

Enhancing solar evaporator efficiency and reducing salt fouling by separating evaporation surface and water pump

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ABSTRACT

Solar-driven evaporation has emerged as a promising approach for sustainable seawater purification. However, most reported methods often suffer from problems such as low efficiency, scaling, and fouling. To address these long-term limitations, this study presents a novel solar evaporator design that separates the evaporation surface from the water pumping column. This design allows the water pumping column to maintain the same salinity and temperature as the bulk seawater, minimizing scaling, fouling and heat loss. The system employs a temperature-responsive hydrogel with aligned pores to facilitate non-powered water transport driven by diurnal temperature fluctuations. Additionally, the design uses a floating graphene composite paper (GCP) evaporation surface that moves with the water level inside the evaporation chamber, automatically adjusting the angle between the evaporation surface and the sun to optimize energy absorption. Experimental results demonstrate that this configuration effectively prevents salt accumulation within the water channels, minimizes heat loss, and achieves enhanced evaporation rates, thereby offering a viable solution for efficient and sustainable seawater purification.

1. Introduction

Water evaporation plays a crucial role in purification and energy storage systems, yet conventional methods demand substantial energy input [1–4]. To support a sustainable, low-carbon future, the development of energy-efficient evaporation technologies is imperative. Recent research has explored various strategies to optimize water evaporation, including the effective utilization of diverse energy sources and advanced surface treatments that enhance condensation and evaporation processes [2–8]. Among these, solar-driven evaporation stands out as a particularly promising approach due to its reliance on renewable energy, eliminating the need for artificial power sources or energy conversion [9–11]. Significant progress has been made in designing passive and active materials to improve evaporation efficiency, such as tunable surfaces that modulate thermal emissivity to minimize heat loss [13,14]. Nanostructured and nanoporous materials have also demonstrated exceptional potential by enhancing heat transfer and evaporation rates due to their unique physicochemical properties [15,16]. Membrane distillation leverages hydrophobic membranes to separate water from contaminants via vaporization at lower temperatures than conventional distillation [17,18].

Solar-based evaporation is especially valuable for seawater

desalination, offering an energy-efficient and sustainable solution for freshwater production [1]. Floating solar evaporators, for instance, provide cost-effective and space-saving configurations. Advanced photothermal materials, such as graphene and carbon nanotubes, have been engineered to maximize light absorption and heat generation [19,20], while other innovative materials further improve solar-to-thermal conversion efficiency [21–23]. Additionally, passive radiative cooling at nighttime enhances system performance by dissipating excess heat, maintaining lower operational temperatures, and improving overall efficiency [24,25]. These advancements have spurred interest in large-scale solar evaporation systems for industrial, agricultural, and decentralized water supply applications in arid regions [26].

Despite these innovations, practical implementation faces persistent challenges, particularly scaling and fouling, which drastically reduce evaporation efficiency [23]. As seawater evaporates, dissolved salts accumulate on surfaces, forming mineral deposits that hinder performance. Current mitigation strategies include designing evaporators with hierarchical pore structures to facilitate salt redissolution through seawater flow [27,28], as well as employing hydrophobic or hydrophilic antifouling coatings [29]. Some studies have combined 3D grooved architectures with antifouling materials to further alleviate scaling [30]. However, these solutions remain insufficient for long-term use—large

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pores, while enabling salt removal, diminish capillary-driven water flux, and antifouling surfaces degrade over time in harsh marine environments [12,15]. Moreover, maintaining direct contact between the evaporation surface and bulk seawater leads to significant heat loss, further compromising efficiency [2,23]. While most of the papers have focused on the maximum evaporation rate, the practical implementation of solar evaporators relies more on these long-term issues which are critically related to the actual evaporation rate.

To address these limitations, this study introduces a novel solar evaporator design that physically separates the evaporation surface from the seawater pathway for the first time, thereby minimizing scaling, fouling, and thermal losses all together. In the seawater pathway, the anisotropic structure of hydrogels facilitates unidirectional water flow driven by diurnal temperature variations without artificial power sources. Since the hydrogel maintains the same salinity and temperature as the bulk seawater, salt crystallization is effectively suppressed. Solar absorption occurs on an isolated surface, allowing crystallized salts to accumulate harmlessly at the chamber's base. This approach not only prevents salt deposition in water pathways but also significantly reduces heat dissipation, offering a robust and scalable solution for sustainable solar desalination.

2. Results and discussion

2.1. Water pumping and evaporation

The evaporator system consisted of a water pump, which operated based on variations in diurnal temperature, a chamber to hold the pumped water, and a solar evaporation surface for the purpose of evaporating water (Fig. 1a). Hydrogels with aligned pores, created using a crystallization-based technique, were utilized in the pumping unit (Fig. 1b). Seawater was drawn into a column composed of poly(ethylene glycol) (PEG) and a composite material termed PNPD, which consists of poly(*N*-isopropyl acrylamide) (PNIPAm) and polydimethylsiloxane (PDMS) during the night. PNIPAm exhibits reversible volume expansion and contraction changes with increasing and decreasing temperature [31]. This seawater was subsequently released during the day as temperatures increased (Table S1). The released water flowed into the chamber, moistening the surface of a graphene-coated paper (GCP, evaporation surface). The water evaporation process facilitated continuous flow from the chamber to the GCP surface. To enable the GCP to float on the pumped water's surface, a small polystyrene foam piece was attached. The GCP surface was pivotally connected to the PEG and PNPD column. As the water level in the system fluctuated, the angle (θ_e) between the evaporation surface and the horizontal plane adjusted naturally, thereby enhancing solar absorption by ensuring that the surface oriented more directly towards the sun (Fig. 2a). GCP was chosen because of its stability and easy preparation method among the

photothermal materials [19,20].

