



# Synchronous steam generation and photodegradation for clean water generation based on localized solar energy harvesting

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## ABSTRACT

Solving the problems of water shortage and water pollution is a vital challenge for sustainable development. Different strategies, such as photodegradation, solar distillation, and filtration, have been proposed to purify contaminated water and generate clean water. Energy-efficient clean water generation technologies play a critical role in augmenting freshwater resources. Solar energy has the potential to increase sustainable clean water production, which is a key facet of the water-energy nexus. In this study, a novel strategy to generate clean water was realized via a synchronous solar distillation and photodegradation technology based on localized solar energy harvesting using a trifunctional solar energy absorbing membrane. The membrane is composed of mixed cellulose ester (MCE) membrane, hedgehog-like hierarchical ZnO particles (HP) and gold nanoparticles (Au NPs). By floating the MCE/HP/Au membrane on rhodamine B contaminated water, the steam generation rate could reach up to  $\sim 8.70 \text{ kg}/(\text{m}^2 \cdot \text{h})$  and the concentration of organic pollutant in the residual water could be reduced to  $\sim 30\%$  within two hours' solar light irradiation. It was found that the coupling between the hierarchical HPs and plasmonic Au NPs could enhance the solar light absorption and energy conversion for photodegradation and thermal generation. This work provides a new strategy for the high efficient utilization of solar energy for the clean water generation.

## 1. Introduction

The rapid growth of modern society and increased energy consumption have resulted in severe environmental problems, which are great challenges for continued human development, especially air and water pollution [1–3]. The contamination of natural water sources has resulted in a shortage of clean water all over the world. In particular, in a few special regions and environments, such as naval island stations, sea voyages, and remote communities, clean water must be produced on-site [4]. One efficient strategy to generate fresh water is to reuse the contaminated water via a series of decontamination steps, including photodegradation [5,6], filtration [7], and distillation [8]. The utilization of solar energy is one of the most promising methods for clean water production because of its sustainability and cleanness [9–11].

One efficient method to utilize solar energy for water purification is photocatalysis, which uses a photocatalyst to absorb solar light and form highly active species (such as  $\text{H}_2\text{O}_2$ ,  $\text{OH}^-$ ,  $\text{O}_2^-$ , and  $\text{O}_3$ ) [12,13]. These active species react with contaminants and degrade the organic compounds into small nontoxic molecules. However, most photocatalysts can only absorb part of the solar light within a certain range of

wavelengths and the remainder of the solar energy cannot be efficiently utilized [14]. For instance, zinc oxide (ZnO) has been extensively studied because of its attractive properties in photocatalysis, which has resulted in its use for the removal of highly toxic and nonbiodegradable pollutants from water [15]. While ZnO has only a narrow light absorption band in the ultraviolet region, which limits the photocatalytic efficiency of ZnO [16]. Different nanostructures, such as nanorods (NRs), nanobelts, nanosphere, show enhanced photocatalytic efficiency. In addition, plasmonic nanoparticles have been introduced to expand the usage of wavelength range outside of the absorption range of the photocatalyst [17]. However, even within the absorption band of the photocatalyst, some of the absorbed solar energy is converted into the thermal energy.

Besides photocatalysis, the use of solar thermal energy for steam generation is also another promising strategy for water purification [18,19]. Unlike photocatalytic reactions, solar energy is converted into heat, which is used for water evaporation to generate clean water. Solar steam generation through localized solar heat has attracted significant attentions recently. In this process, solar irradiation at the water–air interface mediates fast water evaporation [20]. Carbon materials (e.g.,

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carbon black, carbon nanotubes, and graphene) have been applied to absorb solar light and heat the water at the water–air interface to enhance water evaporation [21–23]. Nanofluids, a kind of fluid containing nanometer-sized particles, was defined and proposed by Choi and Eastman firstly [24]. Chen et al. [25] prepared recoverable multi-walled carbon nanotubes (MWCNTs) nanofluids in saline water and the evaporation efficiency was enhanced ~76% during the solar steam generation process. Ghasemi et al. [26] used a double-layered porous carbon foam to absorb solar light and achieved efficient steam generation without the heating of the bulk fluid. Liu et al. [27] developed a paper-based Fe<sub>3</sub>O<sub>4</sub> membrane for wastewater treatment via solar irradiation, in which the magnetic nanoparticles were used as the solar absorber. In addition, plasmonic nanoparticles (NPs) have been introduced into the interfacial evaporation system, and this system realized good water evaporation performance. Zhou et al. [28,29] fabricated a broadband solar light absorber via the 3D self-assembly of plasmonic NPs (Au NPs or Al NPs) and realized a highly efficient seawater desalination method. Amjad et al. [30–32] investigated the use of Au nanofluids for steam generation using volumetric solar heating. Cheng et al. [33] investigated solar vapor generation for seawater desalination with a polypyrrole-coated membrane as an interfacial solar absorber. Ma et al. [34] developed a low-cost waste polyurethane sponge for solar vapor generation in desalination. Ye et al. [35–37] developed a kind of black TiO<sub>x</sub> nanoparticle for highly efficient solar steam generation and investigated the performance of magnetically recyclable Fe<sub>3</sub>O<sub>4</sub> thin film for highly efficient water evaporation by interfacial solar heating.

