

**Moisture-indicated Cellulose Aerogels for Multiple Atmospheric Water  
Harvesting Cycles Driven by Solar Energy**

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## **S1 Supplemental methods and characterization**

### **S1.1 Preparation of microfibrillated cellulose (MFC)**

Kapok fibers were smashed by a pulverizer. Lignin was then removed by the acidified  $\text{NaClO}_2$  method. Concretely, Kapok fibers were treated with a fixed proportion of  $\text{CH}_3\text{COOH}$  and  $\text{NaClO}_2$  in a water bath at  $75\text{ }^\circ\text{C}$  for 2 h until the kapok fibers turn white. Then hemicellulose was removed with 4 wt%  $\text{NaOH}$  at  $90\text{ }^\circ\text{C}$  for 4 h. Finally, the sample was washed with distilled water to neutrality by filtration to obtain kapok fiber-based cellulose. The obtained kapok fiber-based cellulose was formulated into a 0.8% suspension and then ultrasonicated (JY98- IID, Ningbo Scientz Biotechnology Co., Ltd., China) under 1200 W for 25 min in an ice/water bath to obtain microfibrillated cellulose.

### **S1.2 Preparation of LiCl-decorated poly-pyrrole (Cl-ppy)**

2 ml of pyrrole was dispersed in 20 ml of DI water by sonication. Then 1.645 g of  $(\text{NH}_4)_2\text{S}_2\text{O}_8$  and 3 g of anhydrous  $\text{LiCl}$  were respectively dissolved in 20 ml of distilled water. Under stirring, the  $(\text{NH}_4)_2\text{S}_2\text{O}_8$  aqueous solution and the  $\text{LiCl}$  solution were added dropwise to the pyrrole suspension. The product was washed repeatedly until it is neutral. Finally, the purified Cl-ppy was homogeneously dispersed in DI water by sonication to form Cl-ppy solution.

### **S1.3 Preparation of ethanolamine-decorated $\text{CoCl}_2$ (E- $\text{CoCl}_2$ )**

First, 3.82 g of  $\text{CoCl}_2$  was dissolved in 20 ml of ethanol. Then, 480  $\mu\text{l}$  of ethanolamine was added into it under stirring and stirred for 30 minutes. Finally, E- $\text{CoCl}_2$  was obtained after drying at  $80\text{ }^\circ\text{C}$ .

### **S1.4 Characterization**

Scanning electron microscope (SEM) images were acquired on a Quanta 200 (American FEI Company).-The X-ray photoelectron spectroscopy (XPS) analysis was conducted on a XPS spectrometer (250 Xi, Thermo Fisher Scientific, American). Fourier transform infrared spectroscopy (FTIR) was carried out using an infrared spectrometer (TENSOR 27, Bruker, Germany). The rheological experiments were performed by a rheometer (DHR-2, Waters, America) using the parallel plate on a Peltier plate in a frequency sweep mode. The contact angle was measured using Kruss

Drop Shape Analyzer (Kruss GmbH DSA255). The UV-vis-NIR spectra were obtained on a UV-3600Plus spectrophotometer (Shimadzu, Japan). The adsorption isotherms are obtained from a Vapor Sorption Analyzer (AquaLab) at 25 °C. A CEL-S500/350 xenon lamp (China Education Au-light Co. Ltd., China) was used to simulate sunlight. The light intensity was determined using a CEL-FZ-A light power meter (China Education Au-light Co. Ltd., China). The temperature of surface of specimens was measured by an infrared (IR) thermometer (FLIR A35).

## **S2 XPS analysis**

From CA to SMCA, E-CoCl<sub>2</sub> and Cl-ppy were doped into matrix in succession. The binding energy values for O 1s have negative shifts continuously. It may attribute to the fact that O constantly attracting the electrons of Cl and N of E-CoCl<sub>2</sub> or Cl-ppy, resulting in an increase in its electron cloud density. In the N 1s spectrum in Figure 2b, the peak at 401.4 eV corresponds to -NH<sup>+</sup> (N-Q), whereas the peak at 399.2 eV is assigned to N-H/C-N (N-5).<sup>[1]</sup> After E-CoCl<sub>2</sub> is incorporated into the matrix, N has a positive shift because the electron pair effect of N or O and central cobalt ions increase the binding energy values for N.<sup>[2]</sup> After doping Cl-ppy, the intensity of N weakens while the intensity of N-5 increases. It can be ascribed to N of MCA takes away the electrons from Cl of Cl-ppy and meanwhile Cl-ppy brings in N-C.

**Table S1** The equilibrium water sorption capacity of E-CoCl<sub>2</sub>, MCA, SMCA and CA at 25 °C under different RH conditions.

