Cellulose-Conducting Polymer Aerogels for Efficient Solar Steam Generation

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Seawater desalination and wastewater purification technologies are the main strategies against the global fresh water shortage. Among these technologies, solar-driven evaporation is effective in extracting fresh water by efficiently exploiting solar energy. However, building a sustainable and low-cost solar steam generator with high conversion efficiency is still a challenge. Here, pure organic aerogels comprising a cellulose scaffold decorated with an organic conducting polymer absorbing in the infrared are employed to establish a high performance solar steam generator. The low density of the aerogel ensures minimal material requirements, while simultaneously satisfying efficient water transport. To localize the absorbed solar energy and make the system floatable, a porous floating and thermal-insulating foam is placed between the water and the aerogel. Thanks to the high absorbance of the aerogel and the thermal-localization performance of the foam, the system exhibits a high water evaporation rate of 1.61 kg m⁻² h⁻¹ at 1 kW m⁻² under 1 sun irradiation, which is higher than most reported solar steam generation devices.

1. Introduction

Over 2 billion people live in countries experiencing high water stress,[3] and it is estimated that by 2040, with the existing climate change scenario, one in four of the world’s children under 18 will be living in areas of high water scarcity.[2] In order to alleviate the global water shortage, numerous strategies have been devoted to seawater desalination and polluted water purification. Among these strategies, solar steam generation has attracted a great deal of interest as an example of green and sustainable technology, where the sunlight is the only source of energy.[3] In an effort to increase the solar energy absorption, thus to achieve high evaporation rate and solar-thermal energy conversion efficiency, a large variety of photothermal materials, including plasmonic absorbers,[4,5] metallic nanoparticles,[6] carbon-based,[7,8] and composite materials,[9] have been investigated. Beside these solar absorber materials, a floating structure that supplies liquid water to the heated region and thermal insulators that localize the heat in the evaporative region have also been studied to improve the water evaporation rate.[10,11] In recent years, a high solar-to-thermal conversion efficiency (≥90%) has been achieved using the abovementioned materials and strategies (see Table S1, Supporting Information, for a literature survey of the most recent and efficient solar steam generators).[5,7,12,13] Evaporators based on graphene, graphite flakes, or carbon nanotubes, typically requiring expensive laser reduction or vacuum deposition processes, have reached water evaporation rates between 1.35 and 2.10 kg m⁻² h⁻¹.[8,14] Evaporators based on inorganic absorbers and structures have typically reached lower evaporation rates, unless very thick layers are used.[15] However, beside high conversion efficiency, the costs and sustainability of the raw materials as well as their manufacturing are also important considerations.[16] Overall optimization of the abovementioned features still remains a challenge in this field.

Using low cost water-processable materials is an effective method to decrease the cost of steam generators. Water-based poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) can absorb most of the solar energy in the near infrared (NIR) region,[17] unlike classical visible absorbing organic dyes. This unique property is due to the electronic structure of PEDOT that resembles that of a semimetal.[18] Since almost half of the energy in solar radiation lies in...
the infrared region, the use of PEDOT as organic infrared absorber in steam generator is attractive. While a thin film of PEDOT:PSS can only absorb a small amount of energy in the ultraviolet-visible (UV–vis) region, a simple strategy to improve the UV–vis absorption is making a thick or multilayer structure, in which the light can be absorbed by multiple layers after transmission or reflection. Similar IR-absorbing polymers, such as polypyrrole (PPy), have also been used as polymeric solar absorbers, reaching high water evaporation rates (0.92–3.2 kg m$^{-2}$ h$^{-1}$). However, PPy is not water processable, which limits its applicability.

It is possible to make large specific surface area aerogels by simply freeze-drying a PEDOT:PSS suspension, but such aerogels cannot be used as steam generator because of their poor mechanical properties. Nanofibrillated cellulose (NFC) is a water-based ecofriendly material with high strength and stiffness. Because of the good mechanical properties and biocompatibility, NFC has been used as a scaffold to establish aerogels for multiparameter sensors and also substrates for carbon nanotube based ecofriendly material with high strength and stiffness.

