a rough surface (Fig. 3g). Their pore size distribution curves are displayed in Fig. 3h, with average pore sizes and porosity summarized in Table 1. Average pore size and porosity of GA/NL prepared at different rotational speeds. Notably, all samples maintain a consistent porosity level ($\sim 97\%$), showing no significant dependence on stirring speed. This high porosity is extremely beneficial for moisture transport. As the stirring speed increases, the number of macropores within the GA/NL also increases. Their average pore diameters are 3.26, 5.21, 10.61, 11.74, 12.60, and 14.38 μm for GA/NL-3 to GA/NL-8, respectively. The GA/NL-8 displays larger and more open pores compared to GA/NL-3. This occurs because increasing the stirring speed increases the shear force in the liquid. This results in a more pronounced bubble formation process and a corresponding increase in the volume expansion of GO foam (Fig. S3). However, excessively high stirring speed will cause bubbles to burst. During subsequent drying at atmospheric pressure, capillary pressure-induced stresses cause the porous structure to collapse (Fig. S4). Therefore, the maximum stirring speed in this study was set at 8000 rpm. The density of GA/NL-8 is as low as 7.82 mg cm^{-3} (Fig. S5). It showed no visible deformation when placed on a leaf blade (Fig. 3i).

3.3. Chemical properties and hydrophilicity

X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared (FTIR) spectroscopy were employed to analyze the elemental compositions and functional groups of the aerogels. As shown in Fig. 4a, the GA/NL aerogel primarily consists of carbon (C 1s peak at 284.08 eV) and oxygen (O 1s peak at 532.08 eV). The O 1s spectrum (Fig. 4b) resolves three components at 531.48 eV (COO^-), 532.48 eV (C=O), and 533.13 eV (C-O-C). The C 1s spectrum (Fig. 4c) shows peaks at 284.6 eV (C-C) and 286.1 eV (C-O). The FTIR spectrum of GO is shown in Fig. 4d, with characteristic peaks at 3402, 1729, 1623, and 1053 cm^{-1} , which correspond to the presence of -OH , C-O in the carboxyl group, the C=C stretching vibration in sp^2 , and the stretching vibration of C-O in alcohols. These characteristic peaks indicate that the GO structure is rich in oxygen-containing functional groups. In the spectrum of NL, the peak at 3434 cm^{-1} corresponds to the stretching vibration of -OH , and the peaks at 2960 and 1448 cm^{-1} are usually attributed to the stretching vibration of the C-H bond. The GA exhibits the same characteristic peaks as GO, except for a significant decrease in intensity. In the FTIR spectrum of GA/NL the absorption bands appear at 2920, 1639, and 1460 cm^{-1} , which correspond to the characteristic peaks of GO and NL, respectively. This indicates that natural latex is successfully doped into graphene aerogel. The characteristic peaks for the in-plane bending vibration of hydrophilic hydroxyl groups (-OH) at 3404 cm^{-1} and the C-O stretching vibration at 1053 cm^{-1} remain clearly visible.

The graphitization processes of GO and GA were elucidated by Raman spectroscopy. Fig. 4e displays the Raman spectra of GO and GA/NL. Two characteristic peaks at 1350 cm^{-1} (D-band) and 1590 cm^{-1} (G-band) correspond to graphite band (G-band) and defect band (D-band), respectively. The I_D/I_G ratio is conventionally employed to evaluate the defect density and structural disorder in graphene materials [65]. The Raman spectra reveal that the I_D/I_G ratio of GO (0.94) increases to 1.34,

suggesting the reduction of GO. With the addition of NL, the I_D/I_G of GA/NL decrease to 1.1, demonstrating the weaker reduction of GO. This is because when the more alkaline NL suspension was added. It consumed a portion of the reduction agent ascorbic acid leading to more residual oxygen-containing groups on the rGO sheets.

As shown in Fig. 4f, the XRD pattern of GO exhibits a sharp peak at $2\theta = 9.3^\circ$, which corresponds to the (001) crystal plane [66]. A broad peak at 18.4° is observed for NL, indicating its amorphous structure origination from short-range ordering of cis-polyisoprene [67]. When L-Ascorbic acid was acted as a mild reducing agent, it removed a significant portion of oxygen-containing functional groups from GO, resulting in GA with a broad diffraction peak at $2\theta = 23.1^\circ$. This suggests an increase in structural disorder and the introduction of lattice defects during reduction [68]. For the GA/NL, the diffraction peak shifts to 22.9° , indicating an expansion of the interlayer spacing from 0.38 nm to 0.39 nm. This shift can be attributed to the consumption of ascorbic acid by alkaline NL, which leaves residual carboxyl groups that promote interlayer expansion through electrostatic repulsion.

The thermal stability of samples was evaluated by using thermogravimetric analysis (TGA). As shown in Fig. 4g, the GA/NL exhibits two main stages of weight loss stages. The first stage, commencing at 180°C , is attributed to the further reduction of rGO. The second stage, starting at approximately 300°C , corresponds to the thermal decomposition of NL. The observed further reduction during the heating indicates the presence of residual oxygen-containing groups on the rGO sheets. These remaining oxygen-containing groups are likely to enhance the hydrophilicity of materials.

The hydrophilicity of ISSGs ensures a sufficient supply of water during the steam generation process, which is necessary for high-performance solar evaporation [69]. Fig. 4h displays the water contact measurement process for the GA/NL-6. A water droplet upon contact with the sample surface is completely absorbed by the material within 7 s, demonstrating good hydrophilicity.

