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Carbon nanocomposites with high photothermal conversion efficiency

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ABSTRACT Photothermal conversion for water vapor generation is a novel strategy and an efficient way to utilize solar energy, which has great potential for water purification and desalination. In this review, the development of photothermal conversion and the classification of absorbers for solar vapor generation systems are presented, especially in recent development of carbon nanocomposites (carbon nanotubes and graphene) as solar vapor generation devices. Combined with recent progresses and achievements in this field, we discuss the challenges and opportunities for photothermal conversion based on carbon nanocomposites as well as their promising applications.

Keywords: carbon composites, photothermal conversion, solar energy, water vapor

INTRODUCTION

Exploitation of abundant solar energy is an efficient strategy for achieving sustainable development of global resources. So far, the application of solar energy has covered various fields such as solar power generation, photocatalysis, and photovoltaic cells. However, photothermal conversion from solar energy is an essential way to utilize solar energy. This approach has been followed in many areas including power generation [1,2], medical sterilization or sanitization [3], fresh water production [4], and other industrial processes. Among these applications, water vapor generation is often a necessary condition. However, traditional methods cannot meet the demands of high temperature for generating solar vapor under weak solar irradiation (1 kW m^{-2}).

In industrial applications, in order to achieve sufficiently high temperatures for producing solar vapor, numerous optical concentrators (e.g., parabolic troughs, heliostats, and lenses) have been utilized to improve the solar flux by up to tens of times or even thousands of times [5]. However, using optical concentrators as major parts of these solar thermal devices is not only very expensive, but requires a vast amount of space, requiring an enormous investment.

Over the last few decades, fluids seeded with nanoparticles (NPs) as volumetric absorbers (Fig. 1a) have been used to minimize surface energy loss by ensuring a uniform temperature in the fluid and enhancing thermal conductivity [6–8]. Upon illumination, NPs can absorb the incident light and convert solar energy into thermal energy because of their photothermal properties [9–12]. When water directly contacts NPs, thermal energy is rapidly transferred from the NPs to the water, and steam generation occurs on the surface of the NPs. Nevertheless, heat loss *via* thermal radiation from bulk water results in decreased photothermal conversion efficiency [13]. To overcome the issue, the integration of light-absorbing materials into one layer (Fig. 1b) [8,14,15] floating on the surface of the water can improve the conversion efficiency, avoiding heat loss to the bulk water underneath by localizing the heat.

The photothermal conversion efficiency from solar illumination to thermal energy for solar vapor generation is calculated in the filed according to the following equation [16]: $\eta = h_{LV}/q_i C_{opt}$, where η represents the photothermal conversion efficiency and denotes the evaporation rates under illumination, h_{LV} is the liquid–vapor phase change enthalpy, including sensible heat and phase change enthalpy, q_i is the nominal solar illumination of 1 kW m^{-2}, and C_{opt} is a multiple of one sun solar intensity.

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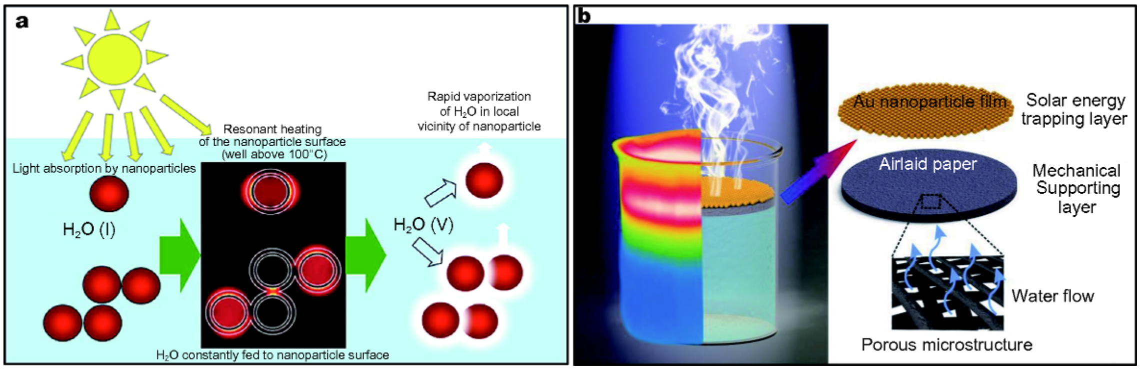


Figure 1 (a) Schematic of nanoparticle-enabled solar steam generation. Reprinted with permission from Ref. [6], Copyright 2013, the American Chemical Society. (b) Schematic illustration of the structure of airlaid-paper-based AuNP film for high efficient photothermal conversion. Reprinted with permission from Ref. [15], Copyright 2015, Wiley-VCH.

