**Graphene and Rice Straw Fibre Based 3D Photothermal Aerogels for Highly Efficient Solar Evaporation**

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### Experimental section

**Materials and chemicals:** Rice straw was obtained from the rice postharvest and byproduct management group of the International Rice Research Institute, Los Baños, Philippines. Reduced graphene oxide (RGO) was supplied by Huasheng Graphite Co., Ltd., China. Sodium alginate (SA) was purchased from Sigma-Aldrich. Sodium hydroxide and calcium chloride dihydrate were purchased from Chem-Supply. Unless otherwise noted, Milli-Q water with a resistance >18.2 MΩ cm–1 was used for all experiments.

**Preparation of cellulose aerogel:** In a 100 mL Teflon-lined stainless-steel autoclave, the cut rice straw (1 g) was added to an aqueous 5 M sodium hydroxide solution (60 mL). The autoclave was sealed, and the mixture was heated at 150 °C for 6 h. Thereafter the autoclave was allowed to cool naturally to ambient temperature before the resulting mixture was washed with water until neutral and centrifuged to separate and recover the solid fibres. The obtained cellulose fibres were then redispersed with ultrasonication (QSonica, model: Q125) in Milli-Q water (15 mL) to generate a homogenous suspension. Thereafter, 8.7 mL of this cellulose suspension (8.7 mL) was added into a 3.2 cm diameter container, pre-frozen (-20 °C) and freeze dried (-60 °C) to generate a cellulose aerogel with dimensions of 3 cm diameter x 1 cm thickness.

**Preparation of RGO-SA-cellulose aerogel:** RGO nanosheets (1 mg mL⁻¹) and SA (5 mg mL⁻¹) were initially combined with a cellulose dispersion, and thoroughly mixed via ultrasonication and stirring to produce a homogenous black RGO-SA-cellulose suspension. Afterward, the
produced black dispersion (8.7 mL) was added into a 3.2 cm diameter container, pre-frozen and freeze dried. The as-prepared aerogel sample was then immersed into an aqueous 5% (w/w) CaCl$_2$ solution overnight, washed with Milli-Q water several times, frozen and freeze dried to generate a Ca$^{2+}$ cross-linked RGO-SA-cellulose aerogel suitable for solar evaporation use. By controlling the amount of the RGO-SA-cellulose dispersion used, RGO-SA-cellulose aerogels of different heights were prepared.

**Preparation of RGO-SA aerogel:** RGO nanosheets (1 mg mL$^{-1}$) and SA (5 mg mL$^{-1}$) were initially mixed thoroughly with Milli-Q water, via ultrasonication and stirring to obtain a homogenous dispersion. Subsequently an aliquot of this homogeneous dispersion (8.6 mL) was added into a 3.2 cm diameter container, frozen and freeze dried. The obtained sample was thereafter immersed into an aqueous 5% (w/w) CaCl$_2$ solution overnight, rinsed for several times, pre-frozen and freeze dried to obtain Ca$^{2+}$ cross-linked RGO-SA aerogel for a comparison study.

**Characterization:** Scanning electron microscopy (SEM) images were obtained on a Zeiss Merlin scanning electron microscope. Transmission electron microscopy (TEM) images were obtained using a JEOL JEM 2100F transmission electron microscope. X-ray photoelectron spectroscopy (XPS) analysis was carried out on a Kratos Axis Ultra with a Delay Line Detector photoelectron spectrometer using an aluminium monochromatic X-ray source. UV-Vis spectra were recorded using a UV-2600 Spectrophotometer (Shimadzu). Infrared photographs were captured using an IR camera (FLIRE64501). A Dataphysics OCA 20 contact angle system was employed to characterize the hydrophilicity of the samples. Initial concentrations of common cations present in seawater (from Semaphore Beach, Adelaide, Australia) were measured using an Inductively Couple Plasma Optical Emission Spectrometry (ICP-OES, Optima 5300V, Perkin Elmer). Following desalination trace levels of residual ion concentrations in the collected clean water were analysed using an Inductively Couple Plasma-Mass Spectrometry (ICP-MS) Triple Quad system (ICP-QQQ, Agilent 8800).

**Solar-driven steam generation:**

Solar-driven steam generation was recorded under laboratory conditions (ambient temperature: 25 °C). A Newport Oriel Solar Simulator (Model: 69907) was used as the light source, and an electronic balance was connected to record the mass change during evaporation. The surface temperature of the photothermal aerogel was monitored by an infrared (IR) camera. A digital temperature and humidity sensor was applied for the real time monitoring of the evaporation
environment. In addition, an aperture with a hole size matching the size of the photothermal aerogel sample was applied to block any extra irradiation from the light source.
Figure S1. SEM images of cellulose fibres in the pure cellulose aerogel with (a) low magnification and (b) high magnification.
Figure S2. EDX spectrum and element mapping of the pure cellulose aerogel.
Figure S3. EDX spectrum and element mapping of RGO-SA-cellulose aerogel.
Figure S4. Digital photograph of the weight of the pendant (a) and copper cube (b) used for testing the mechanical stability of the RGO-SA-cellulose aerogel.
Figure S5. (a) Photograph showing the immersion of a photothermal aerogel in a 45 °C water bath overnight, and after that (b) the aerogel still can safely hold a weight of 206 g.
Figure S6. Photographs of two distinct samples showing differences in diameter (a) and thickness for (b) RGO-SA-cellulose aerogel (left) and RGO-SA aerogel (right) Photograph of the RGO-SA aerogel before, during and after compression of 0.5 kg pressure (c).
Figure S7. Time-dependent average temperature of the top evaporation surface (a) and time-dependent weight loss of water over a RGO-SA-cellulose aerogel (3 cm in thickness) after light-off (b). The values presented in Figure S6b shows the variation in the slope of the weight loss for different time periods.
Calculation:

Energy exchange between the 3 cm photothermal aerogel and the environment:

\[
E_{\text{environment}} = A_1 \varepsilon \sigma \left(T_1^4 - T_E^4\right) + A_2 \varepsilon \sigma \left(T_2^4 - T_E^4\right) + A_1 h(T_1 - T_E) + A_2 h(T_2 - T_E) + q_{\text{water}}
\]  

where \(A_1\) is the area of the top surface of the photothermal aerogel (7.06 cm\(^2\)), \(T_1\) is the steady average surface temperature of the top surface (\(\sim 30.7\) °C), \(A_2\) is the surface area of the side wall of the photothermal aerogel (28.26 cm\(^2\)), \(T_2\) is the average surface temperature of the side wall (\(\sim 19.5\) °C), \(T_E\) is the ambient temperature (25 °C), \(\varepsilon\) is emissivity of the absorbing surface (\(\sim 0.95\)), \(\sigma\) is the Stefan–Boltzmann constant (\(5.67 \times 10^{-8}\) W m\(^{-2}\) K\(^{-4}\)), \(h\) is the convection heat transfer coefficient (assumed to be 10 W m\(^{-2}\) K\(^{-1}\)), and \(q_{\text{water}}\) is the energy change between the photothermal aerogel and bulk water. A thermocouple inserted into the bulk water to monitor its temperature variation during the evaporation process found that after 60 min irradiation (1.0 sun), the temperature of bulk didn’t change, thus \(q_{\text{water}}\) was assumed to be 0 W.

According to equation 4, the radiation and convection loss from the top evaporation surface was 0.024 and 0.04 W respectively. While the energy gain from the environment from the side evaporation surface was 0.241 W, including 0.086 W of radiation energy gain and 0.155 W of convection energy gain.