3.4. Light absorption and photothermal conversion performance

It is well-known that outstanding broadband sunlight absorption is essential prerequisite for ideal ISSGs. To evaluate the optical properties, the UV–Vis–NIR spectrophotometer was used to measure the light transmittance ($\sim 0\%$) and reflection spectrum of samples. The solar absorption spectra and reflection spectra of the representative samples GA/NL-3, GA/NL-6, and GA/NL-8 (fabricated at low, medium, and high shear speeds) are shown in Fig. 5(a and b). It is clearly that all samples exhibit excellent solar absorption and low reflectance ($\sim 7\%$). The average solar absorbance of GA/NL-3, GA/NL-6, and GA/NL-8 are 92.8 %, 93.2 % and 93.3 %, respectively. This low reflectance spectrum and high absorption spectrum lay the foundation for achieving high solar-to-steam conversion efficiency.

Generally, the strong photo-absorption of the materials suggests promising for photothermal conversion performance. To investigate their photothermal properties, the surface temperatures of dry GA/NLs were recorded by an infrared thermal imaging camera under 1sun irradiation. As shown in Fig. 5(c, d), the dry GA/NL samples exhibit a rapid temperature increase within the first 3 min and reach a thermal equilibrium within 10 min. For example, GA/NL-6 heats rapidly from 25.2 to 53.9°C within 3 min and stabilizes at approximately 69.8°C after 10 min.

During the seawater evaporation process, the photothermal materials remain continuously wetted. Therefore, the surface temperatures of the wet GA/NL samples were also recorded in Fig. 5(e, f). Evidently, the wet samples show a rapid temperature rise, reaching equilibrium within 30 min. For example, GA/NL-6 increase from 21.3 to 41.3°C within the first 3 min and eventually stabilizes at around 46.1°C after 30 min. As for ISSGs used in seawater desalination, low thermal conductivity is helpful to heat confinement. As shown in Fig. S6, the GA/NL composites exhibit very low thermal conductivity. Specifically, GA/NL-8 has a

Table 1
Average pore size and porosity of GA/NL prepared at different rotational speeds.

	GA/ NL-3	GA/ NL-4	GA/ NL-5	GA/ NL-6	GA/ NL-7	GA/ NL-8
Average pore diameter (μm)	3.26	5.21	10.61	11.74	12.60	14.38
Porosity (%)	96.53	96.94	96.89	97.58	96.68	97.39

