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A Facile Approach to Fabricate Sustainable and Large-Scale Photothermal Polydopamine-Coated Cotton Fabrics for Efficient Interfacial Solar Steam Generation

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with oven drying method and in situ reduction of Ag⁺ by dopamine, respectively. Thanks to the synergistic effect of PDA and AgNPs, the P-Ag-P@CFs show remarkable light absorption and solar-tovapor efficiency (over 90% for deionized water under 1-sun irradiation), resulting in a high evaporation rate of 1.378 kg m⁻² h⁻¹. Moreover, the P-Ag-P@CFs display an excellent purification

efficiency and degree for several kinds of aqueous solutions, such as



seawater, simulated sewage, and industrial wastewater. The desalinated water is compliant with the standards of drinking water.

INTRODUCTION

Employing solar energy as the energy source, interfacial solar steam generation (ISSG) developed in recent years is regarded as a green, low-cost, and environment-friendly desalination and wastewater treatment technology to meet the ever-growing need for freshwater at minimal energy consumption.¹⁻⁴ Compared with traditional SSG, the key mechanism of ISSG is to confine the heat, converted from solar energy through the photothermal materials, at the water-air surface to enhance the temperature of surface water and accelerate the water evaporation to generate steam, which is then condensed to form freshwater via water collectors.⁵⁻⁷ Recently, many kinds of photothermal materials with high light absorption performance have been employed as solar evaporators in ISSG, for instance, carbon black (CB),^{8,9} carbon nanotubes (CNT),^{10,11} graphene,^{12,13} graphene oxide (GO),^{14,15} reduced graphene oxide (rGO),^{16,17} polypyrrole (PPy),^{18,19} noble metal nanoparticles,^{20,21} and Ti₃C₂-MXene.^{22,23}

Solar evaporators comprised with only the photothermal materials often suffer from many issues, such as dramatic heat loss due to high thermal conductivity, insufficient water supply because of poor hydrophilicity, and ultrahigh manufacturing cost, which significantly limit the large-scale fabrication of solar evaporators. As an economical and efficient strategy, the photothermal materials can be coated on the surface of porous substrates, such as membranes (e.g., filter paper and nonwoven fabric),^{24,25} foams (e.g., polystyrene and melamine foam),^{26,27}

and bio-based materials (e.g., wood and bamboo),²⁸⁻³¹ through surface coating methods (e.g., vacuum drying and freeze drying).^{1,26} The obtained photothermal material-coated substrates as solar evaporators exhibit improved hydrophilicity and thermal insulation properties. Nevertheless, the adhesion stability between photothermal materials and substrates via physical interactions is generally unsatisfactory, thereby leading to a relatively low long-term stability.

Polydopamine (PDA), as a kind of bio-friendly photothermal polymer, is synthesized via the oxidation selfpolymerization of dopamine (DA) in alkaline aerobic conditions.³²⁻³⁴ PDA can be in situ attached to almost all the substrates via the strong interaction (such as hydrogen bonds) thanks to the abundant catechol and amino groups of PDA.³² The PDA-coated materials as solar evaporators exhibit outstanding structural stability, which promotes theirs applications in ISSG. Mi et al. reported a PDA/CB-coated polypropylene nonwoven fabrics with high solar absorption efficiency and good hydrophilicity, of which the solar-to-vapor efficiency was up to 91.5% under 1-sun (1 kW m^{-2})

Received:	October 4, 2022
Revised:	November 16, 2022
Accepted:	November 16, 2022





Figure 1. Schematic illustration of the fabrication process of the P-Ag-P@CFs.

irradiation.³⁵ Yang et al. designed a wood-based solar evaporator using PDA and Ag nanoparticles (AgNPs) as photothermal materials.³⁶ Wang et al. proposed PDA-functionalized cellulose aerogels using waste cotton fabrics as raw materials.³⁷ The PDA-coated materials as solar evaporators attract much attention and become one of the hottest topics in the field of ISSG.^{38,39} The PDA coatings on substrates are normally formed by immersing substrates in DA solution for several hours. However, it is still a big challenge to fabricate PDA-coated solar evaporators in a large scale by employing the traditional immersion method due to the restriction of the container size and limited dissolved oxygen in the DA solution.³²