The pump unit comprised PNPD and PEG, which has been already reported [31], connected to the seawater supply. The PNPD structure included two components: a composite made from PNIPAm and PDMS acting as a valve, and a layer of PNIPAm with aligned pores (referred to as PN) situated above the valve [31]. These aligned pores functioned as water channels, facilitating the diffusion of water (Fig. 1b). The valve was created by infiltrating uncured PDMS into the aligned pores of the PN structure and subsequent crosslinking, resulting in a composite material. Upon the release of water from the PNIPAm phase, the valve composite contracted more rapidly than the PN, which enabled anisotropic water flow into the chamber (Fig. 2b) [32,33]. The aligned pore structures were developed through a process of directional crystallization of water, followed by crosslinking and sublimation of the resulting ice crystals, in accordance with previously established methods [34,35]. Initially, a monomer solution was directionally frozen in alignment with a temperature gradient [36–38]. The subsequent crosslinking of this frozen structure at low temperatures preserved the alignment, which was later achieved by sublimating the ice crystals through freeze-drying [31].

Fig. 2c and c' display the cross-sectional and top surfaces of PN, confirming the presence of aligned pores. Although the structure exhibits a superficially lamellar appearance, it is more intricate, showcasing a fine fishbone pattern, as well as large-scale interconnections among the pores [32,39]. The use of an aligned porous hydrogel possessing a lower critical solution temperature (LCST) (termed PN-low) also revealed a similarly interconnected pore structure (Fig. S1 and Table S1). Previous investigations utilizing the same directional freezing methodology have confirmed that such pores are uniformly distributed and highly interconnected, attributed to a preference for secondary nucleation in crystallization mechanisms [32,36]. Consequently, the uncured PDMS resin could readily infiltrate these interconnected open pores, resulting in co-continuous composites of the hydrogel and PDMS upon curing. As the temperature changes, volume contraction and expansion of PNIPAm on composite surfaces can be observed as previously reported (Fig. 2d and d') [40].

2.2. Responsive hydrogels and GCP

To address the temperature variations across different seasons, a comparative analysis of two pump materials with distinct lower critical solution temperatures (LCSTs)—namely PNPD and PNPD-low—was conducted (Table S1). The adaptation of PNPD-low involved the use of PN-low as its base material. Additionally, a randomly porous hydrogel (RPPN) was synthesized for further comparison. In this context, a valve (PNIPAm/PDMS) was omitted due to the absence of a sufficient open pore structure that would allow for the infiltration of uncured polydimethylsiloxane (PDMS). Furthermore, the non-porous PNIPAm

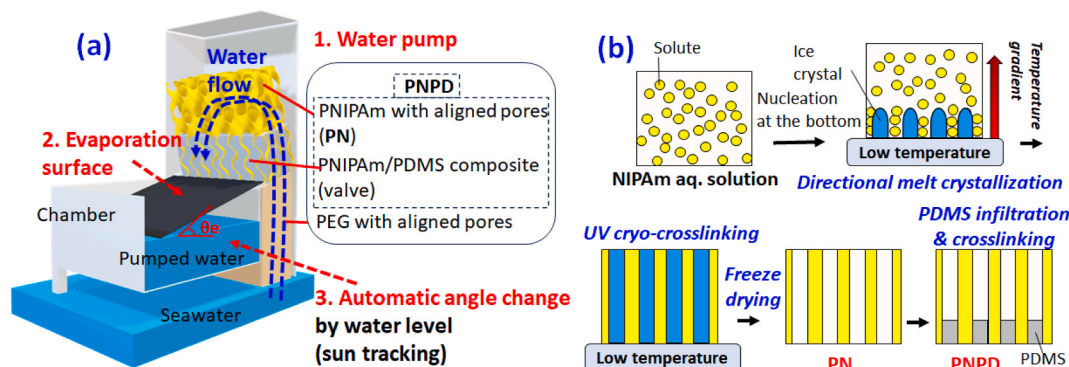


Fig. 1. (a) Structure of solar evaporator having separate evaporation surface and water pump. (b) Preparation of aligned porous PNIPAm (PN) and PNIPAm/PDMS composite (PNPD) by directional melt crystallization, subsequent freeze drying, and infiltration (height of the water pumping column = 4 cm; evaporation surface = graphene composite paper).