The development of sunlight absorbers has become a popular research topic because of their remarkable potential to improve photothermal conversion efficiency. Metallic plasmonic materials [17], ceramic plasmonic materials [18], and carbon-based materials have been demonstrated for efficient solar absorption. Compared to metallic plasmonic materials, carbon-based materials (carbon black, graphite, graphene, carbon nanotubes, carbon composites, amorphous carbons, etc. [19]) have been chosen as superior photothermal conversion materials because of their inherent outstanding properties, which can be widely used in daily life. Excellent photothermal effect, high absorbance, and low-emittance properties are key factors for carbon-based materials that serve as vapor generators. Additionally, carbon-based materials are widely available and very cheap. Furthermore, as mentioned above, light-absorbing materials floating on the surface of the water can efficiently reduce the heat loss. Both their tunable densities and the hydrophobic surfaces are essential for their floatability on water [20]. Herein, we briefly introduce the current development of carbon nanocomposites in the field of photothermal conversion.

CARBON NANOCOMPOSITES

Amorphous carbon can be prepared easily with tunable textural properties and flexible compositions. Cokes from zeolite-catalyzed reactions can be easily converted to porous carbons and deposited on a cellulose membrane for steam generation [21]. In addition, natural wood [22,23] and mushrooms [24] also can be utilized as ideal solar absorbers after a simple carbonization treatment, as shown in Fig. 2.

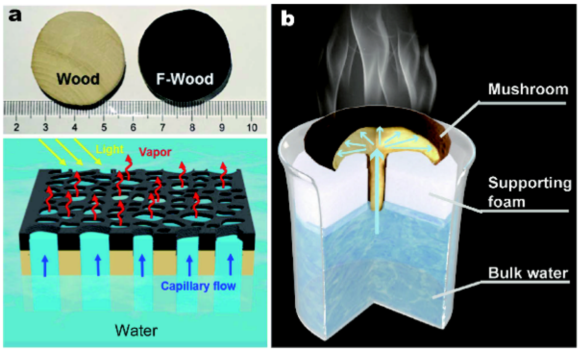


Figure 2 (a) Photographs of blocks of pristine wood and flame-treated wood (F-wood) and schematic illustration of F-wood for solar steam generation. Reprinted with permission from Ref. [23], Copyright 2017, the American Chemical Society. (b) Mushroom-based solar steam generation. Reprinted with permission from Ref. [24], Copyright 2017, Wiley-VCH.

Owing to the D-band's optical transitions, both conventional graphite and carbon black are natural light absorbers over a broad bandwidth. Ghasemi and coworkers [25] reported a double-layer structure consisting of exfoliated graphite used as a top photothermal layer and carbon foam used as a heat barrier (Fig. 3a). Under solar irradiation, a hot spot was formed on the top of the typical double-layer structure because of the inherent absorption capacity of graphite. Carbon foam can transport water efficiently to the hot spot by capillary action and can be used as a heat barrier in order to minimize heat loss. In order to generate steam at both ambient and high pressures (i.e., in a temperature range of 100–156°C and at pressures of 100–525 kPa), Sajadi *et al.* [26] developed an artificially networked structure, which consisted of a porous polymer skeleton coated with exfoliated graphite and artificially networked 3D veins.

