Research article

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Free-standing reduced graphene oxide (rGO) membrane for salt-rejecting solar desalination via size effect

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Abstract: Interfacial solar vapor generation has revived the solar-thermal-based desalination due to its high conversion efficiency of solar energy. However, most solar evaporators reported so far suffer from severe salt-clogging problems during solar desalination, leading to performance degradation and structural instability. Here, we demonstrate a free-standing salt-rejecting reduced graphene oxide (rGO) membrane serving as an efficient, stable, and antisalt-fouling solar evaporator. The evaporation rate of the membrane reaches up to $1.27 \text{ kg m}^{-2} \text{ h}^{-1}$ (solar-thermal conversion efficiency $\sim 79\%$) under one sun, out of 3.5 wt% brine. More strikingly, due to the tailored narrow interlayer spacing, the rGO membrane can effectively reject ions, preventing salt accumulation even for high salinity brine ($\sim 8 \text{ wt\%}$ concentration). With enabled salt-antifouling capability, flexibility, as well as stability, our rGO membrane serves as a promising solar evaporator for high salinity brine treatment.

1 Introduction

Harvesting the abundant solar energy to evaporate water has attracted tremendous attention for solar desalination and water purification [1–11]. In the past few years, remarkable progress has been achieved in promoting solar evaporation rate by rational designs of light absorption [12–14], thermal management [15–17], and/or water path [17–21], etc. However, maintaining the high evaporation rates in long-term operation is still illusive for most reported evaporating systems because of severe salt accumulation/clogging in solar evaporators, which may even destroy the structures when treating high salinity brine. Previous works proposed several salt-rejecting strategies, such as hydrophobic materials to prevent salt adhesion [22, 23] or well-designed water channels for salt ion diffusion [24]. However, conventional hydrophobic material surfaces commonly suffer from insufficient water supply, while the increased salt-diffusing channels would lead to extra conductive heat loss, both of which will reduce the energy conversion efficiency. Enabling effective salt rejection while maintaining a high evaporation rate is still of great challenge for solar desalination thus far.

Graphene-based photothermal materials have attracted tremendous interest in solar desalination because of efficient light absorption, well-controlled microstructures, and facile fabrication [17, 25, 26]. Graphene oxide (GO), one of the most important derivatives of graphene, exhibits flexible tunability of interlayer nanosheet spacing, beneficial for the rejection of hydrated salt ions via size effect [27, 28]. However, it commonly suffers from mechanical instability in aqueous environment [29]. By precisely controlling the reduction process, the structural integrity of the reduced GO (rGO) membrane can be well maintained in water. The sheet

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interspace of rGO can be further reduced compared to the GO counterpart [29, 30], which suggests the improved ability of salt rejection induced by the size effect.

Here, we report a self-floating salt-rejecting rGO membrane by taking advantage of the efficient solar-thermal conversion of the rGO and tailored narrow interlayer nanosheet spacing. Owing to the broadband solar absorption, low cross-plane thermal conductivity, fast water permeation, the rGO membrane can enable efficient solar desalination with an evaporation rate of $1.27 \text{ kg m}^{-2} \text{ h}^{-1}$ (solar-thermal conversion efficiency $\sim 79\%$) under one sun. More strikingly, the narrow interlayer spaces among the rGO nanosheets can exclude most of the salt hydrated ions during water supply, making it a salt-rejecting and antifouling membrane. It is expected that, with the salt-rejecting ability, flexibility, and stability, our rGO membrane is a promising solar evaporator for treating high salinity brine.

2 Results and discussions

2.1 Design of the salt-rejecting rGO membrane

The as-designed salt-rejecting rGO membrane-based solar absorber (Figure 1) consists of massive parallel laminates, and each laminate is composed of numerous well-aligned rGO sheets densely stacked due to strong π - π interactions between adjacent sheets. Since the bottom of the rGO membrane touches the water surface, on the one hand, water molecules can enter the interstice or nanopores, easily pass through the nanochannels (interspacing between the adjacent hydrophilic rGO sheets), and be

pumped up by the capillary force; on the other hand, larger hydrated ions, such as Na^+ and Cl^- ions, are blocked out from the nanochannels by geometrical size effect, as will be discussed. Thus, the rGO membrane can prohibit salt ions from passing through and let water molecules go up, making it a salt-rejecting and antifouling membrane during the desalination process. Once the rGO membrane is self-floated under illumination, it can enable solar-thermal conversion and interfacial vapor generation for water purification.