In this work, we report a cellulose based solar vapor generation system which uses a PEDOT:PSS/NFC aerogel as the solar absorber and water evaporator, with an expanded polyethylene (EPE) foam as the thermal insulator. The solar energy at 1 sun intensity can be almost completely absorbed and converted to thermal energy by the aerogel, where the energy is mostly located. The freeze-dried aerogel has microscale channels that allow water to penetrate into the solar absorber, while providing a large specific surface area for water evaporation. The rough surface of the channel walls further increases the internal surface area for evaporation. The rapid water diffusion and capillary pumping of the microscale channels lead to rapid replenishment of the internal surface area to support a sustained high evaporation rate of up to 1.61 kg m$^{-2}$ h$^{-1}$. This is among the largest water evaporation rates reported for fully organic aerogel based evaporators. We finally demonstrate high feasibility of the aerogel system for water desalination by purifying salt water as well as natural sea water to a much lower salinity than the upper limit for good quality drinking water.

### Figure 1

**a)** Schematic diagram of a typical solar steam generation device. **b)** Optical microscope picture of PEDOT:PSS-NFC aerogel. **c)** Scanning electron microscopy (SEM) image of PEDOT:PSS-NFC aerogel. **d)** Chemical structures of PEDOT:PSS, GOPS, and NFC.

### 2. Results and Discussion

In a stable environment, the evaporation rate depends on the following three key factors: i) the solar-to-thermal conversion ability of the absorber, ii) the interfacial surface area, and iii) the heat localization for evaporation. Herein, we employed PEDOT:PSS for photothermal conversion and NFC as an aerogel scaffold for the fabrication of steam generators, owing to the high absorbance of PEDOT:PSS and hydrophilicity of PEDOT:PSS and NFC. To use such an aerogel in water, the water stability is an important consideration. Although PEDOT:PSS is soluble in water, its water stability can be greatly improved by adding the crosslinker (3-glycidyloxypropyl)trimethoxysilane (GOPS) during the aerogel manufacturing. Figure 1a shows a schematic diagram of a typical solar steam generator along with the chemical structures of the components. A 2 mm thick aerogel is located on top of the steam generator to absorb and convert sunlight and provide a large evaporation internal surface. The converted thermal energy should be located in the aerogel but not transported to the bulk water. Thus, EPE foam, which is a closed pore thermal insulator, is placed between the bulk water and the aerogel to localize the thermal energy. A cellulose-based nozzle protection filter is placed through the middle of the foam in order to transport water to the aerogel. A piece of tissue paper, which is used to spread water to the bottom of the aerogel, is located between the insulating foam and the aerogel. Both the water filter and tissue paper are very hydrophilic, so water evaporated from the aerogel can be rapidly replenished. The PEDOT:PSS-NFC aerogels have an open porous structure that forms pathway channels for water transport, as shown in Figure 1b,c. This porous structure is necessary in order to achieve a high evaporation rate since it provides a large interfacial surface area for water molecules to be evaporated from. However, the aerogel absorbs a very large amount of water (46 g of water per gram of aerogel). Too much water in the aerogel makes the wet aerogel a thermally conductive mixture that has a similar density as water, even though the dry aerogel is not thermally conductive with...
a very low density (~10.8 mg cm\(^{-3}\)). The wet aerogel can float on the water independently, but most of the aerogel is submerged due to the increase in density. Thus, in addition to acting as a thermal insulator, the EPE foam is also working as a floating support to avoid the porous structure of the aerogel immersing into the bulk water.