An efficient strategy was reported for developing a

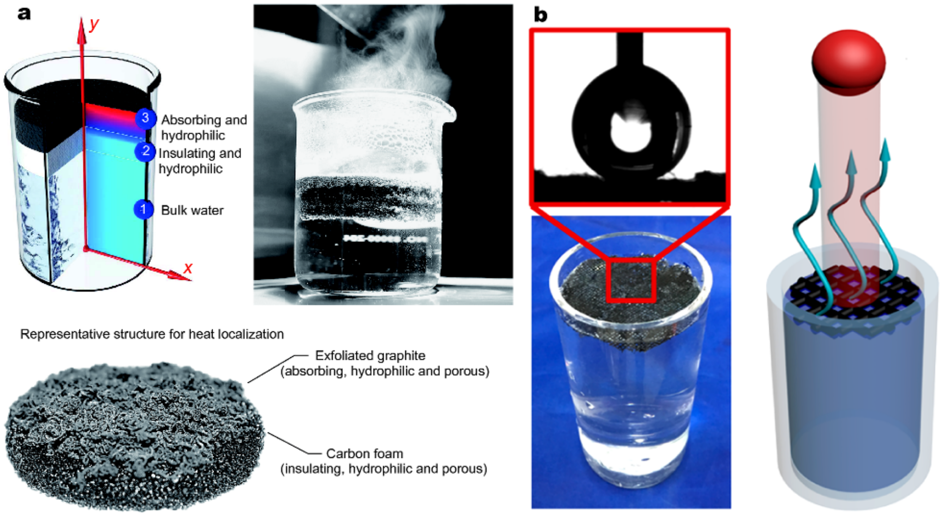


Figure 3 (a) The dual-layer structure consists of a carbon foam for high efficient photothermal conversion. Reprinted with permission from Ref. [25], Copyright 2014, Nature Publishing Group. (b) Schematic diagram of the surface evaporation process of carbon black-based superhydrophobic gauze. Reprinted with permission from Ref. [27], Copyright 2015, the American Chemical Society.

floating black gauze using extremely low-cost materials (carbon black nanoparticles and degreasing cotton gauze) as shown in Fig. 3b. Compared with the traditional process, the evaporation rate of the carbon-black-based superhydrophobic gauze with self-cleaning ability was increased two fold. In addition, the gauze can facilitate recycling and reuse for practical applications [27]. Inspired by the capability of hydrophilic porous paper to transmit water, Gan's group [28] reported the use of carbon-coated paper as a solar vapor generation system. The proposed carbon-coated paper system with a thermal insulation layer (expanded polystyrene foam) realized photothermal conversion efficiency greater than 88% under one-sun equivalent illumination, and the corresponding evaporation rate was 1.28 kg m^{-2} h^{-1}.

However, both carbon black and graphite are limited to moderate reflection values of 5%–10% at the air–dielectric interface. In order to overcome this limit, an efficient approach is to use nanostructures, such as vertically aligned carbon nanotubes (CNTs) or porous graphene [29,30]. CNTs and graphene as outstanding photothermal materials have attracted tremendous attention in extensive studies such as photothermal therapy [31,32], environmental remediation [33], and solar vapor generation [34–36] for water purification/desalination.

CARBON-NANOTUBE-BASED SHEETS

Besides the extremely high broadband light absorption of CNTs over the entire solar spectrum, the main reason why CNTs have been gaining substantial research attention for solar vapor generation is that their attractive properties include physical, chemical, and thermal stability. More importantly, CNT-based thin and porous films or membranes with controlled thickness and pore sizes can be obtained by a series of simple methods such as filtration, spray coating, spin coating, etc.

CNT nanofluids have been investigated for low-temperature solar vapor generation [37]. The solar vapor generation mechanism of CNT nanofluid is shown in Fig. 4a. CNTs absorb and scatter photons in the upper fluid first, and then a fraction of photons are incident on the lower fluid under sun illumination. The well-known disadvantage of CNT fluids is the recovery and reuse of their nanoparticles. Fe_3O_4@CNT nanoparticles (Fig. 4b) based on CNTs decorated with magnetic Fe_3O_4 were introduced for solar vapor generation [38]. Both the separation rate and the recovery rate of Fe_3O_4@CNT from water were faster than those of CNTs according to the magnetic field strength. Moreover, the introduction of the Fe_3O_4 nanoparticles enhanced the photothermal conversion efficiency of CNT fluids owing to the high absorption of Fe_3O_4 nanoparticles. Thus, this work presented an approach to not only significantly reduce material consumption in the design of solar evaporators, but also to realize an outstanding improvement in photothermal conversion efficiency.