2.2 Preparation of the rGO membrane

The proposed free-standing rGO membrane was synthesized from the GO suspension via hydrothermal reduction and freeze-drying methods (Figure 2A). Firstly, the GO sheets with sizes mainly in the range of $1\text{--}15 \mu\text{m}$ (Figure S1) are homogeneously dispersed in water and aligned laterally like nematic liquid crystals [31]. The GO suspension is then treated by the hydrothermal reduction at 120°C , during which process the oxygenated functional groups of the GO sheets are partially removed (transformed to rGO sheets) (see more details in Figure S2), recovering the aromatic structures of the graphene [32]. Thus, due to the stronger π - π stacking interactions, the generated rGO sheets self-assemble into the rGO hydrogel membrane, and the interspaces between the neighbored rGO sheets become even narrower. Finally, by freezing-drying, the water in rGO hydrogel is removed, while the skeleton of rGO hydrogel is perfectly kept (see Materials and methods for more details). Figure S3A and B show a typical optical image of the prepared rGO membrane, which is about several centimeters in diameter and millimeters in

Figure 1: Schematic and mechanism of the salt-rejecting solar desalination process by the free-standing reduced graphene oxide (rGO) membrane.

The blue arrow denotes one possible route for the supply of water molecules.

thickness. The rGO membrane has a density of $\sim 7.6 \text{ mg/cm}^3$ and is also mechanically foldable with robust flexibility (Figure S3C, D, and Video S1, S2), making this membrane a promising candidate for interfacial solar evaporation and desalination. Furthermore, the chemical and mechanical stabilities of the rGO membrane were carefully characterized as well. As it is immersed in different solvents (including some corrosive liquids), the prepared rGO membrane can keep its integrity after 15 days and under mechanically stirring in water (Figure S4 and Video S3).

2.3 Characterization of the lamellar rGO membrane

The microstructure of the rGO membrane was characterized by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). As shown in Figure 2B, the rGO membrane consists of many oriented layered slices, which are connected by small laminates as bridges. It can be clearly observed that each slice consists of several aligned layered laminates (Figure 2C), indicating that rGO nanosheets are stacked compactly during the reduction process due to the strong π - π interactions between adjacent nanosheets. The top view SEM image (Figure 2D) shows that there are no noticeable pores on the surface of the rGO membrane except for some wrinkles and corrugations. Some discrete nanopores (with a size of nanometer degree) can only be observed in TEM images (Figure 2E). The existence of nanopores can also be found from the isotherm of carbon dioxide adsorption. Calculations reveal that only nanopores with diameters of $\sim 1.7 \text{ nm}$ and $\sim 4 \text{ nm}$ are detected among the rGO sheets (Figure 2F), consistent with the TEM observation. As these nanopores are randomly distributed, they serve as effective entrances for the water to get into the pathways (made up by adjacent rGO sheets).

To be an efficient solar evaporator, the rGO membrane is required to possess pronounced solar absorption, sufficient water supply, and good thermal localization, as demonstrated below. Firstly, the rGO membrane exhibits broadband pronounced light absorption. Without adding any absorbing additives, the measured average absorbance of solar energy of the rGO membrane reaches above 94%, as shown in Figure 2G. Secondly, the membrane is hydrophilic (insets of Figure 2H) and exhibits good water absorption (Figure 2H). When the rGO membrane is floating on the water surface, water can be quickly supplied from the bottom to the top. The surface water adsorption ratio (the weight ratio of the rGO membrane

after and before touching the water surface) is also recorded in Figure 2H. The water adsorption reaches a dynamic balance within $\sim 10 \text{ min}$, quite close to the bulk water adsorption when the membrane is immersed in water (Figure S5), which suggests a quick and sufficient water supply during the solar evaporation process. Finally, it is also measured that, without extra thermal insulating floater, the self-floating rGO membrane could suppress the thermal conductance loss down to $\sim 8\%$ (Supplementary Note S1), showing a good thermal localization.

