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Water desalination under one sun using graphene-based material modified PTFE membrane

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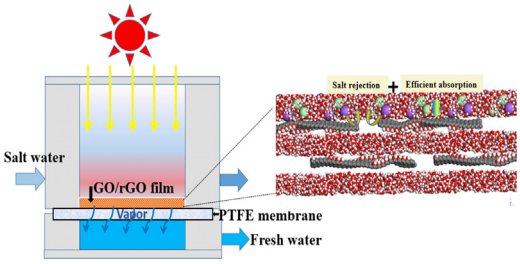
GRAPHICAL ABSTRACT



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ABSTRACT

Harvesting solar energy for water desalination is considered as one of the most important technologies to effectively address global water scarcity. Recently, it was found that water droplets on azobenzene modified anodized alumina membranes and disperse red 1 modified PTFE (polytetrafluoroethylene) membranes can permeate the membrane to be purified and desalinated under light irradiation. Herein, in order to efficiently collect vaporized and condensed water, we demonstrated the immobilization of graphene-based material n hydrophobic PTFE membrane surface for water desalination via photothermal membrane distillation (PMD). An ultrathin graphene-based film, fabricated by a scalable process, serves as efficient solar absorbers (absorption efficiency of rGO/pDA-rGO > 80%), ultrafast water permeable channels, and high salt resistance network. Compared with bare PTFE membrane, water transmembrane flux on the pDA-rGO material modified PTFE membrane can achieve as much as 78.6% enhancement under normal solar illumination. Due to high salt rejection of graphene-based films, the evaporation rate of graphene-based material modified PTFE membrane was unaffected by 4% NaCl solution, which indicated that concentration polarization effect can obviously be eliminated. These results provided new insights into the design and utilization of graphene-based films for effectively solar desalination via PMD.

1. Introduction

As water shortages and pollution become one of most urgent global challenges in our current time [1,2], solar desalination was viewed as an effective solution to produce clean water without extra energy input [3–5]. In recent years, solar-driven water evaporation, as a typical solar-thermal technology, has been extensively applied to water desalination by using novel light-to-heat conversion nanomaterials [6–21]. These localized heating nanomaterials such as porous carbon-based materials [6–17], plasmonic metallic nanoparticles film [18–21] possess broadband and efficient solar absorption and easy accessibility, which significantly improves the scalability and widespread adoption of

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the solar-thermal technology. For example, Ghasemi et al. [6] reported a novel double-layer structure that localized the solar energy and minimized the heat losses, realizing enhanced solar thermal efficiency under one-sun illumination. Ren et al. [8] utilized hierarchical graphene foam to achieve dramatic enhancement of broadband and omnidirectional absorption of sunlight, which can generate a considerable elevation of temperature. In another work, a solar desalination device, with efficient two-dimensional water supply and suppressed thermal loss, could achieve an efficient (80% under one-sun illumination) solar desalination [10]. Compared to conventional bulk water heating-based solar desalination, abovementioned works have revealed that by localizing solar energy at the air-water interface, these solar-absorbing materials can realize efficient and fast-response evaporation performance under one-sun illumination. However, few of above works consider the corresponding water vapor recovery and avoid the purified water back into original brine, which prevented them from applying to industry. Therefore, simpler and more efficient systems of solar desalination, especially in the recovery of purified water, were desired for practical water treatments applications.

In order to more simply collect vaporized and condensed water, an advanced light induced water treatment system was previously reported, where azobenzene modified anodized alumina membranes were utilized [22]. When water droplets on the azobenzene modified anodized alumina membrane were exposed to UV and visible lights, a certain volume of liquid water was recovered under the membrane by water vapor transmembrane diffusion. It is thought that the light induced permeation of water was promoted by water evaporation with the repetitive photo isomerization of azobenzene. When the NaCl solution was used in the membrane permeation process, the penetrated water could be purified and desalinated. According to this point, Fujiwara et al. [23] applied disperse red 1 modified hydrophobic PTFE membrane to replace azobenzene modified hydrophilic anodized alumina membrane. Hydrophobic PTFE membrane is more robust than the anodized alumina membrane and disperse red 1 possesses higher salt rejection than azobenzene, which is more appropriate for practical application. The improved light induced water treatment process was an important step toward the direct solar desalination. However, their selective light induced materials which were restricted to UV and visible light band (300–780 nm) both lacked efficient and broadband absorption capacity of full-spectrum solar energy (300–2500 nm), resulting in a lower efficiency of solar desalination. Meanwhile, to more accurately measure the water evaporation rate, the purified water should be recorded in real time because desalinated water droplets under the membrane were constantly evaporating with time.

