Precision-Engineered, Polymer-Lean, Digital Light Processing 3Dprinted Hydrogels for Enhancing Solar Steam Generation and Sustainable Water Treatment

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Interfacial solar steam generation (ISSG) using hydrogels offers a sustainable approach to desalination, addressing global water scarcity challenges. However, conventional hydrogel fabrication methods, such as moulding or direct ink writing 3D printing, lack the precision to control micro- and/or macrostructures effectively. Digital light processing (DLP) 3D printing has emerged as a powerful alternative, enabling the reproducible and high-fidelity fabrication of hydrogels with precisely engineered structures. In this study, we developed a novel DLP printing "ink" that maintains excellent printability while minimizing precursor concentrations. Using this ink, we successfully printed hydrogels with tunable engineered structures, allowing for precise control over water transport and heat management. These hydrogels demonstrated a high evaporation rate of 3.56 kg m⁻² h⁻¹ and an impressive daily water production rate exceeding 10 L m⁻². This research thus advance the practical application of ISSG technology, providing a cost-effective and sustainable solution for freshwater production.

Introduction

Water scarcity is an urgent global issue, increasingly exacerbated by climate change, environmental pollution, population growth, and industrialization.^{1, 2} Approximately 52% of the global population is projected to reside in water-stressed regions by 2050.³ In particular, climate change is likely to intensify conflicts over water resources, contributing to food insecurity, and pose a significant threat to global peace and public health.^{4, 5} Therefore, to address this challenge, it is essential to develop high-yield, energy-efficient, and cost-

effective technologies for seawater desalination and wastewater treatment. $^{\rm 6\mathchar`10}$

Hydrogel-based interfacial solar steam generation (ISSG) is an emerging technique to harness the abundant solar energy to evaporate seawater or wastewater at the hydrogel surfaces. This approach offers several advantages, including zero energy consumption and rapid production of freshwater.¹¹⁻²² The micro- and macrostructure of hydrogels is closely linked to their ISSG performance, with specific surface patterns enhancing light capture,²³⁻²⁵ internal channels improving water transport,²⁶⁻²⁸ and unique shapes resisting fouling.²⁹ Responsive mold is a common method for constructing specialized hydrogel structures; however, mold selection and preparation can be time-consuming, labor-intensive, and often lack adaptability to new requirements.

The emergence of 3D printing technologies, such as Direct Ink Writing (DIW) and Digital Light Processing (DLP), offers more versatile and scalable approaches for hydrogel fabrication. Among these, DIW enables the extrusion-based construction of hydrogels but often struggles with precise control over internal structures and lacks the high-resolution printing capabilities required for intricate designs.³⁰⁻³⁴ DLP is a new additive manufacturing technique for rapid fabrication of hydrogels via layer-by-layer photopolymerization. Recent advances in DLP 3D printing have enabled the fabrication of high-fidelity hydrogels with precisely engineered structures, offering enhanced mechanical tunability and structural complexity for biomedical applications.^{35, 36} While several research groups have explored potential DLP applications for constructing hydrogels with complex architectures for ISSG,³⁷⁻⁴⁶ current printing inks often contain high precursor concentrations, reaching up to 90 wt%. Elevated precursor content typically increases costs, and the resulting printed hydrogels show reduced swelling ratios, slower water transport, and diminished evaporation rates.^{21, 26,} ⁴⁷⁻⁵⁰ These effects occur because, under the 405 nm near-UV light used in commercial DLP 3D printers, lower precursor concentrations result in fewer propogating chains during the