Herein, as shown in Figure 1, a facile synthesis method of PDA coating was proposed and employed to fabricate sustainable and large-scale PDA and AgNP-coated cotton fabrics (P-Ag-P@CFs) as solar evaporators for efficient interfacial solar steam generation. Cotton fabrics were chosen as substrates due to their sustainability, biodegradability, easy availability, flexibility (portability), and low cost. PDA coatings on cotton fabrics were formed via a dip-coating method followed by oven drying treatment, which was demonstrated as a facile approach to realize the large-scale preparation of photothermal PDA-coated cotton fabrics (P@CFs). The Ag ions (Ag⁺) were adsorbed on the negatively charged P@CFs via electrostatic interactions due to the phenolic hydroxyl groups of PDA and were then reduced to AgNPs with DA as a reducing agent, leading to the formation of Ag-P@CFs. AgNPs possess an outstanding solar absorption efficiency due to the localized surface plasmon resonance (LSPR) and endow solar evaporators with antibacterial properties. Then, another layer of PDA was coated on Ag-P@CFs to prevent AgNPs from being oxidized and enhance the hydrophilicity of Ag-P@CFs, resulting in the final P-Ag-P@CFs as solar evaporators. On account of the synergistic effect of PDA and AgNPs, the obtained P-Ag-P@CFs exhibit superior light absorption (over 93% in the broadband wavelength of the solar spectrum from 300 to 1400 nm) and solar-thermal efficiency, which lead to a high evaporation rate of 1.378 kg $m^{-2} h^{-1}$ and a solar-to-vapor efficiency of over 90% for deionized water under 1-sun solar irradiation. The P-Ag-P@CFs also show a remarkable purification efficiency and degree for several different types of solutions, such as seawater, simulated sewage, and industrial wastewater. The desalinated water obtained from P-Ag-P@CFs can meet the standards for drinking water. In addition, a stable continuously working efficiency and durability are achieved by P-Ag-P@CFs for seawater due to their impressive hydrophilicity and structural stability.

EXPERIMENTAL SECTION

Materials. The cotton fabrics were purchased from Tmall, China. Dopamine hydrochloride (DA), Tris(hydroxymethyl) amino methane (Tris), methyl orange (MO), methylene blue (MB), and Rhodamine B (RB) were all bought from MACKLIN Reagent Co., Ltd. Silver nitrate (AgNO₃, Chron Chemicals, China), potassium dichromate (K_2CrO_7 , KE SHI, China), copper chloride ($CuCl_2$, 3AChem, China), ferric chloride (FeCl₃, 97%, Merck, China), span 80 (Coolaber, China), and milk (YiLi, China) were used as received. MO, MB, RB, and span 80 were used to prepare simulated sewage (1 mg/mL). CuCl₂, FeCl₃, and K_2CrO_7 were employed to prepare simulated industrial wastewater (10 mg/mL). Commercially purchased milk was used as a sample of emulsion. Seawater was collected from the South China Sea (Zhuhai, Guangdong province). The chemical reagents were used without further treatment unless mentioned.

Preparation of Photothermal Cotton Fabrics. *Preparation of P@CFs.* P@CFs were fabricated via a dip-coating method combined with oven drying treatment. Briefly, Tris buffer was used to regulate the pH value of DA solution (5 mg/mL) to about 8.5. The pristine cotton fabrics (with a length over 2.3 m) were rinsed by ethanol and deionized water for 20 min in an ultrasonic water bath, folded, and then immersed into the DA solution for 2 min. The fully wetted cotton fabrics were then unfolded and dried in an oven at 80 °C for 1 h. The above steps were repeated five times to coat enough PDA on the surface of cotton fabrics. In the end, the P@CFs were washed with deionized water to remove undeposited PDA and then dried.

Preparation of Ag-P@CFs. The Ag-P@CFs were obtained by the in situ reduction of Ag^+ on the surface of P@CFs with DA as a reducing agent. P@CFs with a size of 10 × 10 cm were immersed into the AgNO₃ solution (5 mg/mL), and DA solution (5 mg/mL) with a volume ratio of 1 to 50 compared with the AgNO₃ solution was slowly added to accelerate the reduction of Ag^+ . After stirring for 4 h, the cotton fabrics were washed with deionized water and dried to obtain dark-brown Ag-P@CFs. In addition, another Ag-P@CFs sample was fabricated via the similar method without the extra added DA as a reducing agent for comparison.

Preparation of P-Ag-P@CFs. The P-Ag-P@CFs were fabricated by coating another PDA layer on the surface of Ag-P@CFs with the similar procedure to the fabrication of P@ CFs. In general, the Ag-P@CFs were immersed in DA solution for several minutes and then dried in an oven at 80 °C for 1 h. After five times of dip-coating and oven drying, another PDA layer was coated on Ag-P@CFs. In the end, P-Ag-P@CFs were washed with deionized water and dried.

Characterization. The morphologies and elemental distribution of cotton fabrics, P@CFs, Ag-P@CFs, and P-Ag-P@CFs were investigated by scanning electron microscopy (SEM) with energy-dispersive spectroscopy (EDS) (ZEISS Gemini 500, Germany) at an acceleration voltage of 20 kV, and a 5–10 nm layer of gold was sputtered on the samples via a vacuum ion sputtering coater before observation. Powder X-ray diffraction (XRD, BRUKER D8 ADVANCE, Germany) measurements were conducted to assess the crystallinity of the AgNPs in the 5–90° scan range. Raman spectra were performed by a Raman spectrometer (Renishaw inVia microscope, Britain). The X-ray photoelectron spectroscopy



Figure 2. (a) PDA-coated nonwoven fabrics with a size of 40 cm \times 20 cm. (b) P@CFs with a size of 2.3 m \times 1.0 m.