However, a significant portion of the absorbed energy would dissipate into the bulk water in nanofluid solar vapor generation systems, which restricts the improvement of photothermal conversion efficiency. In order to

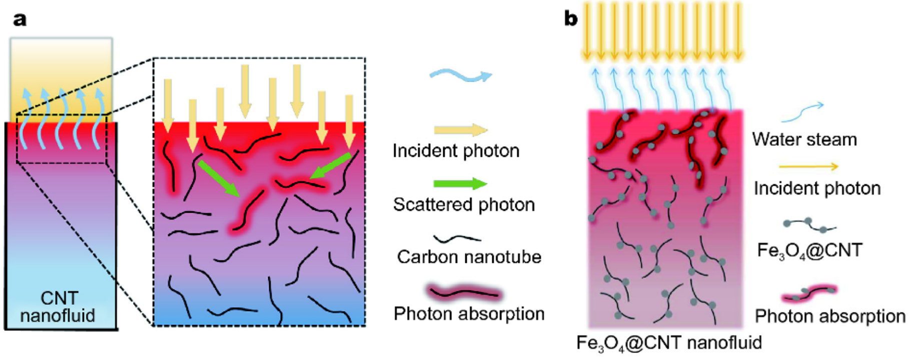
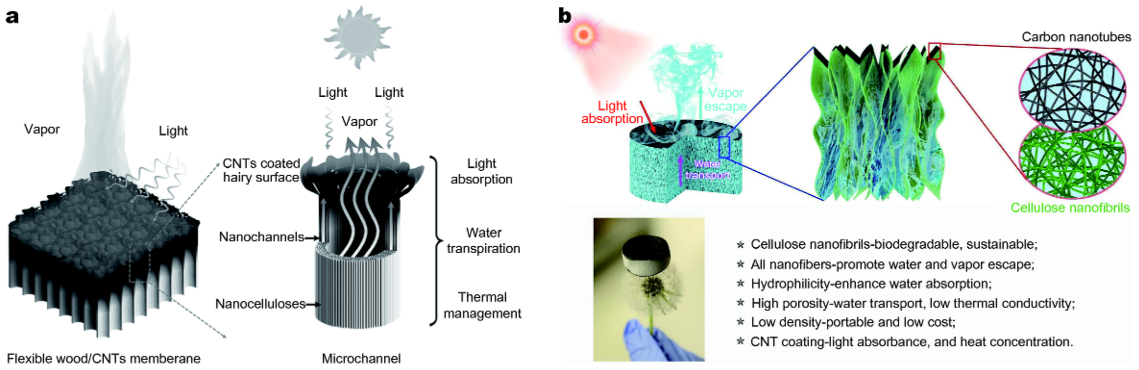


Figure 4 (a) Scheme of localized solar heating and steam-generation mechanism based on CNT. Reprinted with permission from Ref. [37], Copyright 2016, Elsevier. (b) Schematic of the experimental setup for solar steam generation based on Fe_3O_4@CNT. Reprinted with permission from Ref. [38], Copyright 2017, Elsevier.



⋆ Cellulose nanofibrils-biodegradable, sustainable;
⋆ All nanofibers-promote water and vapor escape;
⋆ Hydrophilicity-enhance water absorption;
⋆ High porosity-water transport, low thermal conductivity;
⋆ Low density-portable and low cost;
⋆ CNT coating-light absorbance, and heat concentration.

Figure 5 (a) Graphical illustration of the flexible solar steam made from CNT-coated flexible wood membrane. The flexible wood/CNTs membrane possesses three merits as solar steam—the black hair-like surface coated with CNTs is highly absorbing to incident sunlight, the microchannels and nanochannels can pump water up, while the wood itself has a low thermal conductivity. Reprinted with permission from Ref. [42], Copyright 2017, Wiley-VCH. (b) Graphical illustration of all-nanofiber CNF–CNT aerogel used for solar steam generation. The solar steam generation device is composed of a bilayer structure of a bulk CNF aerogel with a thin CNT coating layer. A photograph of bilayer CNF–CNT aerogel standing on the top of a dandelion demonstrates its lightweight performance. Reprinted with permission from Ref. [43], Copyright 2017, the American Chemical Society.

improve the photothermal conversion efficiency further, reduce costs, and enhance scale up, a series of CNT-based membranes were reported for solar vapor generation [35,39], including ultrathin flexible single-walled nanotube (SWNT)-MoS_2 hybrid film [40] and a vertically aligned CNT [41]. CNT-based membranes floated in the water-air interface can harvest light energy and transfer it almost entirely into the surrounding water, resulting in high photothermal efficiency.

