Supporting Information (SI) for

Modular design of solar-thermal nanofluidics for advanced

desalination membranes

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Section S1: Detailed synthetic procedure of UCP-x membranes

Materials

Pyromellitic acid (BDC-COOH), ZrOCl₂·8H₂O and dimethylsilicone were purchased from Aladdin Industrial Co (China). Benzoic acid (BA), dimethylformamide (DMF), N-methyl-2-pyrrolidinone (NMP), acetone, tetrahydrofuran (THF), Bis(trifluoromethane)sulfonimide lithium salt (Li-TFSI, purity = 99%) were supplied by Zhengzhou Alpha Chemical Co. Ltd (China). Azodiisobutyronitrile (AIBN) was purchased from Tianjin Fuchen Chemical Reagents Factory (China). 1-Vinylimidazole and bromoacetonitrile were purchased from Aladdin Industrial Co (China). All the chemicals were at least analytical grade and used without further purification. Reduced graphene oxide (rGO) and black carbon (BC) were purchased from Nanjing Xianfeng nanometer Co. Ltd (China). Simethicone and lubricating oil were purchased from Huaxia Jujiang Ecommerce Ltd. Multi-walled carbon nanotubes (CNTs) with a diameter of 35-60 nm and length of ca. 30 µm was purchased from Chengdu Institute of Organic Chemistry (China). CNTs were ball-milled at 400 rpm for 6 h before being used, and the length was reduced to 0.3-1 µm. Poly(vinylidene fluoride) (PVDF, diameter = 50 mm, pore size = 0.22μ m) filter membranes were bought from Shanghai Zuofei Experimental Equipment Co. Ltd (China). Wastewater was collected from Huxi river (Wuhan), respectively. Seawater was collected from the South China Sea (near to Hainan). Oil/water emulsion (dimethylpolysiloxane concentration = 10000 ppm) was prepared as simulated wastewater.

Characterization

Powder X-ray diffraction (PXRD) was performed on a Rigaku Smartlab 9 kW X-ray diffractometer at room temperature, in parallel beam geometry employing Cu K α lines focused radiation at 9 kW (45 kV, 200 mA) power. The morphology was observed by means of a field-emission scanning electron microscope (SEM, SU8010) with the accelerating voltage of 5 kV. ¹H nuclear magnetic resonance (¹H NMR) spectrum was executed on Bruker Avance III400MHz. Fourier-transform infrared spectroscopy (FT-

IR) was performed in Shimadzu, Japan, IR-435 at room temperature. The sample was mixed with KBr at the weight ratio of 1:150. The morphology and EDS mapping were observed by field-emission scanning electron microscopy (SEM, Hitachi, S4800ESEM-FEG). N₂ adsorption/desorption experiments were executed on the Micromeritics ASAP 2460 under a liquid nitrogen bath (77 K). The absorption spectrum was tested using a UV-Vis-NIR spectrophotometer (Lambda 750 S) with an integrating sphere. UV-Vis absorption spectrum was performed with UV-Vis spectrophotometer (UV-6100 METASH). Optical images were got by Mshot MS60 optical microscope.

Synthesis of UiO-66-COOH

UiO-66-COOH was synthesized according to reported works^[1]. Briefly, BDC-COOH (0.2920 g), $ZrOCl_2 \cdot 8H_2O$ (0.1823 g) and BA (1.0850 g) were dissolved in 4 mL DMF solution, heated in an oven at 150 °C for 48 h. The as-synthesized UiO-66-COOH was washed by DMF and acetone for three times. Finally, the white UiO-66-COOH powder was dried in vacuum at 40 °C for 16 h.

Synthesis of 1-cyanomethyl-3-vinyl-imidazolium bromide^[2]

1-Vinylimidazole (10 g) and bromoacetonitrile (15 g) were dissolved in 125 mL THF. After heating in oil bath at 60 °C for 10 h, the precipitates, *i.e.*, 1-cyanomethyl-3-vinyl-imidazolium bromide, were vacuum filtered, washed with THF three times, and vacuum dried at 50 °C for 12 h.

Syntheses of poly[1-cyanomethyl-3-vinyl-imidazolium bromide] (PCMVIMBr) and poly[1-cyanomethyl-3-vinylimidazolium bis(trifluoromethanesulfonyl)-imide] (PCMVIM)

PCMVIMBr and PCMVIM were synthesized according to the reported procedures^[3]. 1-Cyanomethyl-3-vinyl-imidazolium bromide (10 g) and AIBN (0.08 g) were dissolved in 100 mL DMSO, heated at 60 °C for 12 h under nitrogen atmosphere. Afterwards, the solution was dropped into excessive THF (500 mL), and the precipitates (PCMVIMBr) were washed with THF three times, vacuum dried at 60 °C for overnight. 10 mL of Li-TFSI (1.4 g/mL) solution was dropped into 300 mL of PCMVIMBr aqueous solution (15 mg/mL). The precipitate (PCMVIM) was collected, washed by fresh water three times, and dried in vacuum at 50 °C for 16 h.