(XPS) data were recorded with a photoelectron spectrometer (AXIS SUPRA, Japan), and the C 1s hydrocarbon peak at 284.8 eV was used to calibrate the binding energies. The contact angles were recorded by an automatic video microcontact angle measuring instrument (OCA15 Pro, Germany). Reflectance (R) and transmittance (T) spectra were measured in the range of 300–1400 nm using a UV–vis absorption spectrophotometer (UV-2600, Japan) equipped with an integrating sphere. The absorbance was calculated by A = 1– R - T. Concentrations of metal ions in different water samples were determined by inductively coupled plasmaoptical emission spectroscopy (ICP-OES, SPECTRO ARCOS MV, Germany).

Steam Generation Experiments. Steam generation experiments were conducted at an ambient temperature of about 15 °C and an ambient humidity of about 70%. A solar simulator (xenon light source, PLS-SXE300, Beijing Perfectlight Technology Co., Ltd.) equipped with an AM 1.5 spectral filter was used to simulate solar irradiation on the tested samples. Solar steam generation experiments were carried out at an irradiation of 1-sun intensity, which was controlled by an optical power meter (PL-MW2000, Beijing Perfectlight Technology Co., Ltd.). The tested samples were cut to a size of 2 cm \times 2 cm and placed on a designed ISSG system with a 2D water pathway. The polystyrene (PS) foam $(2 \text{ cm} \times 2 \text{ cm})$ was wrapped with a piece of nonwoven fabric. The ISSG system was set in water-filled beakers. The real-time mass change of different solutions was measured by a precision balance connected to a computer with a precision of 0.001 g. Furthermore, the variation of the surface temperatures of the samples was recorded by using an infrared radiation (IR) camera (PTi120, Fluke). A lab-made water collector was used to collect the condensation to harvest purified water.

RESULTS AND DISCUSSION

Preparation of Large-Scale Photothermal Cotton Fabrics. The classic DA solution immersion treatment method was first employed to fabricate PDA-coated nonwoven fabrics ($20 \text{ cm} \times 40 \text{ cm}$) as solar evaporators. The nonwoven fabrics made of 55% cellulose and 45% polyester were folded and then immersed in DA solution (the pH was tuned to 8.5 by Tris). After stirring for 24 h, the PDA-coated nonwoven fabrics were obtained and are shown in Figure 2a. Only the edges of nonwoven fabrics were coated with an uneven black PDA layer, and most parts of the nonwoven fabrics, especially in the center area, were almost white due to the limited dissolved oxygen in the DA aqueous solution. Therefore, the conventional DA solution immersion treatment method is not feasible to fabricate PDA-coated substrates in a large scale. Moreover, the nonwoven fabrics suffered severe deformation after stirring for several hours, owing to their poor mechanical properties.

In this study, the sustainable, green, tough, and cheap cotton fabrics were chosen as a substrate, and the dip-coating method was employed to fabricate large-scale PDA-coated cotton fabrics as solar evaporators. As schematically shown in Figure 1, P-Ag-P@CFs were fabricated via a three-step method. The first step involved the dip-coating method followed by oven drying treatment to achieve the large-scale preparation of P@ CFs. In particular, the cotton fabrics were folded and immersed in DA solution for 2 min to be fully wetted. Moreover, the wetted cotton fabrics were unfolded and dried in an oven at 80 °C, which ensure sufficient exposure of cotton fabrics to air and accelerate the oxidation self-polymerization of DA. On account of the hydrogen bond interaction between the -OH groups of cellulose in cottons and the catechol and amino groups of DA, PDA was in situ formed and coated tightly on the surface of cotton fabrics.^{36,40} The above dip-coating and oven drying cycles were repeated five times to deposit sufficient PDA on cotton fabrics, leading to brown P@CFs in a large scale $(2.3 \text{ m} \times 1.0 \text{ m}, \text{ as shown in Figure 2b})$. Notably, the brown P@CFs became black after wetting by water. In the second step, the P@CFs were immersed in the AgNO₃ and DA aqueous solution. The Ag⁺ were absorbed on the surface of P@CFs due to the electrostatic adsorption between Ag⁺ and PDA and were in situ reduced to AgNPs with extra added DA as a reducing agent, resulting in Ag-P@CFs.⁴¹ In the third step, another layer of PDA was coated on Ag-P@ CFs by repeating the dip-coating and oven drying cycles, leading to the final black P-Ag-P@CFs. The outside hydrophilic PDA coating not only boosts the hydrophilicity of cotton fabrics but also prevents AgNPs from being easily oxidized by air. P-Ag-P@CFs thus exhibit excellent stability. Furthermore, the characteristic of facile fabrication in a large area guarantees the promising potential of P-Ag-P@CFs for practical application.