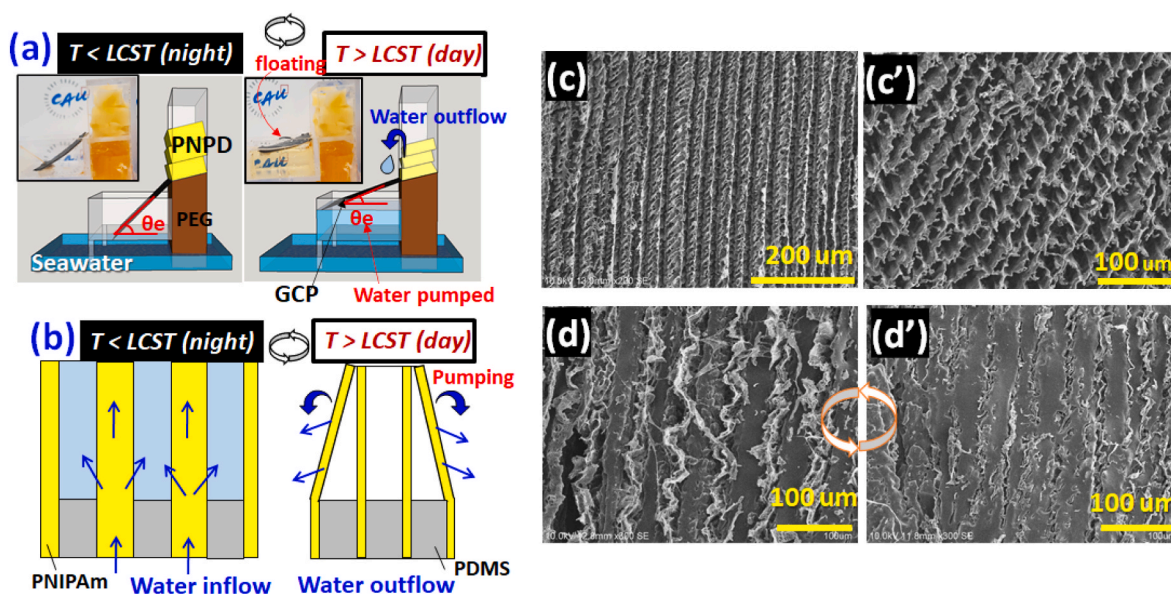


Fig. 2. (a) Powerless angle adjustment of the evaporation surface (GCP) to track sunlight by floating over pumped water (insets: photo images). θ_e is the angle between the GCP surface and the ground. (b) Pumping mechanism of PNPD composite upon diurnal temperature variation. SEM images of (c) cross-section and (c') top surface of PN; cross-sections of PNPD after equilibrium at (d) 4 °C and (d') 40 °C water.

(NPPN) was also examined without a valve since it lacked the requisite large pores to facilitate PDMS infiltration.

The incorporation of aligned pores significantly enhanced the swelling ratio and thermal responsiveness of PNIPAm [32,33,35]. The channel width was determined by the concentration of monomer solution for directional crystallization, which was selected based on a balance of mechanical durability and swelling ratio (Table S1). The hydrogel comprising PEG exhibited aligned pores too, which facilitated water diffusion through the channels connecting PNIPAm and seawater [41]. The composite of PNIPAm and PDMS, which combines a highly hydrophilic polymer with a highly hydrophobic one, demonstrated a notably quicker deswelling rate compared to porous PNIPAm, with deswelling kinetics reported to be three orders of magnitude faster [32]. Consequently, when PNIPAm expelled water in response to increasing temperature, the valve effectively prevented the backflow of water into the seawater reservoir (Fig. 2b), directing the expelled water into a chamber for evaporation. The response times can be found in our previous publication [31,32].

The characterization of the materials constituting the pump unit was conducted to ensure consistency with previous papers [31]. This included measurements of the reversible equilibrium swelling ratio and swelling kinetics to determine the transition temperature (Fig. S2 and Table S1). Both PN and PNPD exhibited a decrease in the swelling ratio at 32 °C, coinciding with the LCST of PNIPAm, while the infiltration of PDMS did not alter this transition temperature. PN-low displayed a lower LCST of 13 °C and a diminished swelling ratio, attributable to increased hydrophobicity from the introduction of additional methyl groups into the co-monomer [42]. Notably, the transition temperatures and equilibrium swelling ratios in seawater were found to be lower than those measured in pure water [41,43].

The observed pumping efficiencies correspond closely with findings from earlier studies (Fig. S3) [31]. The weights of water expelled from the hydrogels (Wd) and the pumped water (Wp) were quantified following periods of swelling and deswelling. The pumping efficiencies recorded were 86.3 % for PNPD, 93.1 % for PNPD-low, 9.2 % for PN, and 3.5 % for NPPN (Table S1). Only the pump units equipped with valves achieved efficiencies exceeding 80 %, consistent with previous reports [31]. In both PN and NPPN models, Wp was significantly lower than Wd due to the lack of a valve to avert the re-entry of water from the reservoir. The Wd and Wp values for PNPD-low were inferior to those of

PNPD, reflecting its comparatively lower swelling ratio, while NPPN exhibited reduced Wd compared to PN as a consequence of its lack of porosity. These findings affirm that the aligned pores in PNPD greatly enhanced water diffusion, resulting in improved Wd, while the valve's rapid deswelling rate contributed to high pumping efficiency. Notably, the water pumping performance remained consistent throughout multiple swelling and deswelling cycles instigated by temperature changes (Fig. S3) [31]. The structured alignment of pores not only expedited water transport from the reservoir to the pump unit but also accelerated the water deswelling process (Fig. 3a and b) [41]. The quantity of evaporated water (Wv) also saw marked enhancements as a result of the implementation of aligned pores, underscoring their critical role in the effective operation of the proposed solar evaporation system.