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photopolymerization within the given exposure period. Consequently, limited chain entanglement yields less cured structures that remain susceptible to further ink absorption, disrupting the curing process.⁵¹ Alternatively, while partial structure can be printed at low precursor concentrations, repeated soaking in the ink can cause excessive swelling and deformation,⁵² disrupting the continuity of the printed structures. Additionally, high water content increases UV light scattering during printing, which reduces precision and weakens interlayer adhesion. In certain non-ISSG applications, researchers have employed heat-assisted anhydrous DLP printing of macromonomers, followed by swelling to introduce water into the polymer network.53 However, this method necessitates a customized heat-enabled DLP printer, which limits its broader applicability and raises costs. Although, some strategies, such as ionic locking $^{\rm 41}$ and directional freezing $^{\rm 46}$ have been investigated to increase porosity, they often lead to longer manufacturing cycle times and higher costs, while also introducing complexity in fabrication. From a materials chemistry perspective, these challenges underscore the necessity of developing a novel DLP-compatible ink formulation that enhances porosity while maintaining scalability and ISSG efficiency, particularly for hydrogels in ISSG applications.

In this study, we present a novel DLP printing ink formulation containing a record-low 20 wt% precursor materials, including 5 wt% long-chain polyvinyl alcohol (PVA), for the 3D printing of ISSG hydrogels. Using this ink, we successfully printed hydrogels with complex macro/microstructures and high shape-fedility in various shapes. We then explored the balance between water and heat management across different hydrogels to optimize their ISSG performance. Our findings indicate that hydrogels with concave surfaces exhibit exceptional water transport properties and reduced heat loss, achieving a high evaporation rate of 3.56 kg m⁻² h⁻¹ under one sun irradiation. Furthermore, the hydrogels demonstrate remarkable daily water production, surpassing 10 L m⁻² in practical seawater desalination, coupled with excellent salt resistance and durability, positioning them as a sustaiable solution to global water scarcity.

Results and Discussion

Development of 3D Printing Ink

We employed a DLP 3D printer in bottom-up configuration (Fig. 1a) to produce micro/macrostructed hydrogels. A 405 nm near-UV light source was positioned beneath the ink tank to cure the precursors layer-by-layer, starting from the bottom and building up on the printing platform.To develop DLP-compatible inks suitable for printing ISSG hydrogels, we systematically screened a range of initiator, monomer and crosslinker combinations with fixed photothermal materials (PTMs). Following numerous iterations, with over a dozen unsuccessful attempts to maintain the total precursor content below 30 wt% (Table S1), we successfully formulated an optimized ink compatible with DLP 3D printing. As shown in Fig. 1b, this ink with a viscosity of 6.67 mPa·s consists of 2-hydroxyethyl acrylate (HEA) monomer, a small amount of long-chain PVA, trace amounts of a cross-linker

(polyethylene glycol diacrylate, PEGDA), a photoinitiator (lithium phenyl-2,4,6-trimethyl benzoylphosphinate, LAP), and graphene oxide (GO) as PTM. Water-based inks are more costeffective and environmentally friendly, and also prevent hydrogel breakage caused by solvent tension during the waterethanol exchange. PVA was incorporated to promote effective physical entanglement, thereby facilitating UV-curing and ensuring high fidelity of each printed layer. GO, selected as the PTM due to its strong solar spectrum absorption, contains surface functional groups (i.e. –OH, –COOH, –C–O–C–), capable of forming H-bonding with polymer chains. These interactions enhance dispersity, provide filler effects, and reinforce the hydrogel's mechanical properties. The high specific surface area of GO nanosheets further increase sunlight absorption, boosting photothermal conversion efficiency.⁵⁴ The GO content was meticulously regulated to prevent excessive UV-light absorption, which could hinder LAP activation.

As shown in Fig. 1c, before 3D printing, randomly tangled PVA chains were uniformly mixed in the ink tank. Upon UV irradiation of each 50 μ m-thick patterned layer, LAP absorbed photons, generating initiating radicals that initiate the polymerization of HEA and PEGDA. This chain propagation proceeded until termination, achieving full layer curing within 25 s under our optimized formulation. The long PVA chains intertwined with the PHEA chains, forming a semi-interpenetrating network. After printing, the hydrogels were subjected to UV irradiation and subsequently treated with glutaraldehyde (GA) as the crosslinking agent, using hydrochloric acid (HCl) as the catalyst to facilitate the crosslinking of PVA chains.