The morphologies and compositions of pristine and modified cotton fabrics were first investigated by SEM and EDS. Figure 3 shows the photographs and SEM images with different magnifications of pristine cotton fabrics (Figure 3a–



Figure 3. Photographs and SEM images with different magnifications of cotton fabrics (a-d), P@C(e-h), Ag-P@C(i-l), and P-Ag-P@C(m-p).

d), P@CFs (Figure 3e-h), Ag-P@CFs (Figure 3i-l), and P-Ag-P@CFs (Figure 3m-p). The pristine cotton fabrics are white, and their striped-shape fibers display a smooth surface. After the coating of PDA, PDA nanoparticles were deposited on the surface of cotton fabrics, leading to enhanced roughness of P@CFs. Such a rough surface of P@CFs is extremely helpful to enhance light absorption performance through the multiscattering and multireflection of incoming sunlight.^{42,43} Moreover, P@CFs turned to brown after the coating of PDA. After the in situ deposition of AgNPs, the Ag-P@CFs turned to dark brown, resulting from the enhanced light absorption due to the LSPR of AgNPs. After the coating of another layer of PDA, the P-Ag-P@CFs turned to dark. According to the SEM images, Ag-P@CFs and P-Ag-P@CFs display similar surface morphologies. The EDS elemental mapping images of cotton fabrics, P@CFs, Ag-P@CFs, and P-Ag-P@CFs are shown in Figure S1. In cotton fabrics, C and O elements were detected. After the coating of a PDA layer, P @CFs display additional N elements, suggesting the successful coating of PDA. Moreover, Ag elements were uniformly found in the EDS of Ag-P@CFs, indicating the deposition of AgNPs on the surface of P@CFs after the in situ reduction of Ag⁺ with DA as a reducing agent. The EDS mapping images confirmed the successful preparation of P-Ag-P@CFs.

XPS spectra were further employed to analyze the surface chemical composition of pristine and modified cotton fabrics. As shown in Figure 4a, C 1s and O 1s (Figure S2a) features are detected in pristine cotton fabrics, and the additional N 1s feature is found in P@CFs. The high-resolution N 1s spectra of P@CFs show a new fitting peak near 402.0 eV, which is attributed to the R–NH₂ bond of PDA (Figure 4b).⁴⁴ The analysis of the high-resolution C 1s spectra also reveals a novel C==N fitting peak at 288.4 eV in P@CFs (Figure S2b,c). The N-containing peaks in P@CFs prove the coating of PDA. Furthermore, the Ag 3d and Ag 3p features are present in Ag-P@CFs. As shown in Figure 4d, the Ag $3d_{5/2}$ and Ag $3d_{3/2}$ features at about 368.5 and 374.4 eV are both detected in Ag-P@CFs, indicating the successful in situ growth of AgNPs.⁴⁵ In



Figure 4. (a) XPS spectra of pristine and modified cotton fabrics. XPS analysis of (b) N 1s spectra of P@CFs and (c) Ag-P@CFs and P-Ag-P@ CFs. (d) XPS analysis of Ag 3d of Ag-P@CFs and P-Ag-P@CFs. (e) XRD patterns and (f) Raman spectra of pristine and modified cotton fabrics.



Figure 5. Water transportation testing of pristine and modified cotton fabrics.

particular, Ag-P@CFs exhibit two unique peaks at 402.9 and 405.8 eV in N 1s spectra, which probably are produced due to the spontaneous reduction and chelation of the AgNPs on the surface of PDA molecules and the residual nitrate $(-NO_3)$ of AgNO₃, respectively.³⁶ These two peaks, however, disappear in the N 1s spectrum of P-Ag-P@CFs, and meanwhile, the Ag 3p spectrum is not found in P-Ag-P@CFs, suggesting that AgNPs are well-protected by the external PDA coating layer.

The XRD patterns and Raman spectra were used to further verify the existence and well protection of AgNPs. As shown in Figure 4e, the pristine cotton fabrics and P@CFs exhibit two overlapping peaks at $2\theta = 14.0-17.8^{\circ}$ and an independent peak at $2\theta = 22.5^{\circ}$, which correspond to the (101) and (101) crystal planes of cellulose I and the (002) crystal plane of the cellulose I crystalline structure, respectively.^{24,46} Furthermore, the Ag-P@CFs and P-Ag-P@CFs display typical crystal peaks of Ag at 38.12, 44.32, 64.44, 77.42, and 81.54° (Figure 4e), which belong to the (111), (200), (220), (311), and (222) crystal faces of Ag, respectively.^{47,48}Figure 4f shows the Raman spectra. The pristine cotton fabrics cannot be detected with Raman signals due to the fact that chemical functional groups of cellulose are too weak. P@CFs exhibit certain absorption at 1413 and 1625 cm⁻¹, which refer to the D and G peaks generated by the stretching and deformation vibrations of the catechol group in PDA molecules, respectively.⁴⁹ After modification with AgNPs, the centers of D and G peaks in Ag-P@CFs are blueshifted to 1394 and 1595 cm⁻¹, and the intensities of both peaks are significantly enhanced. These are because of the fact that AgNPs exhibit a very strong surface enhancement of the Raman scattering signal for PDA molecules, suggesting the strong interaction between AgNPs and the phenolic hydroxyl groups of PDA.50 For carbon materials, the intensity ratio of the D peak to the G peak can reflect the lattice defect density of C-atom crystals,⁵¹ which increases from 0.44 of P@CFs to 0.51 of Ag-P@CFs due to the enhanced lattice defects caused by AgNPs. This intensity ratio, however, decreases to 0.05 for P-Ag-P@CFs, ascribed to the dramatically reduced intensity of the D peak. Therefore, the external PDA coating layer greatly increased the graphitization of photothermal cotton fabrics, indicating the excellent protection of AgNPs.⁵²