The preparation of GCP was validated through a comparative analysis with results from previous studies [44,45]. The reduction of graphene oxide (GO) was evidenced by a noticeable color change to a dark black hue, which was corroborated by Raman spectroscopy (Fig. S4). The Raman spectral analysis further indicated a rise in the I_D/I_G ratio for GCP, attributable to the distortion of the carbon lattice in the sp² domain during the reduction process, which resulted in the emergence of additional graphitic domains within the sp² cluster. Furthermore, scanning electron microscopy (SEM) analysis demonstrated that the cellulose paper exhibited a smooth surface following the graphene coating, indicative of a uniform layer formation. This uniformity contributed to an enhancement in light absorption efficiency. Comparative assessments between the uncoated cellulose paper (CP) and the GCP revealed that the temperature differential exceeded 15 °C, a finding further substantiated by thermal imaging (Fig. S4). Additionally, the GCP demonstrated a greater water evaporation rate (Wv) compared to the uncoated CP.

2.3. Design of solar evaporators

The proposed evaporator was conceptualized based on the temperature-responsive properties of the anisotropic hydrogel column, which facilitated the movement of water into a separate chamber and consequently reduced heat loss to the water reservoir (Fig. 1). In Fig. 3, the evaporation surfaces of B and B', which employed a conventional evaporator design, exhibited significantly lower temperatures compared to those of the separated design (A and A'). Specifically, the surfaces

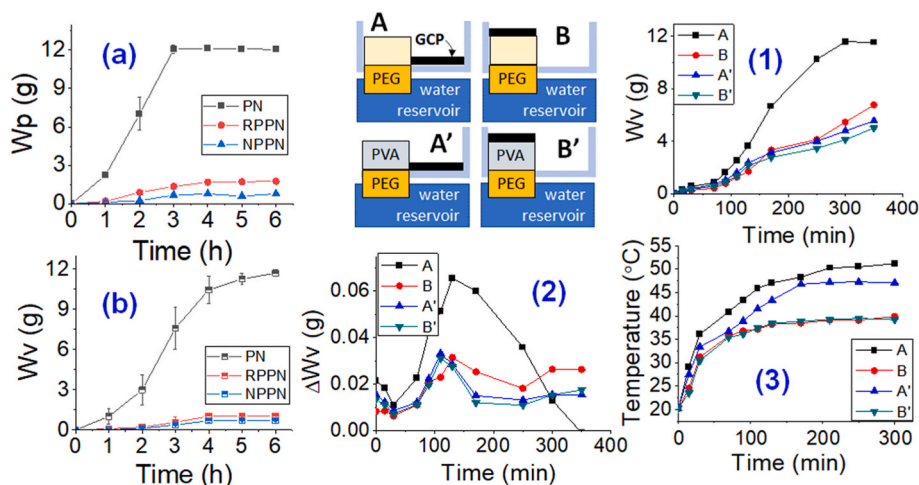


Fig. 3. (a) Weight of pumped water (Wp) and (b) evaporated water (Wv) of evaporators with PN, RPPN, and NPPN at one sun condition. Influence of the relative positions of GCP evaporation surfaces: (1) Weights of evaporated water (Wv), (2) its differentials (ΔWv), and (3) temperatures of GCP surface at one-sun condition. For comparison, PVA hydrogels (A' and B') were used in place of PNPD (A and B).

associated with the pump hydrogels in the separated design recorded temperatures exceeding those of the conventional design by more than 10 °C, a finding further corroborated by thermal imaging (Fig. S5).

In the conventional design, both PNIPAM and polyvinyl alcohol (PVA, a non-responsive hydrogel) demonstrated similar temperature profiles, and evaporation effectiveness was not enhanced (Fig. 3(1-3)). The maximization of water evaporation (Wv) was achieved exclusively with the separated design utilizing temperature-responsive hydrogels (A in Fig. 3). When PVA was utilized within the separated design, no substantial improvement in water evaporation was noted due to the lack of water flow into the evaporation chamber (A' in Fig. 3). Likewise, the combination of a temperature-responsive hydrogel with an evaporation surface positioned atop the hydrogel did not yield significant enhancements in evaporation performance due to considerable heat loss (B in Fig. 3). Thus, it was evident that only the synergistic combination of the separated design and temperature-responsive hydrogels facilitated effective water evaporation.