Natural wood possesses a series of inherent properties, such as high porosity, light weight, low thermal conductivity, and hydrophilicity. These make it an excellent material for solar vapor generation. Hu and Jia [42] first proposed CNT-modified flexible wood membranes as solar vapor generation devices. In Fig. 5a, CNTs coating on the top of the flexible wood/CNTs were utilized to concentrate solar energy. The natural interconnected channels in the wood structure can strengthen water transport. Meanwhile, the low thermal conductivity of wood can minimize thermal loss in the vapor generation process. In addition, inspired by the water transpiration behavior of trees, Hu's group [43] also designed a bilayer-structured aerogel (Fig. 5b) composed of seamlessly integrated cellulose nanofibrils (CNF) and CNTs to form a compressible and efficient solar vapor generator. It was found that a piece of aerogel can stand on the top of a dandelion without bending or deforming the seed hair, providing portability for a solar vapor generator. Like in wood, macro-channel structures in CNF aerogel can contribute to water absorption and transport and guarantee minimum heat loss to the bulk water body or surrounding environment. A CNT coating on the top of the bilayer-structured aerogel was utilized to concentrate solar energy. Photothermal conversion efficiency can reach 76.3% under one-sun illumination.

GRAPHENE-BASED SHEETS

Graphene with light weight has been widely investigated in the field of solar vapor generation owing to their remarkable properties, including large surface area and outstanding photo-harvesting ability [43]. However, the natural properties of graphene (two-dimensional (2D) flat and hydrophobic features) are the main limiting factors in the development of graphene in solar vapor generation. Functionalized graphene by nitrogen doping [44] and hydrophilic treatment [45] has demonstrated remarkable enhancement of photothermal conversion efficiency.

Graphene oxide (GO), with its good dispersibility, is one of the most important derivatives of graphene, presenting a promising opportunity to precisely functionalize graphene. In our previous work, we first prepared GO-Au nanofluids with high efficiency for solar vapor generation under natural sunlight irradiation [46]. Furthermore, Wu *et al.* [47] also demonstrated that reduced GO (rGO)@Fe_3O_4 nanoparticles with recyclability displayed a high solar thermal efficiency (75%) under one-sun illumination.

However, using nanofluids as photothermal devices can result in secondary pollution. Nanofluids in direct contact with bulk water also cause a large amount of heat loss, which can decrease photothermal conversion efficiency. These issues severely restrict the development of the solar evaporation technology. In order to ensure high photothermal conversion efficiency, various rGO composite films for solar vapor generation have been reported [48–51]. Composite films were rationally designed to be po-