Water Pumping and Photothermal Conversion. A labmade device was used to test the water transportation rate of photothermal cotton fabrics (Figure 5). The cotton fabrics, P@CFs, Ag-P@CFs, and P-Ag-P@CFs were cut into a length of 5 cm (Figure S3) and suspended above a beaker containing RB solution as the color indicator. Attributed to the great hydrophilic performance, the water in pristine cotton fabrics reached about 2 cm in 90 s and 5 cm in 385 s. After PDA coating, it took 300 s to wet the P@CFs to 5 cm, suggesting a better water transportation performance than pristine CF. The better water transportation of P@CFs is mainly attributed to the in situ formation of PDA nanoparticles on the surface of CFs, and introducing appropriate nanostructures into microstructures has been demonstrated as an effective approach to improving water transport performance.53-55 After the in situ growth of AgNPs, the hydrophilicity of Ag-P@CFs decreased slightly, resulting in a longer time of 400 s to pump the water to 5 cm. After coating another PDA on Ag-P@CFs, the P-Ag-P@CFs showed better hydrophilicity than Ag-P@CFs due to the PDA layer. The water droplet was sucked by Ag-P@CFs in several seconds (Video S1) while was absorbed by P-Ag-P@ CFs immediately (Video S2). For P-Ag-P@CFs, it only takes about 100 s to achieve a water transportation of 5 cm (Figure 5). The remarkable water pumping ability of P-Ag-P@CFs is attributed to the hydrophilicity of the PDA coating and nanostructures of PDA and Ag nanoparticles.

The light absorption efficiency and solar-thermal performance of photothermal cotton fabrics were investigated. As shown in Figure 6a, pristine cotton fabrics have a very limited light absorption efficiency (about 14%, Table 1) in the wavelength range of 300–1400 nm. P@CFs present an enhanced light absorption performance. The absorbance of P@CFs in the UV–vis (300–760 nm) and NIR (760–1400 nm) spectrum is about 91 and 45%, respectively, indicating that PDA has strong absorption in the UV–vis spectrum,



Figure 6. (a) UV-vis-NIR spectra of pristine and modified cotton fabrics. Surface temperature (b) and (c) IR mapping images of cotton fabrics as a function of irradiation time under 1-sun irradiation.

Table 1. The Light Absorption Efficiency, Evaporation Rate, and Corresponding Solar-to-Vapor Efficiency of Photothermal Cotton Fabrics under 1-Sun Irradiation

	light absorption efficiency (%)			
	UV– vis	NIR	water evaporation rate $(kg m^{-2} h^{-1})$	solar-to-vapor efficiency (%)
cotton fabrics	14.1	14.5	0.949	59.3
P@CFs	91.6	44.5	1.251	81.2
Ag-P@CFs	92.2	83.0	1.330	86.8
P-Ag-P@CFs	95.9	91.6	1.378	90.2

which is attributed to the unique $\pi - \pi^*$ transition of PDA.⁵⁶ The absorbance of Ag-P@CFs in the UV-vis and NIR spectrum rises to 93 and 83%, respectively. This is due to the resonant coupling effect of the metal dipole with incident light caused by AgNPs.⁵⁷ Moreover, the absorbance for the NIR spectrum of the Ag-P@CFs sample synthesized without using the extra DA as a reducing assistant is reduced to about 72% (Figure S4). Therefore, extra DA is required for the reduction of Ag. The UV-vis and NIR absorption of P-Ag-P@CFs further increased to 96 and 87%, respectively, with an average absorption efficiency of over 90%. In particular, the wetted P-