To effectively reject salt ions during the solar evaporation process, the interlayer spacing between the adjacent rGO nanosheets in the laminates plays a critically important role. As shown in the X-ray diffraction (XRD) spectra in Figure 3I, a diffraction peak of the rGO membrane appeared at 24.1° , shifted from the position of 11.9° of the GO membrane, indicating that the interlayer spacing between neighboring rGO nanosheets in the ultrathin laminates was decreased to $\sim 3.7 \text{ \AA}$ from interlayer distance of $\sim 8.8 \text{ \AA}$ in the GO membrane after hydrothermal reduction. This interlayer distance did not change when the membrane went through soaking treatment for 2 h (Figure S8). Such narrow interlayer spaces of rGO nanosheets allow the permeation of monolayer water molecules while exclude the hydrated salt ions such as Na^+ ions and Cl^- ions with a hydrated diameter of 7.16 and 6.64 $\text{\AA}$, respectively [29, 30, 33, 34]. As a result, the rGO membrane can impede salt ions to go into the membrane and prevent the clogging of salt in the membrane and accumulation on the surface during solar desalination.

2.4 The solar desalination performance of the rGO membrane

The solar evaporation experiments of a free-standing rGO membrane under one sun illumination were conducted to demonstrate its solar evaporation performance (Figure 3A). As Figure 3A shows, the absolute evaporation rate of the rGO membrane on pure water reaches $1.37 \text{ kg m}^{-2} \text{ h}^{-1}$ with a solar-thermal conversion efficiency (η) of $\sim 81.5\%$, stemmed from pronounced solar absorption, fast water supply, as well as good interfacial thermal localization. The evaporation rate of the rGO membrane for 3.5 wt% brine maintains $\sim 1.27 \text{ kg m}^{-2} \text{ h}^{-1}$ (referred to energy transfer efficiency of $\sim 79\%$). The ion concentrations in purified water were also carefully tracked to identify the purification effect of desalination for the rGO membrane. It can be seen that concentrations of all the

Figure 2: The fabrication process and characterizations of the reduced graphene oxide (rGO) membrane.

(A) Schematic of the fabrication process for the free-standing rGO membrane. Cross-sectional scanning electron microscopy (SEM) images of the rGO membrane at low and high (B and C) resolution. It shows that each slice consists of numerous layered laminates that are stacked together. (D) SEM image of the surface morphology for the rGO membrane. (E) Transmission electron microscopy (TEM) image of nanopores as the defects on the rGO sheets. (F) Pore size distribution of the rGO membrane calculated by carbon dioxide adsorption data. (G) Solar absorption of the rGO membrane together with the solar intensity. (H) Water adsorption ratio of the membrane. The insets are the water contact angles of the membrane measured at 0 s (the first moment of the water droplet touching the membrane surface) and after 3 s. (I) X-ray diffraction (XRD) spectra of GO and rGO membrane. GO, graphene oxide.

Figure 3: The performance of solar desalination for the reduced graphene oxide (rGO) membrane.

(A) Mass changes of pure water and 3.5 wt% brine under one sun illumination. The mass change of pure water under dark is also shown in figure. (B) Ion concentrations in seawater and in the water after desalination. Seawater with an average salinity of ~1 wt% (collected from the Bohai Sea, China) is used as the water source. The dashed yellow lines show the World Health Organization (WHO) standard of ion concentrations for drinking water. (C) Long-term solar desalination when treating brine with 3.5% salinity. Each cycle lasts for ~1 h. The ambient temperature is kept at a temperature of ~28 °C and humidity of ~30%.

primary ions in seawater (Na^+ , Ca^{2+} , Mg^{2+} , and B^{3+} , with concentration of 9524, 403, 1217, and 5.16 mg L^{-1} , respectively) are significantly reduced (to 0.157, 0.113, less than 0.001, and 0.108 mg L^{-1} , respectively) in Figure 3B, meeting the World Health Organization standard for drinking water [35]. Figure 3C demonstrates the cycling performance of the rGO membrane when treating 3.5 wt% brine. Thanks to the ion sieving of the unique lamellar structure for salt self-rejection, the membrane maintains high and stable evaporation rates during the long-term solar desalination.