Energy balance analysis

The heat flux of sunlight is 1 kW/m², and the energy consumption consists of five components: (1) water evaporation consumption, (2) reflection energy loss, (3) conductive loss from materials to water, (4) radiation loss and (5) convection loss from the materials to environment.^[4] The details of analysis are shown below.

(1) Water evaporation consumption (η_c)

The water evaporation consumption is equal to evaporation efficiency. Therefore, the η_c is 80.2%.

(2) Reflection loss (η_{ref})

The solar absorption of UCP-10 is 99%. Therefore, the reflection loss η_{ref} is 1%.

(3) Radiation loss (η_{rad})

The radiation energy, Q_{rad} can be calculated based on the Stefan-Boltzmann law:

$$Q_{\rm rad} = \varepsilon \sigma (T_1^4 - T_2^4) \qquad (S1)$$

Where ε is the emissivity and σ is Stefan-Boltzmann constant (5.67×10⁻⁸ W/m²/K⁴). The emissivity of UCP-10 surface is 1. T₁ and T₂ are the steady-state surface temperature under one sun irradiation and room temperature (316K), respectively. Therefore, the radiation flux is 74.8 W/m².

Then, the radiation loss can be calculated by:

$$h_{\rm rad} = Q_{\rm rad} / P_{\rm in}$$
 (S2)

Under one sun irradiation, the loss η_{rad} is calculated to be 7.5%.

(4) Convection loss (η_{conv})

The convection heat loss can be calculated by Newton's law of cooling.

$$P_{\rm conv} = hA_{\rm sur}(T_1 - T_2) \quad (S3)$$

where h is the convection heat transfer coefficient (5 W/m²/K), and A_{sur} is the surface

area (530 mm²) of UCP-10. T₁ and T₂ are the steady-state surface temperature. Therefore, the calculated η_{conv} is 3.1%.

(5) Conduction loss (η_{cond})

The heat energy of conduction Q_{cond} , which refers to the heat transferred from the asprepared materials to water can be calculated:

$$Q_{\rm cond} = Cm\Delta T \tag{S4}$$

Where *C* is the specific heat capacity of pure water (4.2 kJ·kg^{-1.o}C⁻¹), *m* is the weights of water (50 g), and ΔT (0.6 °C) represents the increased temperature of bulk water before and after stable solar steam generation. Under 1 kW·m⁻² solar irradiation, the conduction heat loss is 3.5% according to Equation S4.

Based on these methods, the values of refection loss, radiation, convection and conduction are 1%, 7.5%, 3.1% and 3.5%, respectively. The total energy consumption containing energy conversion of water evaporation (80.2%), refection loss (1%), radiation loss (7.5%), convection loss (3.1%), and conduction loss heat (3.5%) under 1 kW·m⁻² solar irradiation is calculated to be 95.3%, which is very close to 100%.





Figure S1 (a) Synthetic route to PCMVIM. (b) ¹H NMR spectrum of PCMVIM using DMSO- d_6 as the solvent. (c) GPC trace of the PCMVIMBr.

Note: the apparent number-average molecular weight and polydispersity index value of PCMVIMBr are measured to be 1.97×10^5 g/mol and 2.67, respectively. Since the PCMVIM was synthesized by anion exchange of PCMVIMBr with Li-TFSI salt in aqueous solution, the apparent number-average molecular weight of the PCMVIM is calculated to be 3.80×10^5 g/mol.



Figure S2 PXRD pattern of UiO-66-COOH.

Please note: the diffraction peaks of UiO-66-COOH match well with the simulated pattern.



Figure S3 SEM image (a) and particle size distribution plot (b) of UiO-66-COOH.

Note: the particle size of UiO-66-COOH is around 35–55 nm.

10

10

10

10

UCP-10

UCP-12.5

UCP-15

UCP-20

= 10 : n : 4.3) membranes. x stands for the mass ratio of CNTs in the MMMs							
Sample name		CNTs (n)	PCMVIM				
(UCP-x)	010-00-00011	CIVI3 (II)					
UCP-0	10	0	4.3				
UCP-2.5	10	0.367	4.3				
UCP-5	10	0.753	4.3				
UCP-7.5	10	1.159	4.3				

1.589

2.043

2.524

3.575

4.3

4.3

4.3

4.3

Table S1 Mass ratio of different parts in UCP-x (UiO-66-COOH : CNTs : PCMVIM)	[
= $10 : n : 4.3$) membranes. x stands for the mass ratio of CNTs in the MMMs	



Figure S4 SEM image of the cross-section of UCP-10 membrane. Please note: the UCP-10 thickness is measured to be $13.7 \,\mu m$.