Characterizations of the 3D Printed Hydrogels

The resulting hydrogels are denoted as 5PVA15PHEA-X, where 5 and 15 represent the weight concentration of the PVA and PHEA respectively, and X stands for the designed shape of the hydrogel. Specifically, C, N, and H represent 3D-printed hydrogels with concave, raised-node, and vertical through-hole arrays on their surface, respectively (Fig. 2). We also 3D printed two hydrogels with higher precursor loading (25 wt%), named 5PVA20PHEA-C and 10PVA15PHEA-C. Comparative analysis of Figs. 2a-c (i and ii) reveals that the 3D-printed SSG hydrogels demonstrate high structural fidelity (Table S2), retaining their designed morphology even after freeze-drying, though a certain degree of volume shrinkage is observed due to water loss during the process (Table S3). Notably, our formulation achieves a printing accuracy exceeding 95% in the horizontal direction, while the vertical accuracy is slightly lower, likely due to the soft nature of the hydrogel, experiences slight compression during the sequential layer-by-layer printing process. Characterization of longitudinal sections shows distinct raised nodes (Fig. 2a-iii), as well as full-pass (Fig. 2b-iii) and half-pass (Fig. 2c-iii) channel structures. The uniform black coloration of the printed hydrogels confirms even distribution of GO nanosheets, ensuring effective photothermal conversion. SEM imaging postfreeze-drying further highlights the internal architecture, displaying a homogeneous pore structure on both the raised nodes and channel walls (Figs. 2a-c, iv and v, Table S4).



Compared to 5PVA15PHEA-C, hydrogels 5PVA20PHEA-C and 10PVA15PHEA-C exhibit thicker pore walls and lower pore size (Fig. S1 and Table S4), attributed to their higher precursor content.

Fourier-Transform infrared (FT-IR) spectroscopy analysis confirmed the successful preparation of the ISSG hydrogels through DLP 3D printing (Fig. 2d and Fig. S2). The characteristic absorption peaks of the vinyl group (-CH=CH₂) in HEA, typically observed around 910 cm⁻¹ and 990 cm⁻¹, were not detected in the FTIR spectra. This absence indicates a high degree of vinyl group consumption during photopolymerization, confirming the conversion of HEA monomers into the polymer network. All hydrogels displayed a characteristic absorption peak around 3,315 cm⁻¹, which is attributed to the stretching vibrations of



Fig. 2 Characterizations of the 3D printed hydrogels. For the 3D printed (a) 5PVA15PHEA-N, (b) 5PVA15PHEA-H, and (c) 5PVA15PHEA-C hydrogels: (i) designed 3D models, (ii) physical images (The 5PVA15PHEA-H sample was photographed top-down in water to emphasize through-pores, with slight pore deformation caused by uneven swelling.), (iii) longitudinal cross section optical images, (iv) cross-sectional SEM images, and (v) the zoom-in SEM images showing the pores on the raised node structure of 5PVA15PHEA-N, the hole wall of 5PVA15PHEA-H, and the concave structure of 5PVA15PHEA-C, respectively. (d) FT-R spectra of the 3D printed 5PVA15PHEA-N, H, C hydrogels. A control sample of pristine 5 wt% PVA hydrogel is included. Physical image of (e) a 3D pattern (with a total height of 4 mm and 2 mm depth of "UTS" patterns) and (f) three ASTM [E8] subsize standard hydrogel specimens for tensile test. (g) Stress-strain curve of the printed 5PVA15PHEA hydrogel in the tensile test. (h) UV-VIS-NIR adsorption spectrum of the 3D printed 5PVA15PHEA hydrogel and the air mass 1.5 global (AM 1.5 G) solar spectrum with normalized spectral solar irradiance density (the light-yellow region) in the wavelength range from 300 to 2,500 nm.

the hydroxyl groups (-OH). Notably, the printed hydrogels exhibited a stronger signal compared to that of the pristine PVA hydrogel, a result of the enhanced O-H vibrations from PHEA. Additionally, distinct peaks at 1,724 cm⁻¹ and 1,070 cm⁻¹ were associated with the vibrations of the C=O groups of PHEA and C-O-C groups of GO, respectively.