Under 1-sun condition, water flow into the evaporation chamber was completed within a duration of 3 h, with the water being fully evaporated within 5 h (Fig. 4a). The pumped water (Wp) exhibited a rapid increase, which was subsequently followed by a significant rise in evaporation rate (Wv). The proposed evaporator thus demonstrated a capacity for complete evaporation of the pumped water. PN-low exhibited faster completion of water flow and evaporation. The surface temperature of the GCP reached 32 °C, in contrast to temperatures of 25 °C observed for both PN and PN-low (Fig. S6).

2.4. Outdoor performance

In outdoor experiments, measurements were taken to assess the temperature of each component and the volume of evaporated water (Fig. 4b-e). Following sunrise, both the PNPD and GCP exhibited increases in temperature, although at differing rates, leading to an expanding temperature differential over time. By 9:30 a.m., the surface temperature of the GCP had risen to 24.7 °C, reaching 50.9 °C by noon, primarily due to the effective radiation absorption properties of graphene. In contrast, the temperature of the PNPD remained below 35 °C during this period (Fig. 4c). In Fig. 4b, the red part of the GCP shows a temperature that is significantly different from the other parts. Throughout the day, the temperatures of both the PNPD and GCP exceeded that of the ambient atmosphere, while the temperature of the water reservoir remained lower than the surrounding air. As the PNPD reached temperatures above its LCST, the amount of pumped water

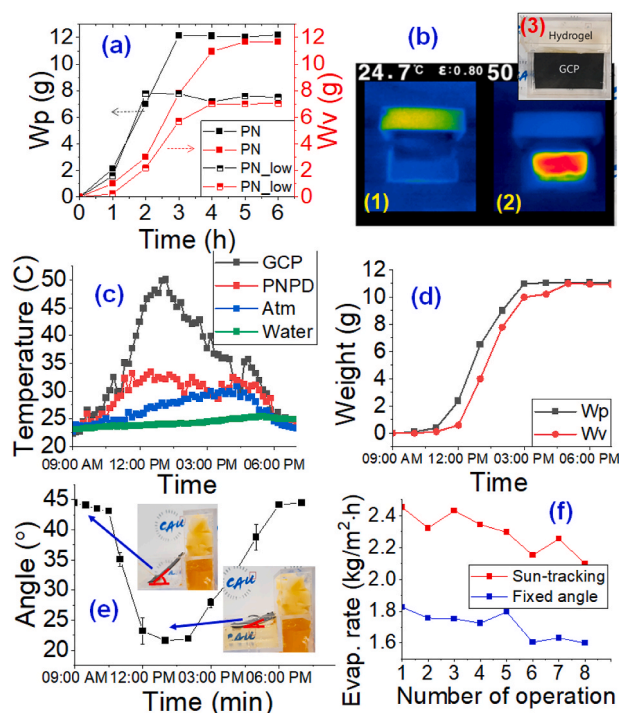


Fig. 4. (a) Wp and Wv in PN and PN-low systems. Outdoor test results for the sun-tracking evaporator, (b) thermal images of the evaporator (1) 0.5 h and (2) 3 h after 9 a.m. with (3) corresponding photographic images, (c) temperature variation of GCP, PNPD, atmosphere (Atm), and water reservoir (Water), (d) Wp and Wv, (e) change in the θ_e of the GCP surface, and (f) evaporation rates of the evaporators with sun-tracking and fixed-angle GCP.

(Wp) increased significantly from 11:00 a.m. to 3:00 p.m., subsequently followed by an increase in water evaporation (Wv) (Fig. 4d). The complete evaporation of water was achieved by 5:00 p.m. This level of performance was consistently observed over three consecutive days of outdoor testing (Fig. S7).

During the evaporation period, the angle of the evaporation surface adapted to the changing water level, orienting the GCP towards the sun, thereby enhancing both temperature and evaporation efficiency (Fig. 4e). The evaporation rate for the sun-tracking design was found to be 30 % greater than that of the fixed-angle design (Fig. 4f and S8).

Initially, the angle between the evaporation surface and the horizontal plane (θ_e) was set at 45° . As the temperature rose throughout the day, water was pumped into the chamber through the PNPD column, allowing the GCP to float on the water's surface while reducing θ_e (Fig. 4e). After evaporation, the angle returned to its original 45° position.

During nighttime, the hydrogels reverted to a swollen state (Fig. S9). In autumn (Fig. S9a), as atmospheric temperatures decreased from 10°C to 4°C between 8:00 p.m. and 8:30 a.m., the swelling ratios for PNPD, PNIPAm, and PN-low reached 98 %, 92 %, and 84 % of their equilibrium values, respectively. In contrast, during summer (Fig. S9b), adequate swelling was observed for PNPD and PN, but not for PN-low. This was attributed to the minimal difference between the nighttime temperature and PN-low's LCST. Therefore, as originally designed, PN-low is considered suitable for use at low temperatures.