2.5 The capabilities of salt rejection and antifouling for the rGO membrane

In addition to the steady-state photothermal performances in solar desalination, a more direct study of the salt-rejecting capability of the rGO membrane was conducted. A GO film-based conventional absorber is chosen for fair comparisons [18]. Note that, the mass gain of the dried absorber after cycles referred to the dried weight of that at the first circle, namely $m_n - m_1$, not $m_n - m_0$, is a justified figure of merit for salt rejection capability because it is obvious that definite mass gain appears when samples contact brine for the first time, due to surface adhesion. After each cycle of the process, the

floating GO film/rGO membrane is dried and weighed for quantifying the salt-rejecting ability. Figure 4A demonstrates the measured $m_n - m_1$ of the GO film and the rGO membrane (dry state) after each cycle of solar desalination with brine sources of different salinities (3.5, 5, and 8 wt%). It is found that the weight of the dried GO film increases significantly (an increment of 2.6 mg cm^{-2} after 10 cycles) while the rGO membrane barely gains (0.19 mg cm^{-2} after 10 cycles) when treating 3.5 wt% brine, due to inevitable defects on the surface. The mass gains of the rGO membrane are stable even when treating brine with even higher salinity (5 and 8 wt%), indicating the excellent salt rejection ability for the membrane. Meanwhile, it is observed that the salt precipitates on the surface of the GO film gradually during cycles of solar desalination while the surface of the rGO membrane is kept clean without any obvious salt aggregation all the time (Figure S6), revealing the antifouling property due to the salt rejection. To further test the performance of the rGO membrane during the practical application, the rGO membrane was used for outdoor solar desalination with 5 wt% brine. After 5 days, the final concentration of NaCl solution increases up to ~8.1 wt%. The normalized weight of the membrane is not increased, and the surface of the membrane is kept clean during the process (Figure 4B), showing a long-term salt-rejecting ability when treating high salinity brine (>5 wt%).

Figure 4: Salt-rejecting ability of the reduced graphene oxide (rGO) membrane during solar desalination.

(A) The mass gain of the dried GO film/r-GO membrane after n cycles of solar desalination compared with that at the first cycle ($m_n - m_1$) under one sun irradiation (each cycle lasts for ~ 1 h) when treating brine with different salinities. (B) The mass gain of the dried r-GO membrane ($m_d - m_1$) during continuous solar desalination out of doors for 5 days (initial salinity of 5 wt%), 8 h each day. The final salinity reaches up to ~ 8.1 wt%. Insets are the photographs of the rGO membrane surfaces over time, yellow color on the edge of the absorber is attributed to the reflection of illumination above.

3 Conclusion

In conclusion, we reported flexible and free-standing salt-rejecting rGO membranes for efficient and stable solar desalination of high salinity brine under one sun. When combined with effective optical absorption, reduced thermal loss, as well as hydrophilic water transport channels, our rGO membrane enables the one-sun evaporation rate of $1.27 \text{ kg m}^{-2} \text{ h}^{-1}$ and energy conversion efficiency of 79% for 3.5 wt% brine. Beneficial from the controlled narrow interspaces between adjacent rGO nanosheets, the membrane can realize long-term stable salt-rejecting solar desalination even when treating high salinity brine (higher than 5 wt%). Therefore, our rGO-based membrane provides a promising alternative for stable, efficient, and antisalt-fouling solar desalination out of high salinity brine.

4 Materials and methods

4.1 Fabrications of the GO membrane and lamellar rGO membranes

GO was synthesized from the natural graphite powders by a modified Hummers' method [36, 37]. GO suspension of 5 mg mL^{-1} was prepared in water. After that, 2.8 mL GO suspension was added into a 50 mL Teflon

line, then put it in an autoclave. The autoclave was put in an oven at room temperature. Then, the temperature was increased to 120°C and kept for 12 h. When the temperature was cooled down, the rGO hydrogel membrane was obtained. The gel membrane was frozen at -30°C . The rGO membrane was produced by freeze drying the frozen gel membrane for 24 h. The rGO membrane with a hundred-micrometer thickness was obtained by putting the freeze-dried rGO membrane in water for several hours and then drying it at room temperature.