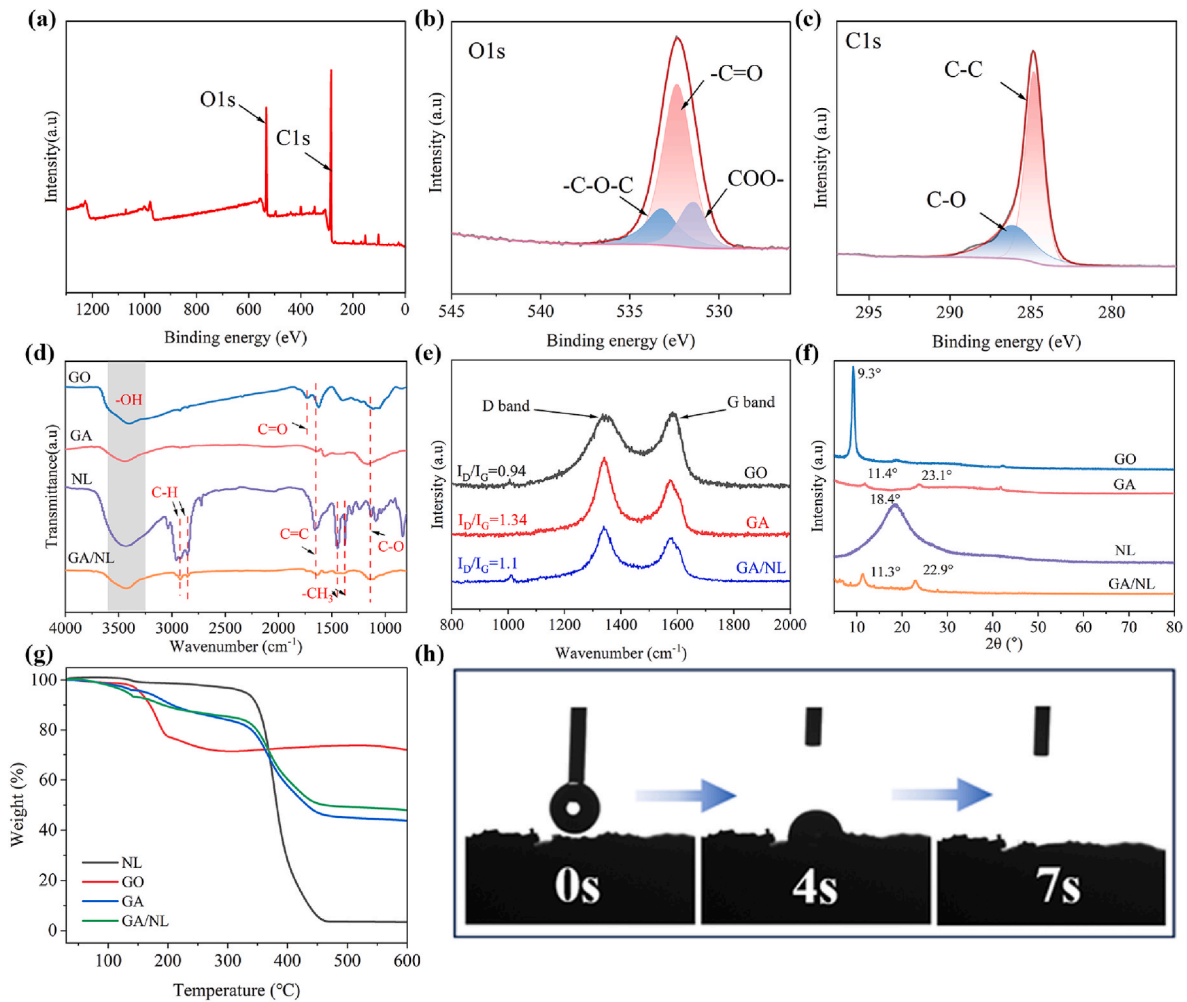


Fig. 4. (a) XPS spectrum of GA/NL. (b) O 1s XPS spectrum of GA/NL. (c) C 1s XPS spectrum of GA/NL. (d) FTIR analysis of GO, GA, and GA/NL. (e) Raman spectra of GO, GA, and GA/NL. (f) XRD and (g) TGA of NL, GO, GA, GA/NL. (h) Water contact measurement process for GA/NL-6.

thermal conductivity of only $0.038 \text{ W m}^{-1} \text{ K}^{-1}$.

These results demonstrate that GA/NL possesses outstanding photothermal conversion performance, providing a solid basis for efficient solar-driven evaporation. Their outstanding performance stems from a dual synergistic mechanism. Firstly, the sp^2 hybridized carbon framework in graphene forms an extensive π -conjugated system. Under sunlight, π - π^* electron transitions efficiently convert photon energy into thermal energy via lattice vibrational. Secondly, the high porosity and rough surface of aerogel form multiple reflective cavities. This structure enables efficient light utilization via multiple reflections within the concave cavities [69]. At the same time, the exceptionally low thermal conductivity minimizes downward heat loss, thereby improving heat localization at the surface.

3.5. Steam evaporation performance

The evaporation performance of GA/NL was assessed via steam generation experiments. As shown in Fig. 6a, when sunlight irradiates the surface of the aerogel, the light is reflected internally several times between the pores within interconnected cavities. This reflection mechanism increases the path length of the light inside the material, thereby improving light absorption. The three-dimensionally interconnected pore network generates strong capillary forces that absorb water from the surroundings.

Evaporation rate and evaporation efficiency are important indicators for judging the evaporation performance. The evaporation rate m in the

material can be calculated from the mass change of the material under illumination. The formula is as follows [70]:

$$m = \Delta m / A \quad (1)$$

where m ($\text{kg m}^{-2} \text{ h}^{-1}$) is the evaporation rate, Δm (kg h^{-1}) is the change in water mass per hour during evaporation, and A (m^2) is the illuminated evaporation area.

The solar-vapor conversion efficiency (evaporation efficiency) can be calculated by the following formula [70,71]:

$$\eta = (m_v \Delta H_v) / q \quad (2)$$

where m_v ($\text{kg m}^{-2} \text{ h}^{-1}$) is the net evaporation rate of the material ($m_v = m - m_{\text{dark}}$), q (kW m^{-2}) is the incident solar power density; ΔH_v (kJ kg^{-1}) is the enthalpy of evaporation of water from the material, which can be calculated by the following equation [72]:

$$\Delta H_0 m_{\text{water}} = \Delta H_v m_{\text{dark}} \quad (3)$$

where ΔH_0 is the theoretical evaporation enthalpy of pure water, m_{water} ($\text{kg m}^{-2} \text{ h}^{-1}$) is the evaporation rate of pure water under dark conditions, and m_{dark} ($\text{kg m}^{-2} \text{ h}^{-1}$) is the evaporation rate of the material under dark condition.

Fig. 6b shows the water mass change curves over time for each sample under 1 sun. Clearly, the evaporation rate of GA/NL increases with higher stirring speeds. The average evaporation rates for GA/NL-3, GA/NL-4, GA/NL-5, GA/NL-6, GA/NL-7, and GA/NL-8 are 1.63, 1.74,