Using this printing ink, we also produced additional 3D objects with favorable model fidelity (Fig. 2e, Fig. 2f and Fig. S3), and used the printed ASTM (E8) subsize standard hydrogel specimens for tensile testing. The stress-strain curve presented in Fig. 2g indicates that the printed hydrogel exhibits a tensile strength of 71.9 kPa and a Young's modulus of 357 kPa. These findings demonstrate that the printed hydrogels remain rigidity even at low precursor concentrations, attributable to the incorporation of PVA and GO. This combination enhances the mechanical properties by promoting effective chain

entanglements and facilitating H-bonding within the matrix. A minimal concentration of GO at 0.25 wt% to create a hydrogel that demonstrates a broad spectrum of light absorption across the entire solar range, from 300 nm to 2,500 nm, achieving an absorption rate exceeding 80% (Fig. 2h). The contact angle data (Fig. S4) demonstrates that the porous structure of the printed hydrogel, combined with its abundant hydrophilic groups, allows for rapid water absorption within 80 ms. These results confirm the successful 3D printing of hydrogels with macro/micro-engineered structures and high porosity using a water-based, cost-effective ink with low precursor concentration. These hydrogels exhibit broad light absorption, high mechanical performance, and super-hydrophilicity, making them well-suited for SSG applications.

ISSG performance of printed hydrogels



Fig. 3 The SSG performance of the printed hydrogels. (a) A home-made set up for the SSG test. (b) Water mass changes under 1 sun irritation condition for 5PVA15PHEA-N, H, C hydrogels, and pure water. (c) The temperature variations of the SPVA15PHEA-N, H, and C hydrogel surfaces and bulk water during the SSG test under one sun irritation. The infrared images showing the temperature equilibrium of these three differently shaped hydrogels after one hour of irritation are displayed on the right. (d) The graph showing the link between the water absorption time and the water content per gramme of the corresponding dry gel. The schematic illustrating of the (e) water transport and (f) thermal transfer in 5PVA15PHEA-N, SPVA15PHEA-H, and 5PVA15PHEA-C. (g) Fitting curves for the 5PVA15PHEA-C hydrogel in the Raman spectrum within the O-H stretching energy region. Free water is represented by the green peaks, whereas intermediate water is represented by the blue peaks. (h) All the printed hydrogels' IW:(IW+FW) ratios calculated from their Raman spectrum, and the equivalent water vaporization enthalpy of the water in the hydrogels calculated from their DSC curves. Each error bar shows the difference from at least two hydrogel samples. (i) Comparison of the evaporation rates and precursor concentrations to state-of-the-art DLP ^{41, 43-46, 55} and DIW ³⁰⁻³⁴ 3D-printed ISSGs (typical three-dimensional ISSGs were not included). Detailed data are listed in Tables S5 and S6.

We then assessed the solar-driven evaporation rates of the 3Dprinted hydrogels at 24 $^{\circ}$ C and 1 atm (Fig. 3a). We monitored the mass changes of bulk water containing hydrogels over two hours of solar irradiation (Fig. 3b and Fig. S7), and calculated the evaporation rate by determining the slope from a linear fit. Among the various structures of 3D-printed hydrogels,

5PVA15PHEA-C achieved the highest evaporation rate of 3.56 kg m⁻² h⁻¹, followed by 5PVA15PHEA-H with a rate of 3.04 kg m⁻² h⁻¹, while 5PVA15PHEA-N had the lowest evaporation rate of 2.86 kg m⁻² h⁻¹, all significantly higher than static water evaporation rate.