Ag-P@CFs exhibit better absorption in the NIR range (about 92%), leading to a superior overall solar absorption efficiency of up to 93%.58 The excellent solar absorption performance gives a promising competitiveness in solar-to-thermal tests for photothermal cotton fabrics. After exposure to simulated sunlight (1-sun intensity) for 300 s, the surface temperature of wetted P@CFs rose to 39.6 °C owing to the strong light absorption and photothermal conversion effect of PDA molecules, much higher than the 23.3 °C of pristine cotton fabrics (Figure 6b,c). Furthermore, Ag-P@CFs and P-Ag-P@ CFs showed a faster solar-thermal rate from 15 to 39.8 and 41.1 °C, respectively. The synergistic photothermal effect of PDA molecules and AgNPs resulted in the shortened thermal response time. On the one hand, the huge amount of heat generated by the LSPR effect of AgNPs is going to be rapidly channeled to the PDA coating. On the other hand, the rate of the electron transition in PDA molecules is accelerated by AgNPs.³⁶ In addition, the synergistic effect also brought more generated heat, maximizing the temperature of P-Ag-P@CFs up to 41.8 °C. Notably, the temperature of bulk water in beakers only reached 16.9 from 15.0 °C after exposure to 1-sun irradiation of P-Ag-P@CFs for 30 min (Figure S5), indicating the outstanding heat-insulating property of the ISSG system.



Figure 7. (a) Schematic illustration of the ISSG system. (b) Mass change of water under 1-sun irradiation and corresponding (c) water evaporation rate and solar-to-vapor efficiency. (d) Enhancement factors of photothermal cotton fabrics compared to the water evaporation of bulk water without an ISSG system.

Solar Steam Generation and Clean Water Collection. The ISSG system consisting of a PS foam wrapped with a piece of nonwoven fabric (Figure 7a and Figure S6) was used to study the evaporation capability of photothermal cotton fabrics. The PS foam was used to provide a sufficient support to float the photothermal layer and reduce the heat loss from the photothermal layer. The nonwoven fabric was employed to facilitate the water transportation from bulk water to the photothermal layer. As shown in Figure 7b, the evaporation rate of deionized water using the solar direct-heating method (bulk water without the ISSG system) is only as low as 0.186 kg $m^{-2} h^{-1}$. However, the ISSG system employing the pristine cotton fabrics as solar evaporators displays a more than five times higher evaporation rate of 0.949 kg m⁻² h⁻¹, indicating the significantly increased efficiency of ISSG compared with bulk water evaporation. After modification with photothermal materials, the water evaporation rates of P@CFs, Ag-P@CFs, and P-Ag-P@CFs further increase to 1.251, 1.330, and 1.378 kg m^{-2} h^{-1} , respectively (Table 1). The solar-to-vapor efficiency is calculated according to the following equation:⁵⁹

$$\eta = \nu (H_{\rm e} + Q) / E_{\rm in} \tag{1}$$

$$H_{\rm e} = 1.91846 \times 10^{6} [T/(T - 33.91)]^{2}$$
⁽²⁾

$$Q = c(T - T_1) \tag{3}$$

where η is the solar-to-vapor efficiency, ν is the evaporation rate (excluding the dark evaporation rate of 0.0885 kg m⁻² h^{-1}), H_e is the vaporization enthalpy of water, Q is the sensible heat of water, E_{in} is the total solar energy input, c is the specific heat capacity of water (about 4.2 J g K^{-1}), T_1 is the initial temperature of water, and T is the equilibrium temperature during the evaporation process. Accordingly, the calculated solar-to-vapor efficiency of bulk water without the ISSG system is 11.7%. The efficiencies of cotton fabrics, P@CFs, Ag-P@ CFs, and P-Ag-P@CFs employing the designed ISSG system are 59.3, 81.2, 86.8, and 90.2%, respectively (Figure 7c), which are 4.09, 5.97, 7.45, and 7.74 times of that of the bulk water without the ISSG system (Figure 7d). The excellent heatinsulating property of the ISSG system and the superior hydrophilicity lead to the impressive evaporation performance of pristine cotton fabrics, and the abundant heat converted from photothermal P@CFs, Ag-P@CFs, and P-Ag-P@CFs accelerates the water evaporation rates. Notably, the efficiency of 90.2% for P-Ag-P@CFs is at a high level compared to the

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Table 2. The Water Evaporation Rate and Corresponding Solar-to-Vapor Efficiency of Ag-P@CFs and P-Ag-P@CFs for	or Saline
with Different Concentrations and Seawater from the South China Sea under 1-Sun Irradiation	

	water evaporation rate (kg $m^{-2} h^{-1}$)		solar-to-vapor efficiency (%)	
	Ag-P@CFs	P-Ag-P@CFs	Ag-P@CFs	P-Ag-P@CFs
deionized water	1.330	1.378	86.8	90.2
1.75 wt % NaCl	1.315	1.372	85.8	89.8
3.50 wt % NaCl	1.248	1.319	81.1	86.0
5.25 wt % NaCl	1.216	1.297	78.8	84.5
seawater from the South China Sea	1.274	1.337	82.9	87.3



Figure 8. (a) Water evaporation rate and solar-to-vapor efficiency of P-Ag-P@CFs for saline with different concentrations and seawater from the South China Sea under 1-sun irradiation. (b) Relative efficiencies of Ag-P@CFs ad P-Ag-P@CFs.