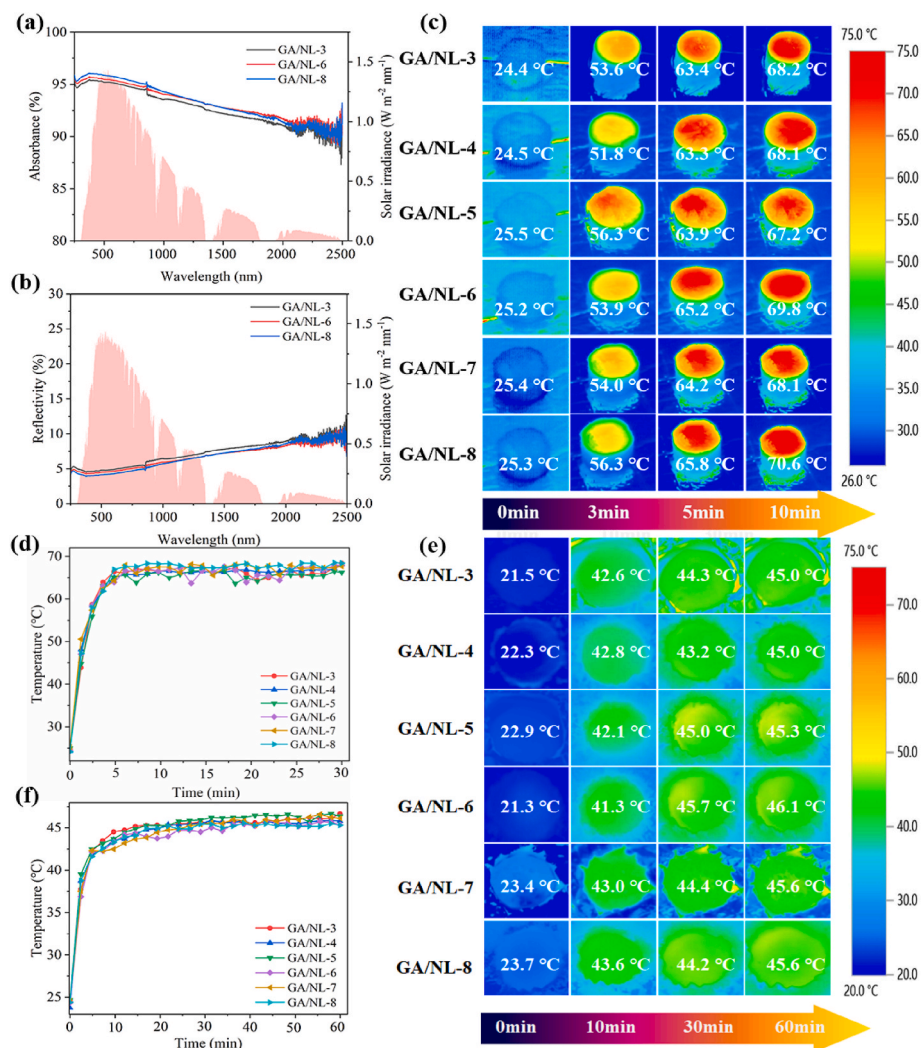


Fig. 5. (a) Absorbance and (b) reflectance of GA/NL-3, GA/NL-6 and GA/NL-8 across the wavelength ranges of 250 – 2500 nm. (c) Infrared images and (d) temperature change curves for the dry samples under one sun irradiation. (e) Infrared images and (f) temperature change curves for the wet samples under 1 sun irradiation.

2.06, 2.25, 2.49, and 2.68 kg m⁻² h⁻¹, respectively. The GA/NL-3 and GA/NL-4 show lower evaporation rates. This is due to smaller internal pores and some closed pores within the aerogel. These features limit rapid internal water transport, causing insufficient water supply at the evaporation surface. The evaporation rates of GA and GA/NL-6 prepared at the same stirring speed are different. The average evaporation rate of GA is lower than that of GA/NL-6. This is attributed to the incorporation of NL-derived hydrophilic groups, which enhance water transport kinetics and diminish hydrogen bonding interactions between water molecules. These effects collectively reduce the energy barrier required for evaporation.

To investigate the effect of natural latex addition on the enthalpy of evaporation of the sample. DSC was used to measure the enthalpy of water evaporation in pure water, GA and GA/NL-6. The measured evaporation enthalpy of pure water is 2422 kJ kg⁻¹ (Fig. S7a). The enthalpy of GA/NL-6 is 2204 kJ kg⁻¹, lower than that of GA (1952 kJ kg⁻¹). DSC fails to precisely measure the heat dynamics during actual evaporation because it only measures the water stored in GA/NL-6, whereas GA/NL-6 continuously supplies water through its unique structure in the actual evaporation process. To more accurately estimate the evaporation enthalpy of the samples, dark evaporation experiments were conducted. After obtaining the dark evaporation rates (Fig. S7b), the equivalent evaporation enthalpy of GA/NL-6 was calculated using

Equation (3). The result was 1577 kJ kg⁻¹, significantly lower than the measured value. The evaporation enthalpy values of other samples measured by the dark evaporation method are summarized in Table S1. The evaporation enthalpy of NL-incorporated samples is significantly lower than that of pure water. This reduction effectively facilitates water evaporation by diminishing the energy barrier that water molecules must overcome for the liquid-to-vapor phase transition.

The average evaporation efficiency of GA/NL is shown in Fig. 6c. GA/NL-3 has the lowest efficiency of 67.84 %. GA/NL-6 showed an efficiency of 83.40 %, while GA/NL-8 achieved the highest efficiency of 94.54 %. This is attributed to the larger internal pore size of GA/NL-8, which facilitates continuous and stable evaporation. The interconnected porous network and capillary action effectively ensure the upward transport of water, enhancing water delivery capacity. Fig. 6 (d, e) show the water evaporation rates of the GA/NL-8 under different irradiances. The evaporation rates under 0.5, 1, 1.5, and 2 sun are 1.30, 1.58, 3.75 and 4.53 kg m⁻² h⁻¹, respectively. It is evident that the evaporation rate increases with the increase in solar intensity, demonstrating stability across a range of irradiance levels and exhibiting good environmental adaptability.

Comparing the evaporation performance of the GA/NL-8 with previously reported ISSGs using graphene as a photothermal conversion material. It is found that the evaporation performance of GA/NL-8 is