Conventional designs for solar evaporators often result in the formation of sea salt crystals along the water flow path, which progressively diminishes evaporation efficiency. In contrast, the proposed design featuring a separated evaporation surface effectively facilitated the crystallization of sea salts outside the water pathway. No salt deposits were observed upon repeated cycles. Salt crystals were only detected in the bottom of the pumped water chamber containing GCP. These crystals were dissolved by an influx of fresh water during the daytime and subsequently recrystallized at the bottom following the evaporation process. As the solar evaporation cycles were repeated, the concentration of sea salts in the pumped water increased. Specifically, during the initial evaporation cycle, only a negligible quantity of salt crystals was noted, while the size of the crystals grew with an increasing number of cycles due to the recrystallization process, leading to significant crystal accumulation (Fig. S10). After several evaporation cycles, large sun-dried salt crystals, similar to those found in natural salt fields, accumulate in the bottom of the chamber [46]. These crystals still do not impede the flow of water and can be conveniently removed manually.

The comparative evaluation of our proposed evaporator design relative to prior models presents significant challenges due to considerable variability in the experimental conditions reported in previous studies. A particular area of complexity lies in assessing the effectiveness of different designs in preventing salt blockages, as techniques employed to mitigate such blockages often result in decreased evaporation rates (Fig. S11). Our innovative approach integrates a 'non-powered' water pump with a solar evaporator, effectively segregating the water channels from the evaporation unit. This design efficiently addresses the issue of salt blockage. Moreover, it minimizes heat loss to the water reservoir, leading to markedly improved evaporation rates (Fig. S11). This aspect is crucial, as many previously reported evaporators have employed strategies to prevent salt blockages through the reflux of water back into the reservoir (seawater), which has been associated with significant heat loss [27,28]. Consequently, the evaporator design presented in this study is more suited for practical applications, as it provides antifouling capabilities while preserving a high evaporation rate. Our system used diurnal temperature variation which impose additional difficulty in direct comparison with the previous reports. Furthermore, our proposed solar evaporator has been studied in the absence of a systematic optimization design between water pumping and evaporation, i.e., water flux optimization to maximize the whole system efficiency. Future optimization of evaporator systems via computational modeling will enable more energy-efficient and cost-effective solution development. Since this approach requires only cheap materials with scalable structure and does not require any extra energy, the possibility of practical implementation could be improved significantly. The amount of graphene required for our system is very small, as it only covers the surface of a filter paper. Other materials, including temperature-responsive hydrogels, are also inexpensive.

3. Conclusions

To address challenges related to water channel blockage by salt crystal formation and heat loss to the reservoir, this study introduces a novel design of a solar evaporator consisting of two separate units: a water pumping unit and an evaporation chamber equipped with a solar evaporation surface. As a result, the water pumping unit can maintain the same salinity as the bulk seawater, thereby preventing salt crystallization. The separate design effectively prevents heat loss from the evaporation surface to bulk seawater. The water pump unit is composed of a temperature-responsive hydrogel that features aligned water channels with an asymmetric structure, which includes a lower valve component. The system achieves water pumping by utilizing diurnal temperature fluctuations without artificial power sources. The design of the segregated water pump effectively prevents the accumulation of salt deposits within the channels. Furthermore, the evaporation surface is engineered to float on the water's surface within the evaporation chamber, allowing it to dynamically adjust its orientation to optimally capture sunlight throughout the day. This adaptability enhances the evaporation rate by ensuring maximum exposure to solar radiation. The separation of components in this design significantly minimizes heat loss from the evaporation surface to the seawater and salt fouling, resulting in improved overall system efficiency. As a result, these innovations contribute to the feasibility and potential application of solar evaporators in practical, real-world environments.

4. Experimental

4.1. Materials

N-isopropylacrylamide (NIPAm, >98 %) and N-tert-butylacrylamide (NTBAm, >98 %) were purchased from Tokyo Chemical Industry (Tokyo, Japan). N,N'-methylenebisacrylamide (MBA, 99 %), ammonium persulfate (APS, >98 %), N,N,N',N'-tetramethylethylenediamine (TEMED, 98 %), poly(ethylene glycol) diacrylate (PEGDA, Mn 575 g/mol), PVA, and L-ascorbic acid (99 %) were purchased from Sigma-Aldrich (St. Louis, Missouri, USA). Irgacure 2959 was purchased from BASF (Ludwigshafen, Germany). Deionized (DI) water, hydrochloric acid (HCl, >35.0 %), and 1,4-dioxane were purchased from Dae Jeong (Daejeon, Republic of Korea). Sylgard 184 A and B were purchased from Dow Corning (Midland, Michigan, USA). A highly concentrated GO dispersion in water (5 g/L, carbon ≥ 46 %, oxygen ≤ 46 %, 0.5–5 μm) was purchased from Graphene Laboratories (New York, NY, USA). Silicon wafers (diameter 100 ± 0.2 mm, P-type, thickness 525 ± 25 μm) were purchased from Pluto (Foshan, China). Sun-dried sea salt was purchased from Young Jin Green Food (Shinan, Jeollanam-do, Republic of Korea). Polystyrene foam (KENIS, Osaka, Japan; density 0.05 g/cm³) and an acrylic chamber (Daihan Scientific, Wonju, Republic of Korea) were purchased from the local hardware store.