To further investigate the impact of designed structures on ISSG performance, their heat and water managements were discussed as follows. IR images were taken during the ISSG process using a handheld infrared camera to record the temperatures of bulk water and evaporation surface (Fig. 3c and Fig. S5). During the initial 5 minutes of exposure, the surface temperatures of all hydrogels exhibited a rapid increase. Then, the surface temperatures of 5PVA15PHEA-N and 5PVA15PHEA-C rose to approximately 43 °C, while the bulk water temperature stabilized at around 32 °C. This result can be attributed to the enhanced light absorption facilitated by the hydrogels' surface patterning (Fig. 3f). In contrast, 5PVA15PHEA-H achieved a lower equilibrium surface temperature of 35 °C due to heat loss through the vertical holes, which transferred more heat into the bulk water, raising the water temperature to 34 °C (Fig. 3f). PVA20PHEA-C and 10PVA15PHEA-C, sharing the same structure as 5PVA15PHEA-C, showed similar temperature variations (Fig. S5).

We also assessed the water uptake of ISSG hydrogels by immersing dried gels in DI water. As shown in Fig. 3d, 5PVA15PHEA-H exhibited the fastest water absorption rate and the highest swelling ratio due to the capillary effect of its 2 mmwide vertical holes (Fig. 3e). The 5PVA15PHEA-C showed a moderate water uptake speed and swelling ratio due to weaker capillary effect of its shorter concave structure. The 5PVA15PHEA-N, which lacks holes, relied solely on water transport through hydrogel matrix. As a result, it showed the slowest water uptake and the lowest swelling. We also found that higher precursor concentrations slowed water transport due to thicker pore walls (Fig. S6).

Considering both heat and water managements in ISSG process, 5PVA15PHEA-C stands out due to its concave structure, enhancing evaporation surface temperature and water transport. Although 5PVA15PHEA-H benefits from rapid capillary water transport through vertical holes, the large volume of water present in these holes results in significant heat loss, which in turn leads to a moderate evaporation rate. In contrast, 5PVA15PHEA-N, with features a raised-node array, attains a higher surface temperature; however, it does not possess an improved water transport structure, resulting in a slow rate that is inadequate for continuous evaporation.

The presence of intermediate water (IW)^{47, 56, 57} is often used to explain why the water absorbed in hydrogels evaporates more readily than bulk water (Supporting information S1.10). Raman spectroscopy was conducted on fully saturated hydrogels, employing peak fitting analysis to quantify the IW content in each sample (Fig. 3g and Fig. S8). The IW content was assessed based on the ratio of IW to the sum of IW and free water (FW) [IW:(IW+FW)], calculated from the integrated areas under the fitting spectral peaks.^{20, 21} As shown in Fig. 3h, the IW contents of 5PVA15PHEA-C, N, and H are almost identical around 0.52, because the same printing ink used. In contrast, hydrogels with higher precursor concentration of 25 wt% show lower IW content, aligning with the observed evaporation performance. Fig. 3h shows the calculated [IW:(IW+FW)] values and the corresponding equivalent enthalpies of water evaporation measured by differential scanning calorimetry (DSC) (Fig. S9). Increased IW content lowers the equivalent enthalpy, thereby enhancing the evaporation rate.

The aforementioned attributes collectively enhance the evaporation performance, underscoring the efficacy of the water-based printing ink for ISSG applications. Compared to state-of-the-art DLP 3D-printed ISSG evaporators, the macro/microengineered hydrogel introduced in this study demonstrates a superior evaporation rate while utilizing the lowest precursor concentration. Additionally, it surpasses DIW 3D-printed ISSGs, which lack the same level of precision in controlling internal structures (Fig. 3i).