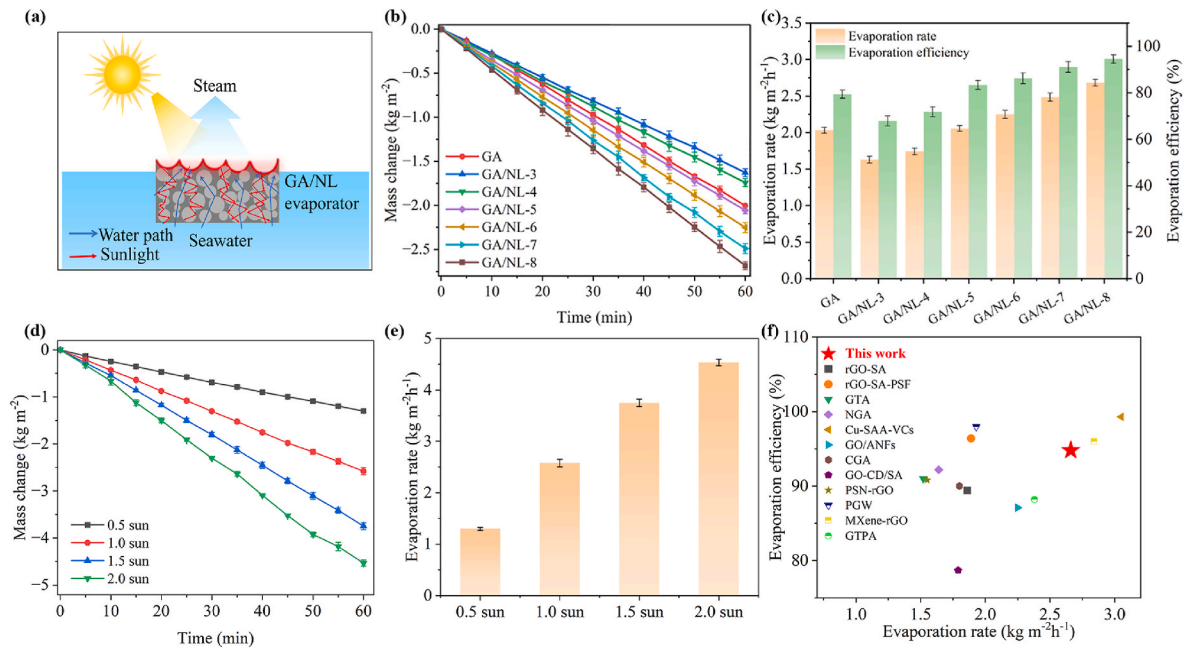


Fig. 6. (a) Schematic diagrams for highly efficient steam generation from GA/NL. (b) Water mass change curves over time for each sample under 1 sun. (c) Water average evaporation rates and efficiency for samples. (d) Water mass change curves over time and (e) average evaporation for GA/NL-8 under different irradiation. (h) Comparison of the evaporation performance of GA/NL-8 with other graphene-based aerogels.

higher than most reported ISSGs (Fig. 6f). More detailed data is in Table S1. This is mainly attributed to: (i) efficient light absorption of GA/NL across the solar spectrum; (ii) a rough and porous surface, which increases the effective area for interfacial evaporation and facilitates the rapid escape of water vapor; (iii) the capillary action and hydrophilic properties of the pore structure within the loosely packed porous network of the aerogel, which accelerate the transport of water to evaporation interface.

3.6. Salt resistance and durability performance

The long-term stability of ISSGs during seawater desalination and the precipitation of salt crystals have always been problems that limit practical applications. To evaluate salt resistance, GA/NL-8 was selected for evaporation tests lasting 6 h in brines of varying concentrations. As shown in Fig. 7a, GA/NL-8 exhibits good evaporation performance in different brine concentrations. In 3.5 wt% brine, it achieved an evaporation rate of 2.52 kg m⁻² h⁻¹ (only 0.05 % lower than in water). Even in 20 wt% brine, GA/NL-8 maintained an evaporation rate of 1.72 kg m⁻²

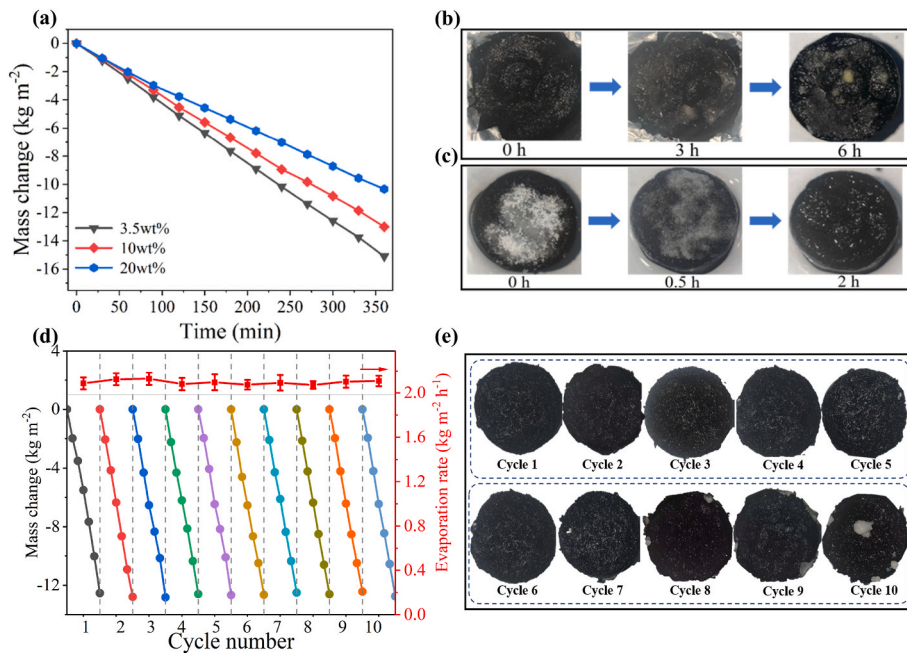


Fig. 7. (a) Mass change of and GA/NL-8 in different concentrations of brine. (b) Images of GA/NL-8 after 6 h in 20 wt% brine. (c) Dissolution of 0.5 g NaCl on the surface of GA/NL-8. (d) Mass change and evaporation efficiency of GA/NL-8 during 10 cycles in 10 wt% brine. (e) Salt precipitation on the surface of GA/NL-8 after 10 cycles.

h^{-1} with merely traces of salt accumulation on its surface (Fig. 7b). Salt dissolution experiments were conducted by placing 0.5 g NaCl on GA/NL-8 (diameter: 2.3 cm) in dark conditions, then immersing it in 3.5 wt% brine (Fig. 7c). After 120 min, surface salt particles gradually dissolved, demonstrating good self-cleaning capability.