The GO membrane was prepared by the following two steps. First, the GO membrane on paper substrate was prepared by spraying the GO suspension onto a filter paper, dried for 1 h, frozen at -30°C for several hours, and then freeze dried. Then, the free-standing GO membrane was peeled off from the paper substrate. This membrane was just used for comparing its structures with the rGO membrane (Figure S1 and S2).

4.2 Characterizations

The flake size of GO sheets and the morphology of the rGO membrane were characterized by a scanning electron microscope (SEM, Dural-beam FIB 235, FEI Quanta 200). The as-prepared GO and rGO membranes were characterized by XRD (Bruker, Cu K α , $\lambda = 0.15406 \text{ nm}$) in the 2θ range of 5° – 50° (with a step size of 0.02° and recording rate of 0.2 s) to study the crystallization of GO and rGO sheets and measure their interlayer spaces. The light absorption of an rGO membrane in a full absorption spectrum was measured by ultraviolet-visible-near-infrared spectrophotometer equipped with an integrating sphere (weighted by AM1.5G solar spectrum). The surface chemical compositions of GO and rGO membranes were analyzed by X-ray photoelectron spectroscopy (Thermo Fisher Scientific,

Al K α source). Contact angle measurements were carried out by a contact angle goniometer. The adsorption–desorption isotherms of carbon dioxide were measured at room temperature with a Quantachrome Autosorb-IQ-2C-TCD-VP analyzer. The pore size distribution was calculated based on the density functional theory model. The concentrations of ions in salt solutions before and after desalination were monitored by virtue of ICP-OES (PerkinElmer Instruments, PTIMA 5300 DV). The surface temperature of the rGO membrane was measured by an IR camera (Fluke TiX 580), and the temperatures at different positions of water were measured by thermocouples.

4.3 Measurement of water adsorption

As the freeze-dried rGO membrane was placed on water, water molecules could permeate into the membrane due to the capillary force, leading to a weight increase for the membrane. Here, the water adsorption is defined as the weight ratio of water adsorbed in membrane to wet rGO membrane.

4.4 Experiments of solar vapor generation

The experiments of solar vapor generation were conducted under 1 sun irradiation (solar intensity of 1 kW m⁻²) with a solar simulator (Newport 94043A). The rGO was floated on different water surfaces (DI water, simulated salinity brine of 3.5, 5, 8 wt% with NaCl). The container of water sources is a 10 mL beaker with a height of 3.5 cm. The real-time weight loss of the water was measured by an electronic balance (accuracy of 0.1 mg) and was recorded by the connected computer. During experiments, the ambient temperature and humidity was kept at 28 °C and ~30%, respectively.

4.5 Calculations of evaporation rate and thermal conversion efficiency

The evaporation rate was calculated from the plot of the curve of mass loss as a function of time, when it reached a steady state (here calculated from 30 to 60 min). The solar–thermal conversion efficiency (η) was calculated by the formula $\eta = \dot{m}h_v / p_m$, where $\dot{m}$ is the evaporation rate, h_v is the total enthalpy of sensible heat and latent enthalpy of the liquid–vapor phase change, and p_m is the solar irradiation energy of one sun (1 kW m⁻²).

4.6 Measurements of salt adsorption and salt precipitation in and/or on the rGO membrane

Firstly, the rGO membrane was floated on surface of brine with different salinities under dark for a certain time. After that, the surface of the membrane was washed with pure water to remove the salt adsorbed on the surface and then dried in an air blowing oven at room temperature for 40 min. The membrane was treated similarly as above after each cycle of solar desalination.

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Supplementary Material: Detailed information is listed or described in supporting material, including experimental methods, GO flake size distribution, X-ray photoelectron spectroscopy (XPS) of rGO membranes, mechanical abilities of the r-GO membrane, rGO membrane in different solvents, water adsorption of the rGO membrane, rGO samples after each cycle of solar desalination with different salinities, heat loss analyses, and heat behavior of the r-GO membranes.

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