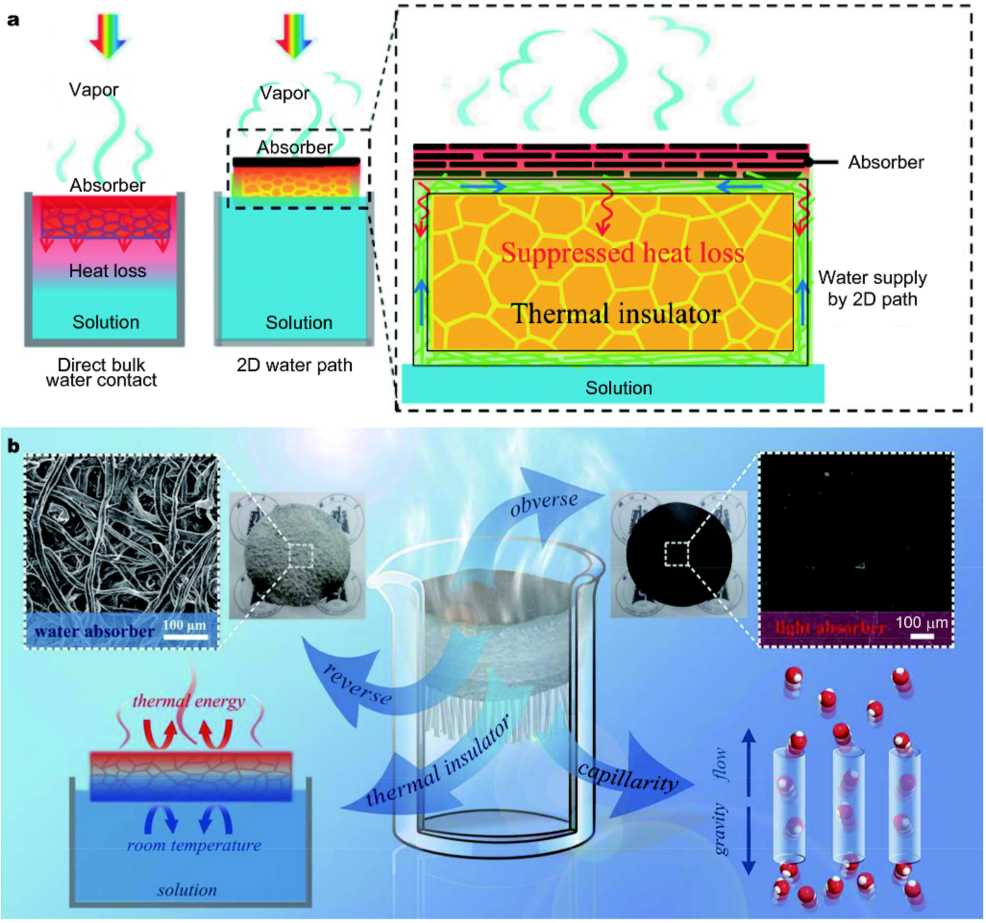


Figure 6 (a) Solar desalination devices with direct water contact (left) and with suppressed heat loss and 2D water supply (right). Reprinted with permission from Ref. [52], Copyright 2016, the National Academy of Sciences of USA. (b) Schematic diagram of the solar vapor generating device with three components: (i) a double-sided rGO film, (ii) a thermal insulator, (iii) the water supply pipelines. Reprinted with permission from Ref. [53], Copyright 2017, the American Chemical Society.