Furthermore, cycling desalination tests were performed using GA/NL-8 to evaluate. As shown in Fig. 7d, each cycle consisted of desalination in 10 wt% brine under 1 sun for 6h, followed by turn off the light for 6h, which was recorded as one cycle. It can be seen that the mass change trend in each cycle was the same, and the evaporation rate was maintained at about $2.10 \text{ kg m}^{-2} \text{ h}^{-1}$. This demonstrates that the aerogel can be used for high-efficiency desalination during the day, and rapid self-desalting during night time. After 9 cycles, no significant salt crystallization was observed. By the 10th cycle, only minimal salt crystallization appeared (Fig. 7e). This demonstrates that GA/NL-8 exhibits excellent reusability, evidenced by sustained performance stability across multiple long-term cycles.

The excellent salt resistance of the material originates from two synergistic mechanisms. Firstly, the rich porous structure within the material provides rapid water transport pathways and salt ion diffusion channels. During evaporation, a concentration gradient is established from the surface to the interior. This gradient drives the reverse diffusion of high-concentration salts back into the bulk solution [73]. Secondly, Hydroxyl (-OH) and carboxyl (-COOH) groups in the sample surround Na^+/Cl^- ions through hydration. This hydration weakens the electrostatic attraction between ions, accelerating the dissolution kinetics of salt particles. Under dark conditions, this effect increases salt dissolution rates by over 40 % [74].

As an interfacial solar steam generator, the structural stability of the material is critical for practical desalination applications. To validate its structural stability, the GA/NL-8 was dried after cyclic evaporation and characterized. As shown in Fig. 8a, FTIR shows retained functional groups: broad peak at 3396 cm^{-1} (-OH stretching), and 1640 cm^{-1} (C=C vibration), confirming hydrogen-bond crosslinking between NL proteins and rGO remains intact after cycles. The characteristic peak at 25.3° (graphite layer stacking) is still retained in the XRD spectrum (Fig. 8b), indicating that the crystal structure has not degraded. As shown in the TGA curves (Fig. 8c), GA/NL-8 still exhibits two weight loss stages. One is associated with the further reduction of rGO, and the other is related to the thermal decomposition of NL. It also demonstrates high thermal stability, with mass loss <5 % below 200°C confirming the stability of the matrix at operating temperatures.

Additionally, mechanical property testing was conducted on the GA/NL-8 after use. The GA/NL-8 was dried after the cyclic evaporation and then tested for mechanical properties. As shown in Fig. 9a, GA/NL-8 remained structurally sound after bending. The stress-strain curve at 60 % strain returned to the origin, with a maximum stress reaching 3.04 kPa (Fig. 9b). This indicates that the GA/NL retains good elasticity after prolonged use. The elasticity of GA/NL-8 was characterized macroscopically by placing 200 g weights. The aerogel shows the same elastic

properties as the unused GA/NL-8, which was compressed by $\sim 15\%$ of the initial height in both axial and radial directions (Fig. 9c). Once the pressure is removed, the aerogel rapidly restores to its initial diameter. This behavior indicates the presence of cross-linking between the latex particles and the rGO sheets. The GA/NL-8 demonstrates stable chemical properties, maintains pore integrity, and exhibits exceptional mechanical performance, enables long-term efficient operation in marine environments.

During solar-driven seawater desalination, the GA/NL-8 evaporator utilizes abundant micropores and larger pores to accommodate salt deposition, preventing internal channel blockage. Larger pores inhibit salt ion accumulation on the evaporator surface and enable downward diffusion along the concentration gradient. Simultaneously, the rich porosity ensures efficient water transport. Under darkness, salt dissolution dominates. The concentration gradient drives dissolved salt ions into bulk water, allowing repeatable evaporator usage. Moreover, it retains robust mechanical properties after multiple uses. Thus, GA/NL-8 demonstrates superior salt resistance and durability performance.

3.7. Solar desalination and dye water performance

To evaluate the practical application potential of the evaporator, outdoor water evaporation was carried out for GA/NL-8. The steam generated by sun irradiance was collected by a custom-built set up where the released steam rapidly condensed on the inclined transparent cover and flowed into the freshwater collection tank (Fig. 10a). In addition, the solar desalination performance was evaluated by analyzing the concentration changes of four major seawater ions from the Bohai Sea before and after treatment. Solar desalination experiments were conducted from 10:00 a.m. to 6:00 p.m. in Chengdu, China. Outdoor temperature, humidity, and solar intensity, and mass changed with time were recorded in Fig. 10b. The calculated evaporation rate was $1.78 \text{ kg m}^{-2} \text{ h}^{-1}$. This evaporation rate is lower than the laboratory rate of $2.66 \text{ kg m}^{-2} \text{ h}^{-1}$ under 1 sun, primarily due to lower average outdoor irradiance. The concentrations of Na^+ , K^+ , Ca^{2+} , and Mg^{2+} in the seawater before evaporation were 11,357, 421, 429, and 1305 mg L^{-1} . After treatment by the GA/NL-8, the concentrations of the four ions in the condensate were reduced to 17.5, 1.89, 2.21, and 0.43 mg L^{-1} (Fig. 10c), achieving removal efficiencies exceeding 99 % for all ions. These values represent reductions of two to three orders of magnitude compared to untreated seawater and are significantly below the drinking water standards established by the World Health Organization (WHO) [75]. The total organic carbon (TOC) value of seawater was measured at 1.62 mg L^{-1} . After purification, the TOC value reduced to 1.53 mg L^{-1} . Both values fall below the WHO standards (Fig. 10f).