Desalination and water purification performance of printed hydrogels

Building upon the successful implementation of the 3D-printed hydrogels in ISSG, we further assessed their potential for seawater desalination under practical conditions. The performance of the 5PVA15PHEA-C, N, and H hydrogels was tested using real seawater sourced from Darling Harbour, Sydney, Australia (E151.20°, S33.87°), as well as a 20 wt% simulated brine solution. The 5PVA15PHEA-C hydrogel achieved an evaporation rate of 3.42 kg m⁻² h⁻¹ in seawater, and sustained a rate of 3.06 kg m⁻² h⁻¹ in challenging high-salinity environment. The performance declined as salinity increased, likely due to salt ion convection during evaporation, which carried away heat from the evaporating surface. Notably, no visible salt accumulation was observed after 8 hours of operation in seawater (Fig. S10). To further investigate its salt resistance, NaCl crystals were intentionally added on the surface to simulate accumulated salts (Fig. S11). For 5PVA15PHEA-C, salt crystals completely dissolved over approximately 200 minutes, aided by the Marangoni effect from its concave structure.^{37, 58} The long-term durability assessment of 5PVA15PHEA-C demonstrated stable evaporation performance of 3.50 ± 0.25 kg m⁻² h⁻¹ over a period exceeding four weeks (Fig. 4b).

To assess the efficiency of desalination and wastewater treatment, a sealed jar was used to collect condensate (Fig. S12). Subsequent analysis of the condensate using ICP-MS analysis revealed a significant reduction in the concentrations of major ions, including Na⁺, Mg²⁺, K⁺, and Ca²⁺, by four to five orders of magnitude relative to untreated seawater (Fig. 4c), well below the World Health Organization drinking water recommendation.⁵⁹⁻⁶² The UV-vis spectrum of the condensate obtained from ISSG of simulate industrial effluents showed the absence of characteristic peak of methylene blue at 663 nm (Fig. 4d), indicating a high level of effluent purification capability.

In practical application, enhancing vapor condensation efficiency is vital for maximizing freshwater productivity.⁶³ To address this, a custom-designed outdoor ISSG device was employed (Fig. 4e and Fig. S14), featuring enlarged 3D-printed hydrogels (Fig. S13), fans, and thermoelectric modules. A solar



Fig. 4 The desalination and water purification performance of the 3d printed hydrogels. (a) The evaporation rates of 5PVA15PHEA-C, N, H in DI water, seawater and simulated brine (20 wt% NaCl solution). Each error bar shows the difference from at least three hydrogel samples. (b) Evaporation rates of 5PVA15PHEA-C in seawater over two weeks. Insets: SSG performance after six and fourteen days of seawater exposure. (c) Four significant ion concentrations in seawater before and after the desalination of 5PVA15PHEA-C, N, and H hydrogel based solar steam generators. (d) The UV-vis spectra of simulated wastewater containing dye (MB) before and after the purification of 5PVA15PHEA-C, hydrogel based solar steam generators. Insert: the optical photos of the MB solution before and after SSG. (e) The environment temperature, humidity, and solar light intensity during the day of outdoor SSG test on 7th October 2024 in Sydney, Australia Insert: The digital photo of our outdoor SSG device. (f) The evaporation and condensation water volume of our SSG device.

panel was employed to provide real-time power to the fans and thermoelectric modules. The outdoor desalination experiment was carried out on a day with an average solar flux of 0.47 kW m⁻² and a peak solar flux of just 0.72 kW m⁻². Despite these conditions, the system demonstrated impressive performance, achieving a daily seawater evaporation rate of 12.73 L m⁻² and freshwater production of 10.36 L m⁻², with a condensation efficiency of 81.4% (Figs. 4e-f).

Conclusions

In this study, we have successfully developed a novel DLP 3D printing ink by incorporating a small amount of long-chain PVA to facilitate the formation of effective physical entanglements, thereby stabilizing the photo-crosslinked polymer network and preserving the integrity of each printed layer. The optimized formulation substantially lowers the overall precursor concentration to 20 wt%, producing 3D-printed hydrogels with

enhanced porosity, rapid water absorption and robust mechanical properties. By precisely adjusting the macro/microstructure of the hydrogels, we achieved a nearoptimal evaporation rate of 3.56 kg m⁻² h⁻¹, enabling freshwater production of 10.36 L m⁻² day⁻¹ with a condensation efficiency of 81.4%. The system effectively reduced major ion concentrations (Na⁺, Mg²⁺, K⁺, Ca²⁺) by 4-5 orders of magnitude, ensuring high-quality desalination performance. This study therefore offers a scalable, cost-effective, and environmentally sustainable solution for freshwater production, contributing to the global effort to combat water scarcity.