For a solar powered steam generation system, the solar absorber is the first consideration factor. Among a variety of light absorbing materials, PEDOT:PSS is one of only a few water processable and biofriendly materials that displays electronic and vibrational transitions in the infrared range.\[^{[24]}\] Furthermore, polymers like PEDOT:PSS have inherently low bulk thermal conductivities (0.1–0.5 W m\(^{-1}\) K\(^{-1}\)).\[^{[25]}\] Hence, to enable broadband light absorption while minimizing heat transfer is a promising strategy to achieve a high solar-to-thermal conversion efficiency. As shown in Figure 2a, a PEDOT:PSS-NFC thin film has very high absorption in the NIR region (800–2500 nm), progressively lowering to 75% in the UV–vis region (250–800 nm). Even though a PEDOT:PSS thin film has lower absorbance in the visible region,\[^{[19]}\] the PEDOT:PSS-NFC aerogel can absorb almost all of the energy in the whole UV–vis–NIR range because the aerogel has numerous layers of thin films layered on each other. A 1 mm thick aerogel can already absorb more than 98% of the light, even in the UV–vis region. By increasing the thickness of the aerogel to 2 mm, over 99% of the light in the whole UV–vis–NIR region is absorbed. After freeze-drying the PEDOT:PSS-NFC water suspension, only PEDOT:PSS and NFC are left as continuous film sheets, whereas the water is evaporated, forming a microscale porous structure.\[^{[21,26]}\] Once the aerogel is exposed to solar light, most of the energy is readily absorbed by the first film sheet, as indicated by the absorption of the thin film. The unabsorbed energy passes through this sheet and is refracted to another sheet. By repeating this absorption-refraction process, most of the light is captured and absorbed by numerous PEDOT:PSS sheets inside the aerogel, as shown in Figure 2b. The efficient infrared absorption of the porous PEDOT:PSS-NFC is in drastic contrast with other porous plastic layers that do not absorb in the infrared range and rather reflect the infrared radiation to promote a cooling effect.\[^{[27]}\]

The solar energy absorbed by the PEDOT:PSS-NFC aerogel is converted directly to thermal energy via photothermal effect.\[^{[28]}\] The photothermal properties of the aerogel were investigated by illuminating both a dry and a wet sample with a solar simulator, followed by measuring their temperature with a forward-looking infrared (FLIR) camera. The results are shown in Figure 2c–e. The dry aerogel absorbs the energy from the light and converts it directly into heat, which is transferred to the
environment through thermal radiation and heat exchange. The temperature of the aerogel stabilizes with the highest temperature reaching up to 90 °C. In the case of wet aerogel, most of the heat from the aerogel is then transferred to the water which then evaporates. Thus, the highest temperature of the wet aerogel is only 39 °C, which is less than half what we measured for the dry aerogel. We believe that such a high temperature difference between the dry and wet aerogel is partly due to the porous structure of the aerogel and the endothermic process of evaporation. The porous structure (pore size =17 μm, Figure S1, Supporting Information) provides a large interfacial area between the water and PEDOT:PSS-NFC, therefore exchanging heat fast and efficiently. Importantly, the aerogel also provides a high surface area for the evaporation of water, which consumes most of the produced heat. Figure 2e shows the side view of the water evaporation setup with illuminating solar light from the top. The EPE foam localizes the heat efficiently between the aerogel and the bulk water, enabling a high energy conversion efficiency.

The water evaporation rate was recorded by measuring the mass loss as a function of exposure time using a computer controlled electronic balance. The mass change of water with time is shown in Figure 3a. By illuminating simulated solar light on pure water in the container, the evaporation rate of the water is 0.39 kg m⁻² h⁻¹. In comparison, the aerogel on top of the water increases the evaporation rate to 1.12 kg m⁻² h⁻¹, due to the much higher solar absorption and increased interfacial surface area. As discussed above, a part of the absorbed solar energy will be transported to the bulk water because of the good thermal conductivity of water (615 mW mK⁻¹ at 30 °C). This energy will be then lost to the environment through heat transfer and radiation. Therefore, a higher evaporation rate was recorded by adding an EPE foam under the aerogel to effectively reduce the heat transport from the aerogel to the bulk water. This achieved the highest evaporation rate of 1.61 kg m⁻² h⁻¹, which is over 4 times higher than the evaporation rate of pure water (see Table S1, Supporting Information). The stability of the aerogel by measuring the evaporation rate in multiple cycles is shown in Figure 3b. Thanks to the presence of GOPS that acts as a crosslinker,[9] the aerogel has good mechanical properties,[21,26] and the pore size as well as the whole aerogel size does not change before and after water evaporation (Figures S2 and S3, Supporting Information). Therefore, the evaporation rate is not affected by cycling the water evaporation (i.e., repeat wet–dry–wet process), which demonstrates that the aerogel is durable and reusable repeatedly without noticeable changes in performance.