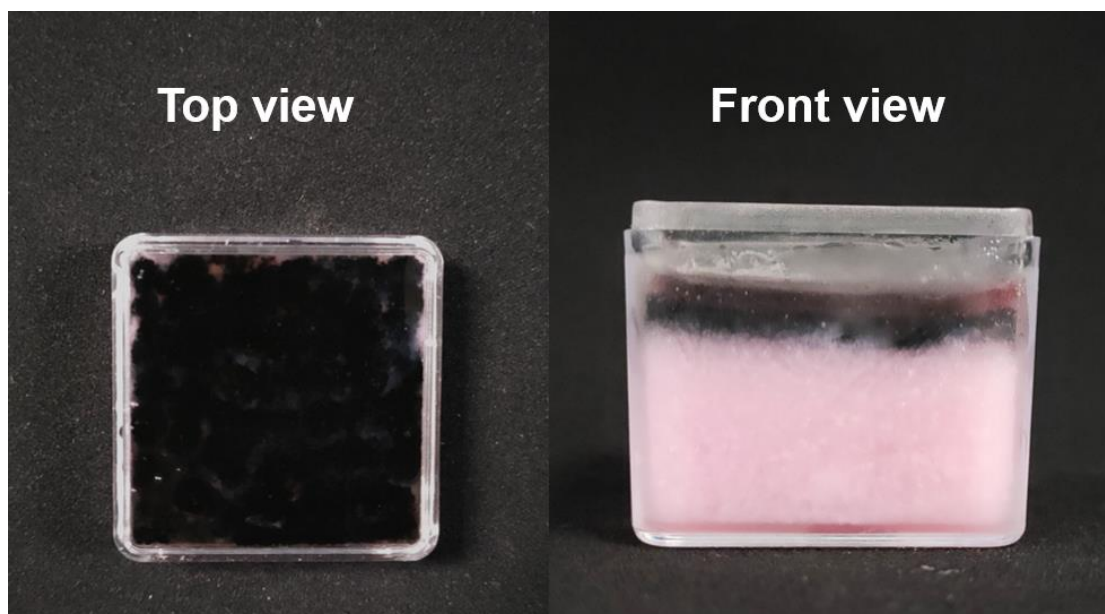
	RH 25%	RH 45%	RH 65%	RH 85%
E-CoCl <sub>2</sub>	0.076 g/g	0.52 g/g	0.94 g/g	1.3 g/g
MCA	0.49 g/g	0.69 g/g	1.12 g/g	2.52 g/g
SMCA	0.49 g/g	0.69 g/g	1.07 g/g	2.38 g/g
CA	0.048 g/g	0.033 g/g	0.083 g/g	0.26 g/g

**Table S2** The consecutive test of SMCA.

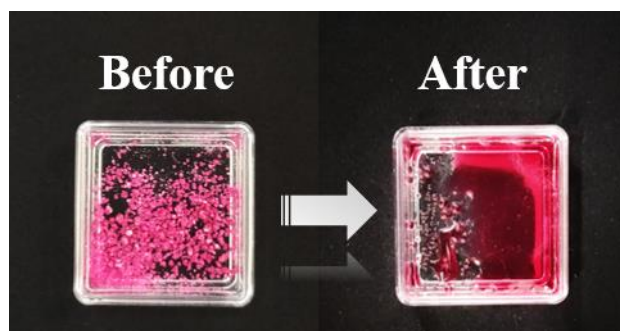
Date	weather	The max temperature, °C	RH at 12:00, %	The mass of collected water for the cycle 1, g	The mass of collected water for the cycle 2, g	The mass of collected water for the cycle 3, g	The total mass of collected water, g
July 22, 2021	Cloudy	32	68	9.73	5.58	2.48	17.79
July 23, 2021	Cloudy	33	65	10.04	5.72	2.57	18.33
July 24, 2021	Sunny	34	51	9.67	5.92	2.95	18.54
July 25, 2021	Cloudy	32	59	9.31	4.78	2.68	16.77
July 26, 2021	Cloudy	33	70	11.02	5.58	2.52	19.12