Experimental Section

Materials

All the chemicals, including 2-hydroxyethyl acrylate (HEA, 96 wt% in DI water), PVA (MW = 89,000-98,000 g mol⁻¹), poly(ethylene glycol) diacrylate (PEGDA, average M_n = 575 g

mol⁻¹), lithium phenyl-2,4,6-trimethylbenzoylphosphinate (LAP), graphene oxide (GO, powder, 15-20 nanosheets, 4-10% edge-oxidized), glutaraldehyde solution (GA, 25 wt% in DI water), hydrochloric acid (HCl, 32 wt% in DI water), sodium chloride (NaCl), and methyl blue (MB) were purchased from Sigma-Aldrich Australia and used directly without any further purification.

Development of 3D Printing Inks

The printing ink of 5PVA15PHEA was chosen as a sample to present the detailed preparation process, where 5 and 15 represent the weight concentration of the corresponding precursor. 10 g PVA powder was dissolved in 100 mL deionized (DI) water at 90 oC for 5 hours to get a 10 wt% PVA solution. 100 mg GO powder was evenly dispersed in 100 mL DI water via sonication for 2 hours to afford a 1 mg mL⁻¹ GO solution. 15 mL PVA solution and 15 mL GO solution were mixed together in a light-proof container, then 4.5 g HEA, 67.5 mg PEGDA and 60 mg LAP were added. The mixture is stirred for 5 minutes at 2000 rpm to obtain the 3D printing ink, denoted as 5PVA15PHEA. Compared to the 5PVA15PHEA printing ink, 10PVA15PHEA contains double the amount of PVA, while an additional 1.5 g of HEA and 22.5 mg of PEGDA are added to 5PVA20PHEA.

3D Printing of Hydrogels

Different targeted 3D structures were designed by Pro/ENGINEER software and saved as 'obj' files. After opening the 'obj' file with Anycubic Photon Workshop 3D Slicer Software, the following printing parameters were set: the z-axis moving rate was 1 mm s⁻¹, the layer thickness was 50 μ m, and each layer's normal exposure time was 25 s. The bottom 10 layers had an exposure time of 30 s. A USB flash drive was used to store the defined slicing 'dl2p' file for 3D printing utilizing a DLP 3D printer (Anycubic Photon D2) with a near-ultraviolet light (λ = 405 nm). The intended printing program was then executed once the prepared photo-responsive ink was poured into the DLP 3D printer's resin tank.

After printing, the remaining ink on the surface of 3D printed objects was cleaned gently with DI water. And then the 3D printed hydrogels were then further cured in a UV curing machine (Anycubic) for 30 minutes to fully cure the uncrosslinked components. To crosslink the PVA chains in the hydrogels, the 3D printed hydrogels was submerged in solution containing 0.75 wt% GA and 0.16 wt% HCl for 30 minutes, and then the process was repeated twice. After washing the hydrogels again with DI water to remove all the residuals. The resultant 3D printed hydrogels were frozen at -18 °C overnight and freeze-dried prior to characterization. Additionally, they were all thoroughly saturated in DI water, seawater or a 20 wt% NaCl solution piror to the solar steam generation test.