Figure S5 FT-IR curves of UiO-66-COOH, PCMVIM, PCMVIM (NH₃-treated), and UCP-10 membrane.

Note: after the treatment in NH₃ (0.2 bar) at 25 °C for 14 h, two new absorption peaks at 1015 and 1621 cm⁻¹ were observed in the PCMVIM (NH₃ treatment) and UCP-10 membrane, corresponding to the symmetrical stretching vibration (v_s) and stretching

vibration (v) of N-C=N in triazine ring structure, respectively. These results prove the crosslinking of the cyan groups in PCMVIM.^[5]



Figure S6 N₂ adsorption isotherms of (a) UiO-66-COOH, (b) UCP-10 membrane and (c) Pore size distribution plots of UiO-66-COOH nanoparticle (black line) and UCP-10 membrane (red line).



Figure S7 (a) SEM image of UCP-10 membrane. (b) Energy dispersive x-ray (EDX) spectroscopy maps of O elements in the UCP-10 membrane.

Note: the uniform distribution of N and Zr elements illustrates that UiO-66-COOH and PCMVIM are mixed homogeneously in the UCP-10 membrane.



Figure S8 SEM images of (a) the DMF/H₂O (v/v = 1:6) solution of UCP-10 and (b) UCP-10 membranes (no NH₃ treatment) after ultrasonication and (c) surface of UCP-10 membrane (no NH₃ treatment) after ultrasonication.



Figure S9 Photographs of various hybrid membranes before (top) and after (middle) being treated by ultrasonication in DMF/H₂O (v/v = 1:6) for 10 mins. Vials in the bottom pannel are supernatant DMF/H₂O solution after ultrasonication (UCPEG, UCPVA, UCPAN, UCPEGME, UCBP-10 and UrGOP-10 are abbreviation of UiO-66-COOH/CNTs@PEG, UiO-66-COOH/CNTs@PVA, UiO-66-COOH/CNTs@PAN, UiO-66-COOH/CNTs@PEGME, UiO-66-COOH/Carbon Black@PCMVIM-10, and UiO-66-COOH/rGO@PCMVIM-10, respectively).



Figure S10 SEM images of UCP-10 membranes (a) before and (b) after being treated by ultrasonication in DMF/H₂O (v/v = 1:6) for 10 mins.

Section S3: Interfacial solar steam generation using UCP-10 membrane



Figure S11 Vis-NIR absorption spectrum of UCP-10 membrane.



Figure S12 Photograph (a) and schematic illustration (b) of interfacial solar steam generation instrument used in this work.



Figure S13 Water mass change using UCP-10 membrane, controlled membranes and bulk water under 1 kW/m² (UP and CP-10 are the abbreviation of UiO-66-COOH@ PCMVIM and CNTs@PCMVIM-10).



Figure S14 (a) PXRD patterns before and after solar steam generation. (b) SEM image of UCP-10 membrane after solar steam generation.

Note: the crystal structure of UiO-66-COOH is maintained after solar steam generation and the morphology of UCP-10 membrane shows no obvious changes after solar steam generation.



Figure S15 Water evaporation rate using UCP-10 membrane for different sources of water under 1 kW/m^2 .

Table S2 Comparison of solar steam generation performance of UCP-10 membrane with some recently reported photothermal materials under 1 kW/m². This table is corresponding to Figure 5c.

Γ.	Photothermal	Evaporation	Flexible or	Stability in	Efficiency	Reference
Entry	material	rate (kg/m²/h)	non-flexible	acid or base	(%)	in SI

1	UCP-10	2.56	Flexible	Stable in acid	80.2	This work
2	Hierarchical	1.52	Flexible	/	81.2	[6]
	3D					
3	hierarchical hydrophilic carbon felt	1.56	Non- flexible	/	98.1	[7]
4	3D polyacrylamid e-radial aerogel	2	Non- flexible	/	85.7	[8]
5	Poly(anilineco- pyrrole) hollow spheres /aerogel	1.83	Non- flexible	/	82.2	[9]
6	3D graphene foam	2.4	/	/	~100%	[10]
7	Polystyrene sulfonate@HK UST- 1/SWCNT	1.38	Flexible	Stable in acid	90.8	[11]
8	Hydrophilic ultralong hydroxyapatite /CNT bilayer aerogel	1.34	Non- flexible	Stable in base	89.4	[12]
9	Surface- modified	2.6	Flexible	/	91	[13]