In the present work, a combination of photodegradation and solar distillation was proposed to realize water purification by expanding the range of usable solar energy, which otherwise could not be used by the photocatalyst. Furthermore, a superhydrophobic property was introduced to ensure that the solar absorber can float for long periods, allowing long-term, localized solar heating at the water–air interface. The localized solar heating means that the solar light could be absorbed and converted into heat locally at the water–air interface in the floating porous solar absorber [26]. Specifically, a trifunctional membrane was designed and fabricated to realize combined solar steam generation for water purification and the solar degradation of aqueous contaminants, synchronously. The membrane is composed of hedgehog-like hierarchical ZnO particles (HP) and Au NPs, in which the ZnO acts as the photocatalyst, Au NPs act as the solar thermal converting media, and the hierarchical structure produces the superhydrophobicity. Thus, a trifunctional membrane integrating superhydrophobicity, photocatalysis, and photothermal conversion ability was designed for the generation of clean water. Because the trifunctional membrane floats on contaminated water and is irradiated by solar light, the photocatalytic function of ZnO was able to degrade the contaminants in the water, thus cleaning it. At the same time, the steam generation function of the membrane via the localized solar heating of the Au NPs can produce clean water vapor, which can be condensed to yield purified water. This trifunctional membrane could generate different degrees of pure water and be used for different applications through this synchronous process of steam generation and photodegradation. This is an alternative approach to enhance the efficiency of solar energy utilization in solar-driven clean water generation.

## 2. Experimental section

### 2.1. Materials

The polystyrene microbead dispersion ( $\mu$ PSs, diameter: 1  $\mu$ m, concentration:  $4.55 \times 10^{10}$  particles/mL) was obtained from Polysciences, Inc., Warrington, PA, US (Catalog No.08226-15). Chemical reagents such as zinc oxide nanoparticle dispersion, zinc nitrate hexahydrate, hexamethylenetetramine, tetrachloroauric acid, sodium citrate dihydrate, Rhodamine B (RhB), 1H,1H,2H,2H-perfluorooctyl triethoxy

silane (POTS), sodium chloride (NaCl), polydimethyldiallyl ammonium chloride (PDDA), and Poly(sodium-p-styrenesulfonate) (PSS) were purchased from Sigma-Aldrich and used as received. Mixed cellulose ester (MCE) membrane (pore diameter = 1  $\mu$ m) was obtained from Shanghai Xinya Co., Ltd. Double deionized water (DDI water) was produced by a water purification system (Sartorius, Arimium® Mini; 18.2 M $\Omega$ ).

### 2.2. Preparation of HP

The preparation of HP via a sonochemical method has been previously reported in detail [38,39]. In a typical process, 125.0  $\mu$ L of polystyrene microspheres ( $\mu$ PSs) was mixed with 4 mL of a zinc oxide nanoparticle (ZnO NP) dispersion (0.025 wt%) to ensure that the  $\mu$ PSs were covered with ZnO NPs. Meanwhile, 20.0 mL of zinc nitrate hexahydrate (Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, 25 mM) was mixed with 20.0 mL of hexamethylenetetramine (HMT, 25.0 mM) as a reaction precursor. Then, the ZnO coated  $\mu$ PSs were dispersed in the reaction precursor, and the reaction was carried out by a heat-assisted sonochemical method. The ZnO NRs grow from the seeds on the  $\mu$ PSs, forming a hedgehog-like structure. The gold nanoparticles (Au NPs) were synthesized by the reduction of tetrachloroauric acid with sodium [40]. In this process, a 950.0 mL tetrachloroauric acid (0.458 mM) aqueous solution was heated to boiling and 50.0 mL trisodium citrate dihydrate aqueous solution (34.0 mM) was added. The mixture was kept boiling until the solution became wine-red, indicating that the tetrachloroauric acid had been reduced by the trisodium citrate; thus, the Au NPs dispersion was obtained.

### 2.3. Preparation of the MCE/HP/Au trifunctional membranes

A simple vacuum filtration method was applied to prepare the trifunctional MCE/HP/Au membrane, where MCE was used as a porous supporting layer. According to the contrary surface charge of HP (positive) and Au NPs (negative), a layer-by-layer strategy was applied to form a stable MCE/HP/Au membrane. First, 3.0 mL negatively charged PSS (1.0 wt%) was filtered through the MCE to make it negatively charged. Then, the 5.0 mL HP dispersion (1.0 mg/mL) was filtered for deposition on the surface of the MCE to obtain an MCE/HP membrane. Subsequently, 3.0 mL of the positively charged PDDA (1.0 wt%) was filtered through the MCE/HP membrane to enhance its positive surface charge. Finally, 50.0 mL of the Au NP solution (0.9 mg/mL) was filtered, and the Au NPs were deposited on the top surface, thus yielding the MCE/HP/Au membrane. POTS was used to modify the surface properties of MCE/HP/Au membrane to realize a superhydrophobic surface, which arises from a combination of the hierarchical structure and low surface energy of the modified membrane.

### 2.4. Characterization

The structures of the MCE, MCE/HP, and MCE/HP/Au membranes were characterized by field emission scanning electron microscopy (FESEM, SUPRA 55, ZEISS, Germany). The water contact angles of the MCE/HP/Au membranes were tested with a contact angle meter equipped with a high-speed CCD camera (JCY-2, Shanghai Fangrui Instrument Co. Ltd., Shanghai, China). The surface structure and roughness were characterized by 3D optical microscopy (OLS3000, Olympus Inc., Japan).

### 2.5. Solar steam generation tests

The experimental setup for the solar steam generation test consisted of one solar light simulator (CEL HXF300, CeAuLight Co., China), one electronic balance (Practum313-1CN, Sartorius, Germany), one data acquisition system (34972A, Agilent Technology, US), one computer, and a foam covered acrylic tube (height = 80 mm and inner

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