4.2. Preparation of the pump unit

An aqueous NIPAm solution (12.5 wt%) with a crosslinking agent (MBA, 0.25 wt%) and a photoinitiator (Irgacure 2959, 0.5 wt%) was poured into a silicone mold (12.5x50x6 mm) on a silicon wafer (Fig. 1b). The concentration of monomer was selected based on the previous research [32]. A liquid nitrogen reservoir was placed under the wafer (0.5 cm between the wafer and the surface of the liquid nitrogen) for the directional melt crystallization (DMC) of water. The frozen solution was irradiated with a UV lamp (365 nm, EN-180/FE, Spectronics, Westbury, NY, USA) for 24 h at -20°C to initiate cryo-crosslinking (3 cm between the solution and the lamp). The irradiated side was flipped after 12 h. PN was prepared by removing the ice crystals from PNIPAm using a freeze-dryer (trap temperature: 80°C ; pressure: 10 Pa; EYELA, FD-2200; Seongnam, Republic of Korea) for 18 h. To remove the residual reagent, the hydrogel was immersed in excess 4°C DI water for 3 days.

The same aqueous NIPAm solution with the crosslinking agent and photoinitiator in the silicone mold with a cover glass plate was irradiated with the UV lamp for 10 h at 4 °C (3 cm between the solution and the lamp). The irradiated side was flipped after 5 h. The resulting nonporous crosslinked PNIPAm (NPPN) was put in a freezer for 20 h at −20 °C. After crystallization, the randomly porous PNIPAm hydrogel was prepared by removing the ice crystals from PNIPAm using the freeze-dryer for 18 h. The prepared randomly porous hydrogel (RPPN) was immersed in 1 L of DI water for 3 days at 4 °C (Table S1).

The PVA hydrogel was prepared from an aqueous solution of PVA (10 wt%, $M_w = 13000\text{--}23000$ g/mol, 98 % hydrolysis), glutaraldehyde (GA, 2.5 vol% of the PVA solution), and HCl (1.2 M, 50 $\mu\text{L}/1$ mL PVA solution) in the same silicone mold. Gelation was carried out for 2 h. The obtained hydrogel was soaked in DI water (1 L) for 3 days at 4 °C and placed on top of a liquid nitrogen reservoir (0.5 cm between the wafer and the surface of liquid nitrogen) for DMC. The porous hydrogel was prepared by removing the ice crystals using a freeze-dryer (trap temperature: 80 °C; pressure: 10 Pa; EYELA, FD-2200; Seongnam, Republic of Korea) for 18 h.

A degassed mixture of Sylgard 184 A and B (10:1 wt ratio) was poured into a silicone mold (6x6x0.2 cm) on a glass plate, and then PN was placed on the mixture. When the mixture infiltrated the PN to a height of 2.5 mm, the glass plate was placed in an 120 °C oven for 20 min, resulting in the PNPD composite (Table S1 and Fig. 1). A thin 0.25-mm section was removed from the bottom of the PNIPAm/PDMS composite using a razor blade. To remove unreacted molecules, the composite was immersed in 4 °C DI water for 3 days.

To prepare the aligned porous hydrogel with a low LCST (PN-low) (PNIPAm-co-PNTBAm), a monomer mixture of NIPAm and NTBAm (7:3 wt ratio, 9.25 wt%), a crosslinking agent (MBA, 0.25 wt%) and a photoinitiator (Irgacure 2959, 0.5 wt%) were dissolved in 1,4-dioxane, followed by the process for PN described above. Its composite with PDMS (PNPD-low) was prepared using the same infiltration process.

4.3. Preparation of the solar evaporator

Aligned porous PEG was used as a water channel between the seawater reservoir and the thermo-responsive hydrogels. PEGDA (15 wt %) and APS (0.3 wt%) were dissolved in DI water for 6 h under stirring. After the addition of TEMED (50 $\mu\text{L}/10$ mL), the mixture was poured into a mold (12.5x 50x 21 mm) on a silicon wafer and crosslinked at room temperature for 1 h. Aligned pores were introduced using the same DMC process as previously described. PNPD was stacked on top of the aligned porous PEG, and the PDMS composite component of PNPD was attached using a silicone adhesive (Kafuter, K-704, Nakhon, Thailand) to the hand-made acrylic chamber (Fig. 1).

GCP was employed as the solar evaporation surface (Fig. 1 and S4) [44,45]. Cellulose filter paper (Advantec, Tokyo, Japan), cleaned in 500 mL of 0.1 M aqueous HCl solution for 3 h, was immersed in 500 mL of DI water for 3 h and dried in a vacuum oven for 1 day. An aqueous GO dispersion (0.45 mL, 3 mg/mL) was deposited onto the paper (5 × 3 cm) for 1 h, which was flipped over using tweezers every 5 min to ensure the homogeneous deposition of GO. The GO-coated cellulose paper was dried at 100 °C for 2 min using a hotplate and then reduced in a 50 mM aqueous L-ascorbic acid solution at 80 °C for 3 h, followed by drying at 100 °C for 2 min. The GCP was attached to the chamber via threading so that it could move as if it were fixed with a hinge. Closed-cell extruded polystyrene foam (0.3 g) was attached to the surface of the GCP 2 mm from the lower end (Fig. S8).