	hydrogel					
10	Carbon foams	1.03	Non- flexible	/	80.1	[14]
11	Carbon black- cellulose sponge system	1.12	Non- flexible	/	82.2	[15]
12	Carbon sponge	1.39	Flexible	/	90	[16]
13	Ordered PCMVIMlar array of graphene framework	2.1	Non- flexible	Stable in acid and base	95	[17]
14	MoS ₂ /C@ Polyurethane	1.95	Non- flexible	/	88	[18]
15	Solar absorber gel with dispersed AuFs	1.36	Non- flexible	/	85	[19]
16	Graphene foil supported porous graphene sponge	2.61	Non- flexible	/	91.7	[20]
17	Activated carbon fiber	1.22	Non- flexible	/	79.4	[21]
18	Composite- embedded cellulose	1.59	Flexible	/	~100	[22]

	sheets					
19	p-PEG diacrylate- PANi	1.4	Flexible	/	91.5	[23]
20	Hydrogel polyvinyl alcohol/rGO	2.5	Non- flexible	/	95	[24]
21	Hydroxyapatit e nanowires/CN T photothermal paper	1.09	Flexible	/	83.2	[25]
22	Geopolymer- biomass mesoporous carbon composite	1.58	Non- flexible	Stable in acid	84.95	[26]
23	Carbonized mushrooms	1.475	Non- flexible	/	78	[27]
24	Hierarchical graphene foam	1.4	Flexible	/	93.4	[28]
25	GO-sodium alginate-CNT	1.622	Non- flexible	/	83	[29]
26	Flexible- wood/CNT	0.99	Flexible	Stable in acid and base	65	[30]
27	Vertically aligned	1.62	Non- flexible	Stable in acid and base	86.5	[31]

	graphene					
	sheets					
	membrane					
28	rGO-400	1.14	Non-	/	89	[32]
20			flexible			
	Graphene		Non			
29	oxide/nanofibri	1.25	1N011-	/	85.6	[33]
	llated cellulose		Tlexible			



Figure S16 Water evaporation tests of UCP-10 membrane for seawater. Please note: there are some crystals on the surface of UCP-10 membrane after 2 hours under 1 sunlight irradiation (b) and the salt crystals are dissolved within 0.5 hour when the membrane was placed in dark (c).



Figure S17 Self-cleaning process of UCP-10 membrane for NaCl crystals in dark. Please note: 0.2 g of NaCl crystals were placed on top of UCP-10 membrane (Figure S17a). These crystals are completely dissolved after placing the membrane in dark for 1.5 hours (Figure S17b-d).



Figure S18 Optical images of oil/water emulsion before and after solar-thermal evaporation purification using the UCP-10 membrane.

Note: from the inserted vials, the disappearance of Tyndall effect illustrates that the oil molecules were removed clearly.

Section S4: Molecular Dynamics (MD) simulation of water transportation in UiO-66-COOH



Figure S19 Hydrogen bonds distribution in bulk water. Please note: the length of hydrogen bond is 1.5-2.5 Å nm according to statistical result of Molecular Dynamic (MD) method.



Figure S20 Dark evaporation rate of bulk water, and water in UCP-10, UP, CB-10 (UP and CB-10 are the abbreviation of UiO-66-COOH@PCMVIM, and CNTs@PCMVIM-10 membranes).

Please note: 1) the enthalpy values are calculated by the formula of $E_{equ} = E_w * (R_w/R_m)$ according to dark evaporation rate, where E_{equ} is the evaporation enthalpy of water in UCP-10 membrane, E_w is the enthalpy of bulk water, R_m is the water evaporation rate of the UCP-10 in dark, and R_w is the dark evaporation rate of bulk water. Based on this formula, the enthalpy value of water in UCP-10 is 1.329 kJ/g; 2) the enthalpy values were calculated by integration for endothermic peak of bulk water and the water in UCP-10 based on DSC result, and the enthalpy values were 2.407 kJ/g and 1.309 kJ/g respectively.

Section S5: Interfacial solar steam generation of UCP-*x* membranes with different contents of CNTs



Figure S21 Surface temperature of UCP-*x* membranes with a different amount of CNTs in (a) wetting or (b) dry condition under 1 kW/m^2 .

Note: with the increasing amount of CNTs, the temperature of UCP-x membranes increases in both dry and wet states.



Figure S22 Effect of carbon nanotubes content in UCP-x membranes in water evaporation rate under 1 kW/m² irradiation.

Please note: the water evaporation rate is 2.56 kg/m²/h when CNTs content is 10 wt%, above which the evaporation rate starts to decrease. A part of UiO-66-COOH particles in unit area is replaced by CNTs and the water channels of UiO-66-COOH was partly reduced. So, the formation of water clusters and transportation of water nanofluidics are inhibited partly and further decrease the water evaporation.

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