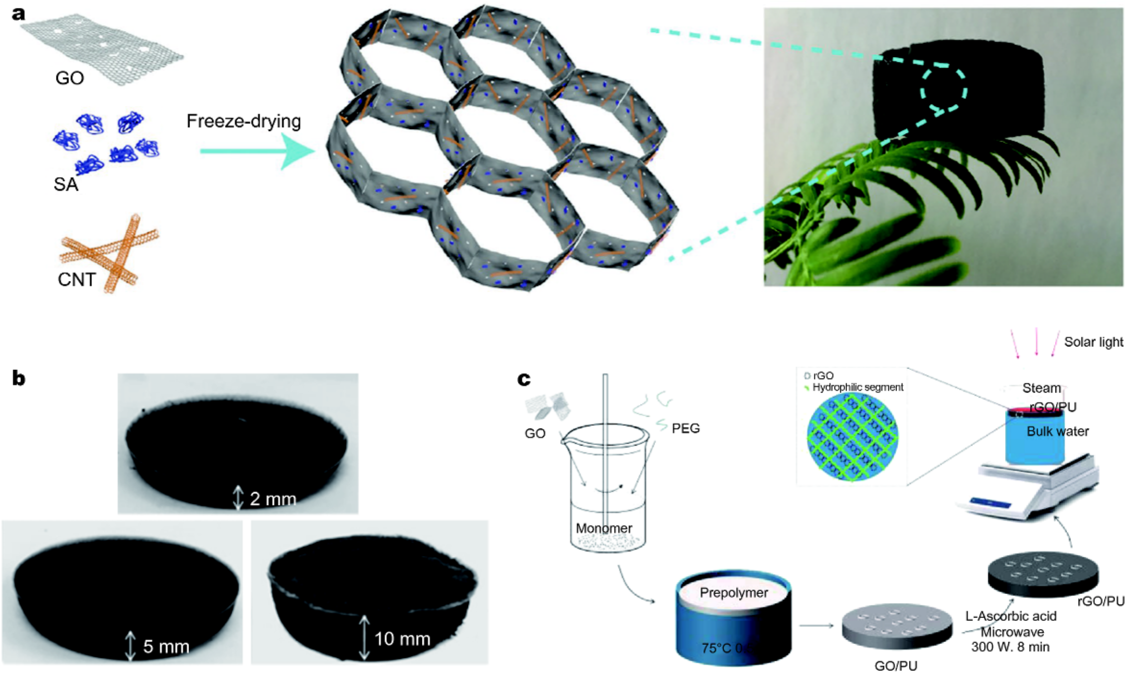


Figure 7 (a) The fabrication process of GO-based aerogels. GO, SA, and CNT were first mixed in water and followed by a freeze-drying process, and then the GO-SA-CNT aerogels were obtained. Reprinted with permission from Ref. [36], Copyright 2017, Wiley-VCH. (b) Optical photos of GO aerogels with different thicknesses. Reprinted with permission from Ref. [56], Copyright 2017, the American Chemical Society. (c) Schematic illustration showing the fabrication of rGO/PU and setup for solar steam generation using the rGO/PU. Reprinted with permission from Ref. [58], Copyright 2017, the American Chemical Society.

sitioned on the top of porous materials with low thermal conductivity to simultaneously attain limited heat loss and efficient water transport.

Typically, a GO film deposited onto a porous mixed cellulose membrane is separated by a thermal insulator (a polystyrene foam with a thermal conductivity of about 0.04 W mK^{-1}), and not in direct contact with bulk water (Fig. 6a) [52]. In addition, cellulose wrapped over the foam functions to create 2D water transport paths *via* capillary force. Another compact device using rGO and paper fibers was designed by our group [53]. Our solar vapor generator (Fig. 6b) consists of water supply pipelines (numerous capillary tubes), a thermal insulator (expanded polyethylene foam), and a double-sided absorbing film combining rGO and fiber. Compared with “directly floating on bulk water,” the heat dissipation from the absorber to bulk water was minimized because of the separation between rGO-based film and bulk water.

More strikingly, novel bilayered structures based on graphene were introduced for fast-response solar vapor generation systems through heat localization at the evaporation surface, such as bilayered biofoam composed of bacterial nanocellulose and rGO [34], wood–GO composite [54], and rGO film on polystyrene foam [20]. Unlike conventional structures, the lower layer of these devices serves two functions. One aims to reduce heat loss by acting as insulation. The other is that porous structures within those devices act as water transport channels to move water from bulk water to the light absorption layer at the top of the devices.

Various developed porous graphene aerogels (Fig. 7a, b) [36,53–57] have also been applied for vapor generation. These as-prepared graphene aerogels have micrometer-sized pores with excellent broadband absorption, low heat capacity, and low heat conductivity. The most important function of micrometer-sized pores within graphene aerogels is that micrometer-sized pores can serve as water transport channels, aligning with the necessary conditions for high-efficiency photothermal conversion. A novel strategy was introduced by our group to prepare rGO/polyurethane (PU) foam (Fig. 7c) [58] for high-efficiency photothermal conversion. This foam consists of rGO and a PU matrix. The photothermal efficiency of rGO/PU foam can be up to about 81% at a light density of 10 kW m^{-2}. Owing to the covalent connection between rGO nanosheets and the PU matrix, the photothermal device provides excellent stability and broad optical absorption. Moreover, the interconnected