The increasing societal demand has led to the widespread use of industrial dyes in sectors such as papermaking, textiles, and coatings. The discharge of dye wastewater causes severe environmental pollution and resource waste, making its treatment a significant challenge [76, 77]. In this study, methylene blue (MB) and methyl orange (MO)

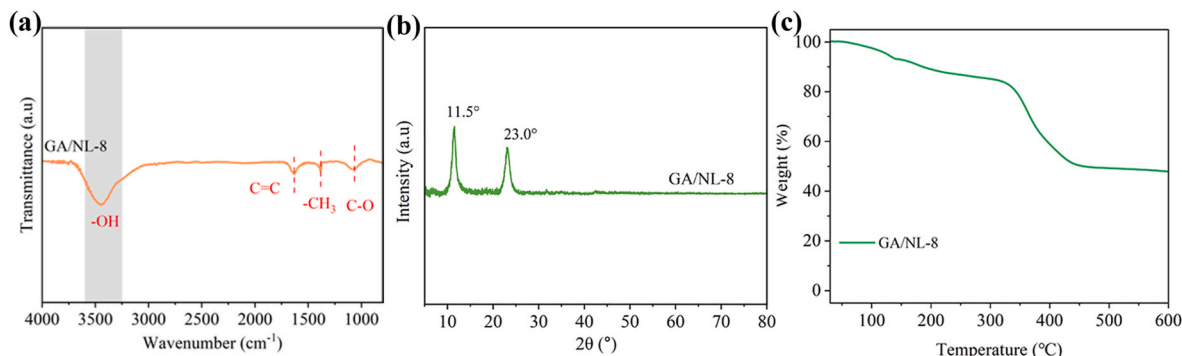


Fig. 8. (a) FTIR, (b) XRD and TGA curve of GA/NL-8 after use.

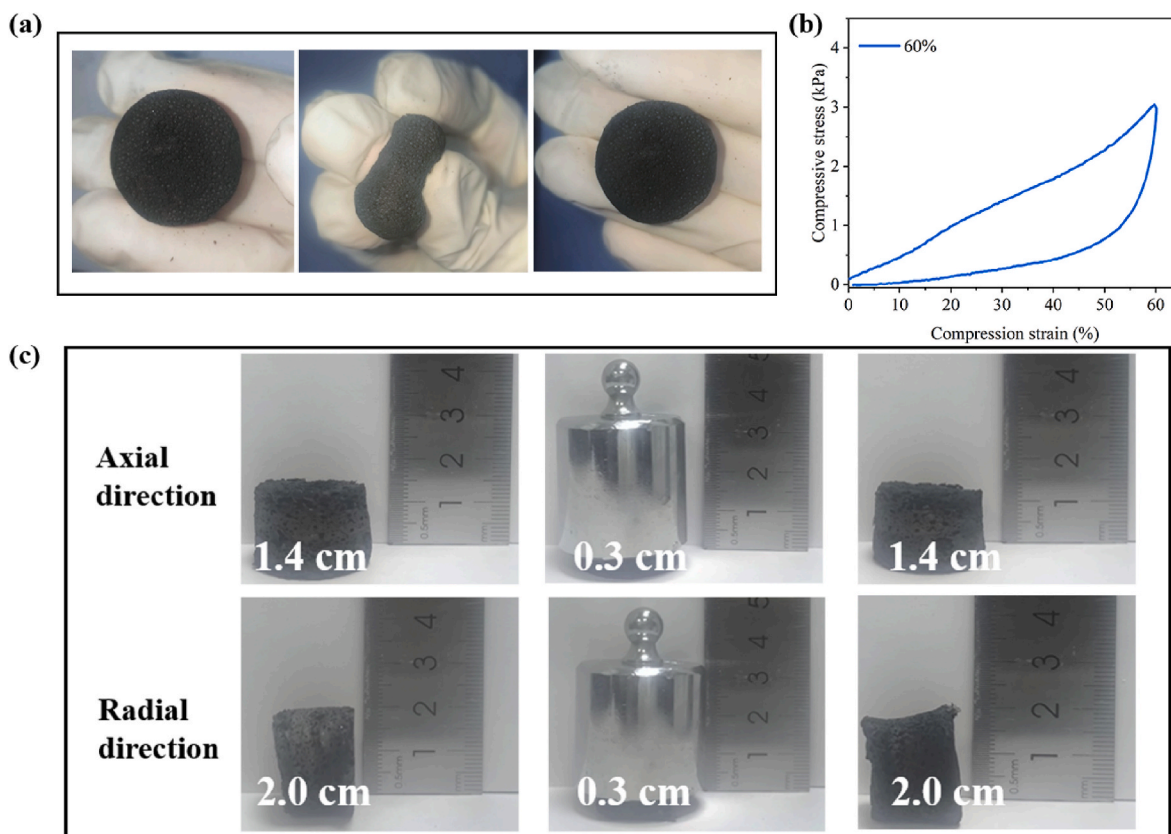


Fig. 9. (a) Schematic diagram of bending for GA/NL-8 after 10 cycles. (b) Stress-strain curve of GA/NL-8 under 60 % compressive strain after 10 cycles. (c) Schematic diagram of axial and radial compression for GA/NL-8 after 10 cycles.