The solar energy conversion efficiency is calculated using the equation of \[ \eta = \frac{m h_{\text{LV}}}{q_i C_{\text{opt}}} \] where \( \eta \) is the conversion efficiency, \( m \) is the evaporation rate after subtracting the evaporation rate under dark field, \( h_{\text{LV}} \) 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 \( C_{\text{opt}} \) is the optical concentration.[5] The pure water evaporation rate in dark is measured as 0.33 kg m⁻² h⁻¹. We further assume that the steam was generated at 39 °C, since that is the measured temperature of the illuminated aerogel surface. Thus, the calculated solar energy conversion efficiency is 81%. Both the evaporation rate and energy conversion efficiency of this work are relatively high level when compared with previous studies.[5,29]

Solar light is distributed so that approximately 5% of the total energy is contained in the ultraviolet range, about 45% in the visible range, and about 50% in the infrared range.[30] Classic black absorber materials such as graphene absorb mostly the visible light,[31] however, in the infrared range the absorption of such materials are relatively low. In contrast, as a conductive polymer pristine PEDOT with approximately 33% oxidation level has a wide absorption band in the infrared range thanks to the existence of polaron and bipolarons.[32] Hence, employing conductive polymers as solar absorbing materials is a promising strategy to optimize the energy utilization of solar light. To investigate the contribution of infrared light to water evaporation with our evaporator, we recorded the evaporation rate using two light sources with different spectral distributions: 1 sun intensity directly obtained from a solar simulator and 1.4 sun intensity from the solar simulator followed by a Schott KG3 1R cutoff filter (Newport). The former contains UV–vis–NIR light while the latter does not contain infrared light since the KG3 filter absorbs almost all of the light in the range above 800 nm. We employed a 1.4 sun intensity with the KG3 filter to ensure that the total optical

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**Figure 3.** a) Mass change versus exposure time for pure water, water + aerogel, and water + aerogel + foam under 1 sun irradiation. The measurement was started 10 min after switching the solar simulator on in order to stabilize the evaporation rate. b) Evaporation rate for pure water, water + aerogel, and water + aerogel + foam under 1 sun irradiation for ten cycles shown in (a).
power in the range below 800 nm was equal in both situations, as shown in Figure 4a. The power difference of these two situations is thus due to the absence or presence of infrared light. Figure 4b is the recorded mass change with exposure time, which shows the evaporation rate of 1.61 kg m\(^{-2}\) h\(^{-1}\) with 1 sun illumination while the evaporation rate without infrared light is 1.18 kg m\(^{-2}\) h\(^{-1}\). Hence, with our solar simulator, the water evaporation contributed by three parts: 1) heat from the environment, which is 0.39 kg m\(^{-2}\) h\(^{-1}\) (24%); 2) energy from UV–vis light, which is 0.79 kg m\(^{-2}\) h\(^{-1}\) (49%); and 3) energy from infrared light, which is 0.44 kg m\(^{-2}\) h\(^{-1}\) (27%). This confirms that one of the advantages of using a conductive polymer as the solar absorber is the high absorption of infrared light. Even through the infrared light can be absorbed by a thick classic black material, conductive polymers provide a possibility to decrease the thickness of the solar absorber.