4.4. Characterization of the materials

For the SEM analysis (S-3400N, Hitachi, Japan; 10–20 kV), dried samples of PN and PN-low were immersed in liquid nitrogen for 30 min and cryo-fractured. Composites of PNPD and PNPD-low were first cut using a razor blade and immersed in 4 or 40 °C water for 6 h, followed by

freezing in liquid nitrogen for 30 min and freeze drying for 18 h. To analyze salt accumulation, GCP removed immediately after the evaporation test was dried in a vacuum (10 Pa) for 24 h. All of the samples were coated in Pt for 120 s (10 nm/min) for SEM analysis. Thermal images were taken using a thermal imaging camera (FLIR E5, Teledyne Technologies, Thousand Oaks, CA, USA).

The reduction of GO was confirmed by Raman spectroscopy (DXR2xi, Thermo Fisher, Waltham, MA, USA) using samples that were vacuum-dried for 6 h. The light absorbance of GCP was measured using UV–vis–NIR ATR (Lambda 950, PerkinElmer, Waltham, MA, USA) for wavelengths from 300 nm to 2100 nm using samples wetted by seawater.

To measure the swelling ratio, the hydrogels were first immersed in 500 mL of DI water at 5 °C for 12 h, and then the temperature was raised from 5 °C to 45 °C at 5 °C intervals using a thermostat (Jeio Tech, Daejeon, Republic of Korea; accuracy ± 0.3 °C). The equilibrium swelling ratio was calculated as follows:

$$\text{Equilibrium swelling ratio (\%)} = (W_s - W_{dr})/W_{dr} \times 100$$

where W_s is the weight of the swollen hydrogel, and W_{dr} is the weight of the dried hydrogel. To assess the swelling kinetics, the hydrogel samples equilibrated in 500 mL of DI water at 40 °C for 12 h were transferred to 500 mL of 4 °C DI water or seawater, followed by weight measurement.

The water pumping efficiency was calculated as follows:

$$\text{Pump efficiency (\%)} = (W_d - W_r)/W_d \times 100$$

where W_d is the total weight of water released from the deswelled hydrogel, W_r is the weight of the released water moving back to the PEG and water reservoir. The weight of the pumped water (W_p) was calculated as $W_d - W_r$ (Fig. S3). The difference in the weight of the porous PEG before and after PNPD deswelling was converted to W_r . Seawater (3.5 wt% aqueous solution of sun-dried sea salt) was used, and three repeated measurements were used to calculate the average value. After the hydrogels were equilibrated in seawater for 6 h, the amount of water was measured for 6 h under 1 sun.

4.5. Solar evaporation tests

The evaporation performance of the proposed system was tested under 1 sun (1 kW/m^2) using a solar simulator (XC-100E, Technox, Incheon, Republic of Korea; distance between the lamp and the evaporation surface = 8.9 cm). The angle of the solar simulator was adjusted every hour based on the altitude of the sun in the Republic of Korea (37°1'N, 126°58'E). The angle between the evaporation surface and the sunlight (θ) was related to the angle between the evaporation surface and the horizon (θ_e) and sun altitude (θ_a) data from the Korea Meteorological Administration, θ (°) = $180 - (\theta_e + \theta_a)$ (Fig. 4 and S8). An aqueous solution of sun-dried sea salt (3.5 wt%) was added to a 5 L water reservoir. In order to simulate night conditions, the temperature and humidity were adjusted to 19 ± 3 °C and 30 ± 10 RH%, respectively. The evaporation rate was calculated as follows: evaporation rate = (weight of evaporated water)/(evaporation area × time).

Outdoor evaporation tests were conducted on the roof of Chung-Ang University Building 207 (37°30'13.0"N, 126°57'28.3"E). The test was conducted from September 9 to 11 (September 9: average temperature of 23.5 °C, minimum temperature of 19.1 °C, maximum temperature of 29.1 °C, average wind speed of 1.5 m/s, relative humidity of 76.5 %, 8.0 sunlight hours, cloudiness of 6.5 %; September 10: average temperature of 23.6 °C, minimum temperature of 19.5 °C, maximum temperature of 28.9 °C, average wind speed of 1.9 m/s, relative humidity of 76.0 %, 6.4 sunlight hours, cloudiness of 3.9 %; September 11: average temperature of 24.0 °C, minimum temperature of 19.9 °C, maximum temperature of 29.4 °C, average wind speed of 1.7 m/s, relative humidity of 73.1 %, 8.7 sunlight hours, cloudiness of 2.0 %). To evaluate the heat loss from the evaporation unit, a conventional solar evaporator design using PNPD,

which was dipped into a seawater reservoir, was employed as a comparison (Fig. S5) [47].

CRedit authorship contribution statement

Hoyeon Kim: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **GyuHeon Im:** Formal analysis, Data curation. **Jonghwi Lee:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: none.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.mtsust.2025.101206>.

Data availability

No data was used for the research described in the article.

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