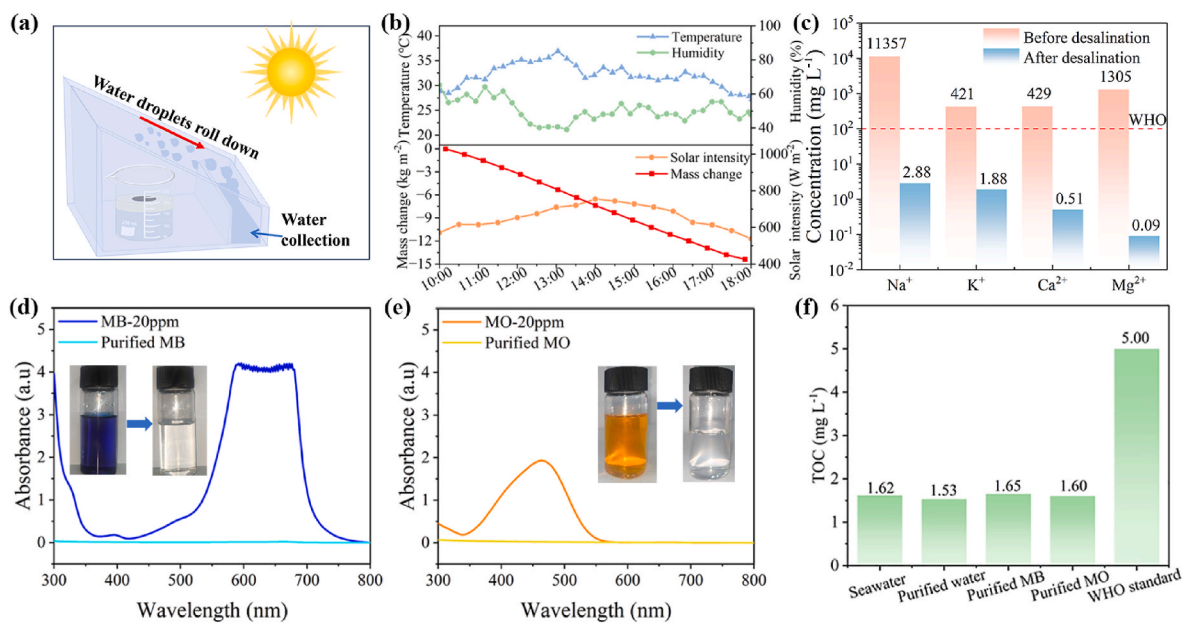


Fig. 10. (a) Schematic of steam collection device. (a) Temperature, humidity, solar intensity, and mass change with time. (c) The concentration changes of four major ions before and after treatment. (d) UV-Vis absorption spectra of methyl blue (e) methyl orange before and after purification. (f) TOC values of the seawater, purified water, MB, and MO.

solutions (concentration: 20 ppm) were used to simulate dye wastewater for evaluating the purification performance of GA/NL. The results showed that the condensate treated by the aerogel appeared transparent. The characteristic absorption peaks of methylene blue (550–700

nm) and methyl orange (350–550 nm) in the UV-vis spectra completely disappeared (Fig. 10(d, e)), and the organic dye removal rate reached 99 %. The TOC values of MB and MO after purification were 1.65 mg L⁻¹ and 1.60 mg L⁻¹, which were below the limits specified by the WHO

standard (Fig. 10f). These results demonstrate that GA/NL-8 possesses excellent wastewater purification capability and can be effectively applied in diverse aqueous environments.

4. Conclusion

This study prepares a graphene/natural latex aerogel (GA/NL) using a bubble-templating approach via mechanical stirring and ambient-pressure drying. The three-dimensional interconnected pore structure within the aerogel enhances its wettability and mechanical properties. The aerogel demonstrates the following outstanding properties:

- (1) Exceptional mechanical properties: GA/NL-6 withstands compressive strains up to 90 % without fracture, achieving a peak stress of 66.9 kPa. GA/NL-8 reaches 3.04 kPa at 60 % strain after evaporation. Structural stability of the material is maintained after ten consecutive brine evaporation cycles.
- (2) Superior photothermal conversion: The GA/NLs display a broadband absorption (~93 %) and lower reflectance. Besides, GA/NL-8 has a low thermal conductivity of approximately $0.038 \text{ W m}^{-1} \text{ K}^{-1}$, which can effectively convert light energy into heat energy confined to the surface.
- (3) Excellent evaporation performance: The GA/NL-8 achieves an average evaporation rate of $2.68 \text{ kg m}^{-2} \text{ h}^{-1}$ and an evaporation efficiency of 94.54 % under 1 sun. It maintains an evaporation rate of $1.84 \text{ kg m}^{-2} \text{ h}^{-1}$ in 20 wt% brine.
- (4) Salt resistance: After 10 evaporation cycles in 10 wt% brine, the evaporation rate of GA/NL-8 shows no significant decline.
- (5) Multi-pollutant remediation: The GA/NL-8 achieves removal rates of up to 99 % for metal ions (Na^+ , K^+ , Ca^{2+} , Mg^{2+}) in seawater and for organic dyes in dye wastewater.

Therefore, the GA/NL-8 holds great practical promise as an interfacial solar steam generator for desalination.

CRedit authorship contribution statement

Wenfeng Zhou: Writing – original draft, Validation, Methodology, Investigation, Data curation. **Xiaohan Li:** Writing – review & editing, Methodology, Conceptualization. **Nan Zhang:** Writing – review & editing, Supervision, Resources. **Zhaoli Zhang:** Writing – review & editing, Validation, Supervision. **Yangping Yuan:** Supervision, Resources, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.solmat.2025.113933>.

Data availability

Data will be made available on request.

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