A miniature greenhouse was built to purify three water samples from different sources, as shown in Figure 5a. The top image shows the side view of the system illuminated with a solar simulator. The bottom image shows the composition of the system: a source water zone to store and provide dirty or salty water to the aerogel and a purified water zone to collect the evaporated clean water. The steam generated from the aerogel is condensed on the surface of the glass dome and flows down to the purified water zone. Salt and other solid particles that do not evaporate remain in the source water zone and the aerogel. Thus, the purified water is separated from the source water as there is a wall between these two zones. GOPS ensured that the aerogel is washable and reusable as the mechanical properties are good (Figure S3, Supporting Information). Scaling up of the aerogel manufacturing is possible in order to answer the demand for large greenhouses as a real-world application. As shown in Figure 5b, the salinities of the three source water samples and the corresponding purified samples were measured. The first water sample is 0.6 mol L\(^{-1}\) NaCl with a salinity of 35 080 mg L\(^{-1}\), which is close to the average ocean salinity.[33] The second sample is seawater collected from the Baltic Sea having a salinity of 6658 mg L\(^{-1}\). The third sample is tap water with a salinity of 175 mg L\(^{-1}\). In contrast, the salinities of the corresponding purified water samples are 23, 22, and 22 mg L\(^{-1}\), respectively, indicating no correlation with the original salinities. Note that precipitation of salt inside the aerogel
eventually clots the porous structure responsible for water transport, thus slightly reducing the water evaporation rate by about 3.7% in 24 h. However, the initial performance is readily recovered after washing the aerogel with salty water. This process can be repeated multiple times which is necessary for any practical application (Figure S3, Supporting Information). In addition, the extremely low salinity of the purified water shows that the aerogel evaporation system operates efficiently.

3. Conclusion
In summary, we reported a high-performance aerogel solar steam generator based on purely organic materials. The aerogel comprised a cellulose scaffold decorated with PEDOT:PSS as the efficient infrared solar absorber. Thanks to the good absorption properties of PEDOT:PSS and the multilayer structure of the aerogel, a 2 mm thick sample was sufficient to absorb over 99% of the energy in the solar spectrum. The high absorption and porous structure of the aerogel yield large water evaporation rate of up to 1.61 kg m⁻² h⁻¹. The aerogels possessed good mechanical properties, enabling them to be washable and reusable. The solar steam generator was shown to purify salted water as well as natural sea water to a much lower salinity than the upper limit for good quality drinking water, demonstrating high feasibility of the aerogel system for water purification.

4. Experimental Section
Preparation of PEDOT:PSS-NFC Aerogel: The aerogel was fabricated by using PEDOT:PSS (Heraeus Clevios, PH1000, 1.3 wt% of PEDOT:PSS), NFC (carboxymethylated, 0.5%), and GOPS (Alfa Aesar, 97 wt%). These three components were first mixed as a solid ratio of 1:1:2 in suspension using PEDOT:PSS (Heraeus Clevios, PH1000, 1.3 wt% of PEDOT:PSS), followed by freeze drying (Benchtop Pro, SP SCIENTIFIC) under −60 °C for 30 min to crosslink the GOPS with the NFC. The aerogel comprised a cellulose scaffold decorated with good absorption properties of PEDOT:PSS and the multi-layer structure of the aerogel, a 2 mm thick sample was sufficient to absorb over 99% of the energy in the solar spectrum. The pore size of the aerogel was measured by an automated pore-volume distribution apparatus.

Temperature Measurements: The temperatures of the dry and wet aerogel with illuminated with simulated solar light were measured using an infrared camera (FLIR systems, ThermoVision A320C).

Water Evaporation and Salt Water Desalination Setup: The container for water evaporation was 3D printed. The setup for salt water desalination comprised of two parts: the middle (source water zone) and the collecting brim (clean water zone). They were 3D-printed from thermoplastic polyurethane (TPU 95A, Ultimaker B.V.) with an Ultimaker 2+ Extended printer. For evaporation rate measurement, the 3D printed container with deionized water was placed on an electronic balance (Ohaus Scout, SKX Portable Classroom Scale). A solar simulator (Newport, model 94011A) was employed to provide the simulated solar light vertically. The evaporation rate was recorded 10 min after the solar simulator was turned on.

Salinity Measurement: The seawater was collected in Arkösund (58°29′18″N 16°56′36″E) on 21st of May 2019. Salinities of the source water samples and the corresponding purified samples were measured with a salinity detector (NeuLog, Salinity logger sensor NUL-228) at room temperature.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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