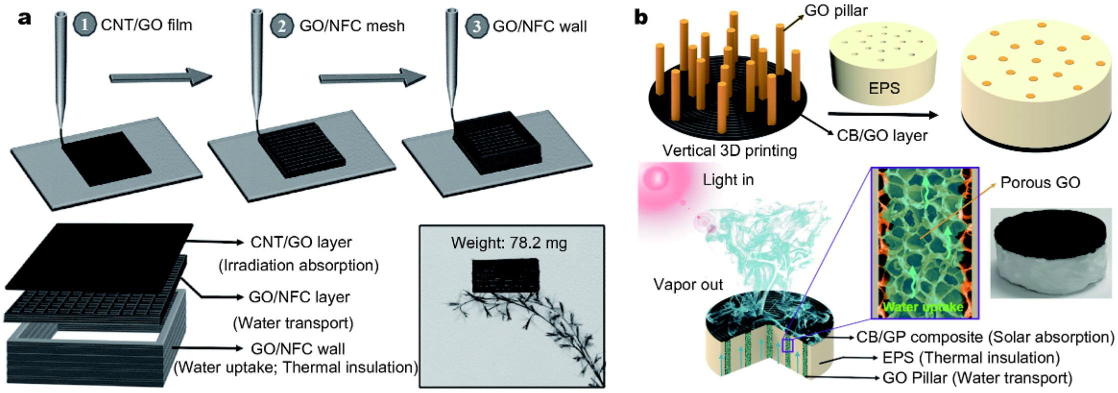


Figure 8 (a) Schematic illustration of 3D printing process of evaporator and sample structure. Reprinted with permission from Ref. [60], Copyright 2017, Wiley-VCH. (b) Schematic showing the structure and principle of a jellyfish-like solar steam generation of the evaporator. Reprinted with permission from Ref. [61], Copyright 2017, Elsevier.

pores of rGO/PU can serve not only as water channels for transporting water to the surface of the rGO/PU foam, but also as thermal insulation resulting in a rapid increase of local thermal energy under illumination.

3D printing, which can be used to fabricate arbitrary structures, has attracted extensive attention [59]. Recently, Li *et al.* [60] utilized 3D printing technology to fabricate a vapor generator with a concave structure (Fig. 8a) which consists of a CNT/GO layer, a GO/nano-fibrillated cellulose (NFC) layer, and a GO/NFC wall. The 3D-printed porous structure not only effectively transports water to the CNT/GO layer, but also prevents heat from escaping into the water. As a result, the 3D-printed evaporator has a high photothermal conversion efficiency of 85.6% under one-sun illumination. Furthermore, Hu's group [61] also designed a jellyfish-like solar vapor generator using an advanced 3D printing technique. As shown in Fig. 8b, a jellyfish-like solar vapor generator consists of a porous carbon black/GO (CB/GO) composite layer (absorbing sunlight), aligned GO pillars (transporting water), and an expanded polystyrene (EPS) matrix (thermal insulator). The all-in-one structure design using the 3D printing technique displays an outstanding photothermal conversion efficiency of 87.5% under one-sun illumination (1 kW m^{-2}).

PROSPECTS

Carbon materials possess inherent physical and chemical properties that make them promising solar vapor generator materials for high-performance photothermal conversion, which can be utilized in water purification and/or desalination, etc. In particular, we focused our discussion on two essential kinds of carbon nanocomposites, CNTs and graphene, whose inherent properties make them ideal solar vapor generator materials. Many reports are becoming available regarding the design of novel and rational floating devices for meeting the demands of high-performance photothermal conversion. Despite current achievements and promising prospects, photothermal conversion is still in its early stages. There remain challenges and opportunities for scientists to improve upon the state of the art. In addition to improving the efficiency of photothermal conversion, industrialization applications should be considered. The key considerations for photothermal conversion systems are low cost, reusability, chemical stability, portability, and suitability for mass production. We believe that carbon nanocomposites serving as photothermal conversion devices could be scalable for commercial applications.

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具有高效光热转换性能的碳基纳米复合材料

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摘要 光热转换产生水蒸汽是一种高效的、全新的太阳能利用方式, 在污水处理和海水淡化等领域具有广泛应用前景. 本文介绍了光热转换的发展以及用作太阳能水蒸汽产生系统的吸光材料的种类, 进一步深入分析碳基纳米复合材料(碳纳米管和石墨烯)制备水蒸汽的转换效率, 并结合碳基纳米复合材料在光热转换领域的相关研究成果, 提出了碳基纳米复合材料在光热领域面临的各种挑战和机遇.