Figure 9. Evaporation rate and solar-to-vapor efficiency of (a) the continuous 8 h test (inset with the images of P-Ag-P@CFs after testing and leaving overnight) and (b) the durability test (10 cycles, once a day, the mass loss curves of the inset) using the collected seawater from the South China Sea as bulk water.

similar type of two-dimensional membrane and can also meet or even exceed many reported three-dimensional materials (Table S1).

The desalination applications of Ag-P@CFs and P-Ag-P@ CFs were further studied. The average mass fraction of the major impurity (NaCl) in seawater is about 3.5%, which is constantly changing along with geographical differences and seasonal ocean currents. Therefore, NaCl solutions with different mass concentrations (1.75, 3.5, and 5.25 wt %) were first used to test the evaporation performance of Ag-P@ CFs and P-Ag-P@CFs. As shown in Figure S7, the evaporation rate of Ag-P@CFs decreases from 1.330 kg m⁻² h⁻¹ for deionized water to 1.315, 1.248, and 1.216 kg m⁻² h⁻¹ for 1.75, 3.5, and 5.25 wt% for NaCl solutions, respectively, corresponding to solar-to-vapor efficiencies of 85.8, 81.1, and 78.8% (Table 2). Similarly, the evaporation rate of P-Ag-P@ CFs decreases from 1.378 to 1.372, 1.319, and 1.297 kg m⁻² h⁻¹, respectively. The corresponding efficiencies are 89.8, 86.4, and 84.5% (Figure 8a). These results reveal that the evaporation performance of the photothermal cotton fabrics



Figure 10. (a) Water mass change of different solutions under 1-sun irradiation and corresponding (b) evaporation rate and solar-to-vapor efficiency. (c) Concentrations of four primary ions $(Na^+, Mg^{2+}, K^+, and Ca^{2+})$ in a standard seawater sample before (original) and after evaporation desalination. (d) Concentrations of Cu^{2+} , Cr^{6+} , and Fe^{3+} before and after evaporation purification. (e) Absorbance of MB, RB, and MO solution before and after evaporation purification.

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decreases with the increase of salt concentration (from 0 to 5.25 wt %). This is because of the precipitated salt crystals on photothermal cotton fabrics. The higher the salt concentration, the more salt crystals will be precipitated during evaporation. Moreover, these salt crystals will block the pores of cotton fabrics, causing a decrease in the evaporation rate. Figure 8b shows the relative efficiency of Ag-P@CFs and P-Ag-P@CFs, defined as the ratio of the solar-to-vapor efficiencies of different concentrations of saline to deionized water. For the 5.25 wt % saline, the efficiency of the Ag-P@CFs sample decreases by a ratio of nearly 10%, while that of the P-Ag-P@CFs sample is only about 6%. This is because of the outstanding hydrophilicity of P-Ag-P@CFs, which results to a high-level efficiency stability for saline. In addition, the evaporation rates of P-Ag-P@CFs for the collected seawater from the South China Sea are about 1.337 kg m^{-2} h^{-1} with a solar-to-vapor efficiency of 87.3%, indicating the excellent solar steam generation ability for practical desalination.

The continuous working efficiency and the cycling rate are also two of the key factors affecting the desalination application. As shown in Figure 9a, the evaporation rate of P-Ag-P@CFs for seawater is 1.333 kg m⁻² h⁻¹ at the beginning and then gradually decreased to 1.230 kg m⁻² h⁻¹ after 8 h, of which the corresponding solar-to-vapor efficiency decreased from 87.1 to 79.8%. The slight efficiency decline is related to the precipitated salt crystals on the edges of P-Ag-P@CFs after the 8 h test. These salt crystals were dissolved and disappeared after leaving the evaporator overnight, which did not affect the solar steam generation on the next day. As shown in Figure S8, 0.5 g of NaCl crystals on the surface of P-Ag-P@CFs was completely dissolved after floating the solar evaporators in seawater for 7 h. The excellent structural stability of P-Ag-P@ CFs benefits the cycling stability of the steam-generation performance. As shown in Figure 9b, P-Ag-P@CFs exhibit

stable evaporation rates for 10 cycles, with an average of 1.334 kg m⁻² h⁻¹, which corresponds to an efficiency of 87.2%. The reliable cycling performance confirms the long-term working potential of P-Ag-P@CFs.