Charaterizations

The viscosity of the 3d printing ink was confirmed by a Brookfield DV2T viscometer at a constant temperature of 25 °C using a spindle SC4-29. The spindle speed was controlled at 120 rpm. The printed structure and pore structure of the hydrogels were investigated using a Zeiss scanning electron microscope (SEM) at an operating voltage of 10–30 kV. Fourier transform infrared (FT-IR) spectra were acquired with a Shimadzu MIRacle

10 FT-IR system. UV-vis-NIR absorption spectra, covering the wavelength range of 300-2500 nm, were recorded using a Perkin Elmber Lambda 950 UV-visible-NIR spectrophotometer. The tensile test of the printed hydrogel was conducted by Shimadzu AGS-X Universal Tester (Max load 10 kN) with a strain rate of 50 mm min⁻¹. The dimensions of the tested samples specimens were length = 100 mm, width = 6 mm, and thickness = 5 mm according to ASTM (E8) subsize standard. Raman spectra were collected via a Renishaw Raman spectrometer. The contact angle test was conducted by an Attension contact angle meter. Thermal changes in the hydrogels, from room temperature to 200 °C, were monitored with a NETZSCH DSC300 Supreme at a heating rate of 5 °C min⁻¹. An Agilent 7900 ICP-MS was employed to analyze the concentrations of the four primary ions in both seawater and desalinated water. An Agilent Cary 60 UV-Vis Spectroscopy was used to detect the MB in water before and after SSG purification.

Interfacial Solar Steam Generation

A solar simulator (NBeT HSX-F3000 xenon light source) was used to simulate solar radiation. With the aid of a thermal power sensor and a portable power and energy meter (PM100D and S405C, Thorlabs, Germany), the solar irradiance on the hydrogel surface was calibrated to one sun (1 kW m⁻²). The test 3Dprinted hydrogels were placed on the surface of a floating EPS foam atop a beaker filled with DI water, seawater, or a 20 wt% NaCl solution. Several tissues passing through a central hole of the EPS foam were used to supply water to the bottom of the testing hydrogels. Using an electronic mass balance (OHAUS Pioneer IC-PX 124), water mass loss was tracked over time during SSG tests. An EPE foam layer was placed between the balance and the beaker to stop heat transfer. By using linear fitting to calculate the slopes of the mass loss curve over time, the evaporation rates were ascertained. Using a Fluke PTi120 pocket thermal imager, the temperatures of both the bulk water and the hydrogel surface were measured every five minutes during the test.

Outdoor Desalination Experiments

The outdoor solar steam generation test was performed using a custom-built evaporation device, which includes ventilation and cooling features (As shown in Fig. 4e and Fig. S14). Specifically, clear acrylic sheets (thickness: 5 mm) were cut into designed shapes and sticked together with water-proof glue. The assembled device was divided into two separate chambers: a larger evaporation chamber and a smaller condensation chamber. Two 6 cm × 6 cm square fans were embedded in the partition between these two chambers, with the airflow directed from the evaporation chamber toward the condensation chamber. Two TEC1-12706 thermoelectric (TE) modules were embedded at the other end of the condensation chamber, with their cooling sides fully immersed in the condensation chamber, while the heating sides were located outside the device and cooled by a water-cooling system to prevent overheating. The evaporation chamber contained an independent seawater reservoir, where a piece of expanded polyethylene (EPE) foam completely covered the seawater, and several tissue strips passed through small holes in the foam to supply water to the hydrogel. The hydrogels, electric fans, and 5. cooling side were positioned on the same horizontal plane to ensure that a larger amount of vapor could be transferred to the condensation chamber and promptly condensed by the cooling side. The independent seawater evaporation chamber 7. could also condense vapor around its perimeter due to the lower temperature of the seawater, making it easier to collect 8. any vapor not transferred to the condensation chamber. The TE module, electric fans, and water-cooling pump were all 9. connected to the output of a Powertech Portable 155W Power Centre as the charger controller, powered in real-time by a 12V 10W solar panel.

Author contributions

Shudi Mao contributed methodology, software, validation, inverstigation, visualization, analysis, and writing-original draft. Xin Stella Zhang, Yihan Shi, and An Feng performed investigation, analysis, and resources. Casey Onggowarsito contributed to resources and analysis. Xiaoxue Helen Xu and Lisa Aditya carried out investigation. Youyi Sun and Long D. Nghiem conducted writing-review & editing. Qiang Fu approved funding acquisition, methodology, conceptualization, visualization, and writing—review & editing.

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Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the Supplementary Information.

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