In this paper, we demonstrate that the immobilization of graphene-based material on hydrophobic PTFE membrane surface can be used to construct an efficient and fast solar desalination system under one-sun illumination. Herein, rather than disperse red 1 modified hydrophobic PTFE membrane, graphene oxide (GO) or reduced graphene oxide (rGO) membrane is chosen as the absorber of the solar desalination system for several reasons including low-cost and scalable fabrication process, efficient broadband absorption efficiency, ultrafast water permeable channels, and high salt resistance network [8,10,24–27]. These excellent performance makes them more suitable to modify hydrophobic PTFE membrane for solar desalination via PMD. A GO/PTFE membrane is fabricated from GO solutions using a vacuum-assisted filtration method. The resulting GO film is reduced to rGO/PTFE membrane using hydriodic acid (HI) steam. Finally, the surface of the resultant rGO membrane is coated with a polydopamine (pDA) thin layer, obtaining the pDA-rGO/PTFE membrane. Comparing with bare PTFE membrane, high evaporation efficiency is realized in these three graphene-based material modified PTFE membrane under normal solar illumination. Furthermore, when 4% NaCl solution (slightly larger than 35,000 ppm salt content of seawater) is desalinated by the home-made photothermal membrane distillation evaporation system, concentration polarization effect has been successfully eliminated. Moreover, in order to accurately measure the evaporation rate of different composite films, the whole evaporation system is placed on a precise electronic balance which is connected to a desktop computer and the weight change is monitored and recorded by the computer in real time. Considering the simpler recovery systems of the treated water and excellent performance of selective evaporation materials, our proposed PMD evaporation system would further accelerate the practical applications of solar-driven water treatments technology under one-sun illumination.

2. Experimental process

2.1. Synthesis of GO

The preparation of GO was carried out by a modified Hummers' method [28] using natural flake graphite powder (325 mesh), which was exposed to a mixture of sodium nitrate, concentrated sulfuric acid and potassium permanganate for oxidation. Typically, 5 g of graphite flakes, 2.5 g $NaNO_3$, and 115 mL concentrated H_2SO_4 ($\geq$98%) were added into a 1000 mL beaker under stirring in an ice bath to ensure the temperature < 5 °C. 9 g of $KMnO_4$ was slowly added to control the temperature of solution lower than 20 °C for about 1 h. The solution was then transferred to a water bath at 35 ± 3 °C. After stirring for 1 h, the solution was slowly diluted by 200 mL of deionized (DI) water, causing violent effervescence and an increase in temperature to 90 °C. The diluted solution, now brown in color, was maintained at this temperature for 30 min. At the end of 30 min, the solution was then further diluted by approximately 250 mL DI water followed by injecting 5 mL of H_2O_2 dropwise, turning the solution color from dark brown to bright yellow. After settled overnight and removed supernatant liquid, the remaining solid material was then washed and subsequently centrifuged (5000 rpm for 10 min) by turns to remove the acid until the solution became neutral (pH = 6.0–7.0). The remaining solid was vacuum-dried overnight at 45 °C to obtain graphite oxide flakes.

2.2. Preparation of GO/PTFE & rGO/PTFE & pDA-rGO/PTFE membranes

The 30 mg of graphite oxide flakes was dispersed in 100 mL of cyclohexanone containing 1 mg of PVDF powder as a binder material by ultrasonication for 5 h to make a stable GO dispersion [29]. Then, 1 mL of GO solution at the concentrations of 0.3 mg/mL was deposited onto a porous hydrophobic PTFE membrane filter (50 mm in diameter, 0.22 μm pore size) to form a GO/PTFE film through vacuum filtration. Next, the stable GO/PTFE composite film was obtained by vacuum drying at 45 °C for > 6 h [30,31]. To reduce the resultant GO laminates on the PTFE membrane, the GO laminates were exposed to hydriodic acid (HI) vapor until the GO membrane completely turned black within 3–5 min, indicating that GO was reduced to rGO by HI vapor [32,35]. The rGO membrane was left at room temperature for 5 h to remove the HI acid. The rGO membrane was then fully dipped in a 2 mg/mL dopamine solution (pH 8.5) for 1 h and the dopamine solution was prepared by adding dopamine hydrochloride to a Tris-HCl buffer [32]. The resulting pDA-rGO/PTFE membrane was vacuum-dried overnight at 45 °C. The specifications of all used chemicals were tabulated in Table S2 (Supporting Information).

2.3. Membrane characterization and property measurement

The characteristics (e.g., surface morphology, hydrophilicity and functional groups) of the prepared GO, rGO and pDA-rGO membranes were investigated. The membrane hydrophilicity was determined by measuring the contact angle using a Contact Angle Meter (DMo-701SA, Japan). Fourier transform infrared spectra were separately measured in a FTIR spectrophotometer (FTIR 5700, Nicolet) over the range of 4000–500 cm^{-1} to compare the functional groups of different films. The optical absorption of dry GO (or rGO、pDA-rGO)/PTFE composite films were measured from 300 to 2500 nm wavelength using the UV–vis