The steam generation performance of P-Ag-P@CFs used for different solutions was further investigated, including simulated sewage (MO, MB, RB, and span 80, 1 mg/mL), simulated industrial wastewater (CuCl₂, FeCl₃, and K₂CrO₇, 10 mg/mL), and emulsion (milk). As shown in Figure 10a,b, P-Ag-P@CFs exhibit similar evaporation rates to deionized water for the simulated sewage and industrial wastewater, ranging from 1.357 to 1.373 kg m⁻² h⁻¹. The corresponding solar-to-vapor efficiencies were higher than 88%. Especially, the evaporation rate of P-Ag-P@CFs for the emulsion is about 1.284 kg m⁻² h^{-1} with an efficiency of 83.6%. The relatively low efficiency is attributed to the large amount of ingredients in milk, such as emulsifiers and proteins. After water evaporation, these ingredients of milk are precipitated and accumulated on the surface of P-Ag-P@CFs, which can block the pores. Moreover, the purification performance of P-Ag-P@CFs on various solutions was evaluated employing a lab-made water collector. The metal ion concentrations of collected water were determined through inductively coupled plasma-optical emission spectroscopy (ICP-OES). As shown in Figure 10c, the concentrations of four primary ions (Na⁺, K⁺, Ca²⁺, and Mg²⁺) in seawater decrease with an ion rejection beyond 99.9% after desalination, and the desalinated water can meet the salinity levels defined by the World Health Organization (WHO) and the standards of the US Environmental Protection Agency (EPA). Figure S9 shows the electrical resistance of different aqueous solutions, which further verifies the purity of the desalinated water obtained by P-Ag-P@CFs. Figure 10d evaluates the quality of simulated industrial wastewater and collected purified water. The relatively high

ion concentrations of 10,000 mg/L (Cu^{2+} , Cr^{6+} , and Fe^{3+}) are drastically dropped to 0.074, 0.008, and 0.007 mg/L, respectively, of which purification efficiencies are close to 100% (Table S2). Figure 10e displays the visible absorbance of MB, RB, MO solution, and purified water, indicating an almost 100% removal of MB, RB, and MO during solar evaporation. In addition, no oil droplets could be observed in the purified water obtained from milk (Figure 10f). These results demonstrate the enormous potential of P-Ag-P@CFs for many kinds of wastewater treatments in the field of ISSG.

CONCLUSIONS

In this study, a facile approach to fabricate photothermal cotton fabrics in a large scale as solar evaporators for efficient ISSG was proposed. The cotton fabrics were chosen as substrates due to their sustainability, easy availability, flexibility (portability), and low cost. A PDA layer was coated on the surface of cotton fabrics via oxidization self-polymerization of DA by dip-coating in DA solution and oven drying cycles, leading to uniform P@CFs in a large area (2.3 m \times 1.0 m). Then, AgNPs were deposited on P@CFs via the reduction of Ag⁺ by DA to enhance the light absorption (especially in the NIR region) due to the LSPR of AgNPs, and another layer of PDA was coated on Ag-P@CFs to prevent the oxidation of AgNPs, further enhance the light absorption, and improve the water transportation efficiency. The as-prepared biodegradable and portable P-Ag-P@CFs exhibit excellent solar-thermal performance, of which the surface temperature reached 41.8 °C under 1-sun irradiation in virtue of the synergistic light absorption (>93%) and photothermal effects of PDA molecules and AgNPs. When combined with the ISSG system with 2D water pathway design, a high evaporation rate of up to 1.378 kg m⁻² h⁻¹ is achieved by P-Ag-P@CFs, corresponding to a solar-to-vapor efficiency of 90.2%. Similar evaporation performance ranging from 87 to 90% was also demonstrated on different aqueous solutions, such as seawater, simulated sewage, and industrial wastewater. In particular, P-Ag-P@CFs perform outstanding continuous working stability and durability during the seawater tests. The average evaporation rate in the 10-day cycle test is up to 1.334 kg m⁻² h⁻¹, with an efficiency of 87.2%. This is due to the remarkable hydrophilicity and excellent structural stability of P-Ag-P@CFs. Notably, the desalinated water obtained from P-Ag-P@CFs can meet the WHO and EPA standards for drinking water. These results indicate the promising practical application value of the large-scale PDA-coated cotton fabrics in the field of ISSG.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.iecr.2c03477.

EDS mapping images, XPS analysis, light absorption comparison, and IR mapping image of photothermal cotton fabrics; schematic diagram and photograph of the ISSG system; water evaporation rate and solar-to-vapor efficiency of Ag-P@CFs for saline; purification performance of P-Ag-P@CFs for different water samples (PDF) (Video S1) Water droplet absorbed by Ag-P@CFs (MP4)

(Video S2) Water droplet absorbed by P-Ag-P@CFs (MP4)

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Notes

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ACKNOWLEDGMENTS

The authors wish to acknowledge funding support from the Science and Technology Program of Guangzhou (no. 2019050001), the Yunnan expert workstation (no. 202005AF150028), the Foundation of Key Laboratory of Pulp and Paper Science & Technology of the Ministry of

Education of China (no. KF202020), the Guangdong Basic and Applied Basic Research Foundation (nos. 1914050005542 and 2022A1515010559), the Natural Science Foundation of Guangdong Province (no. 2021A1515010623), the Guangdong University Research Findings Commercialization Center and Education Bureau of Foshan Innovation Research Program (no. 2020JNHB09), the Guangzhou Basic Research Program (no. 965), the 2021 Innovation Program of Guangdong Province Regular University (no. 2021KTSCX027), the Guangdong Provincial Key Laboratory of Optical Information Materials and Technology (no. 2017B030301007), and ScienceK.

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