**Figure S1** Digital photo of device of indoor solar-powered vapor evaporation.

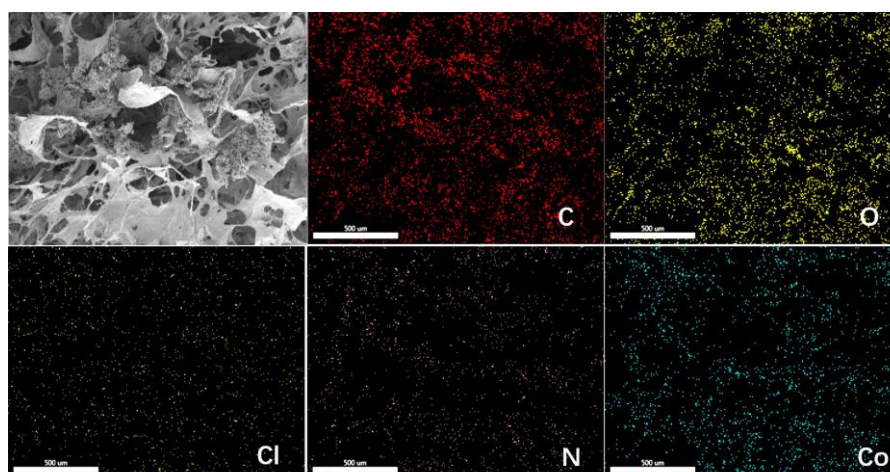


**Figure S2** The mixture of MFC and E-CoCl<sub>2</sub> can support Cl-ppy suspension very well.

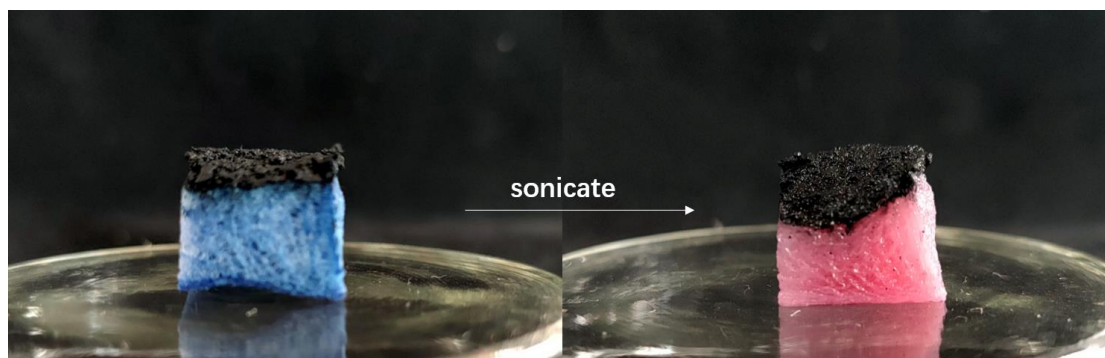


**Figure S3** Photograph of pure  $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$  changing to liquid after capturing moisture at 65%.

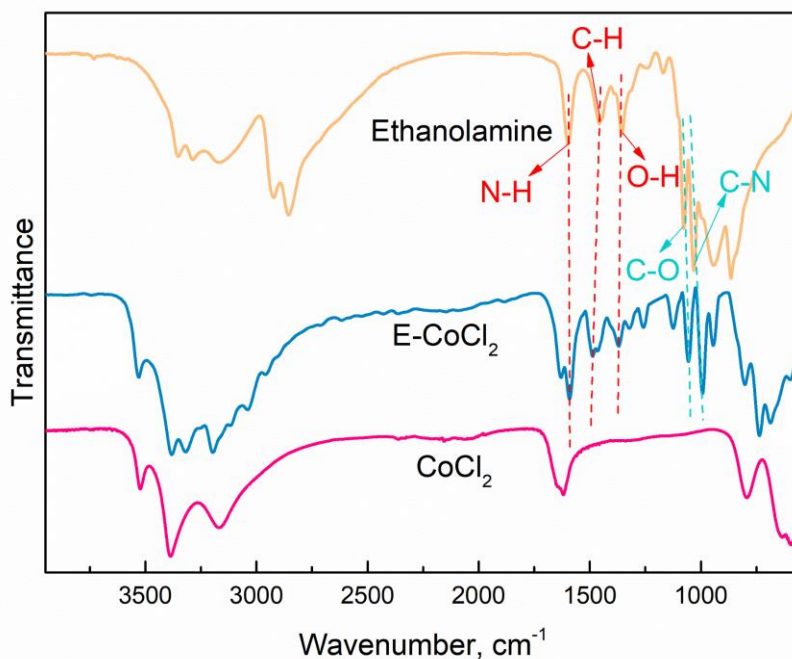
Pure  $\text{CoCl}_2$  becomes liquid after absorbing moisture. The hydrophilicity and porous structure of MCA-x can compound it easily, resulting to a perfect solution to the problem of leakage.



**Figure S4** Mapping of SMCA.



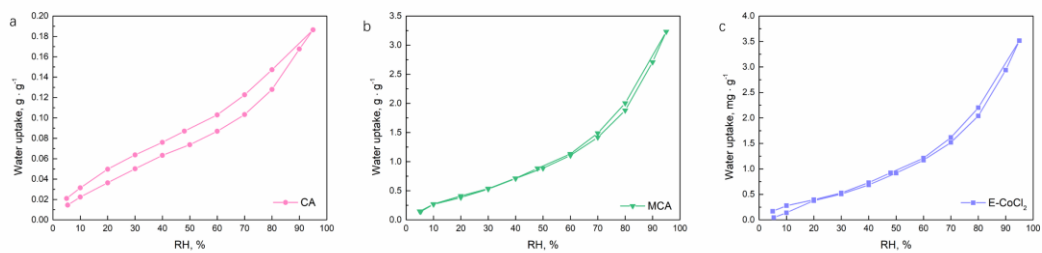
**Figure S5** The bilayer structure maintained well after sonicating in water for 10 mins verifying the strong entanglement between Cl-ppy and cellulose aerogel.



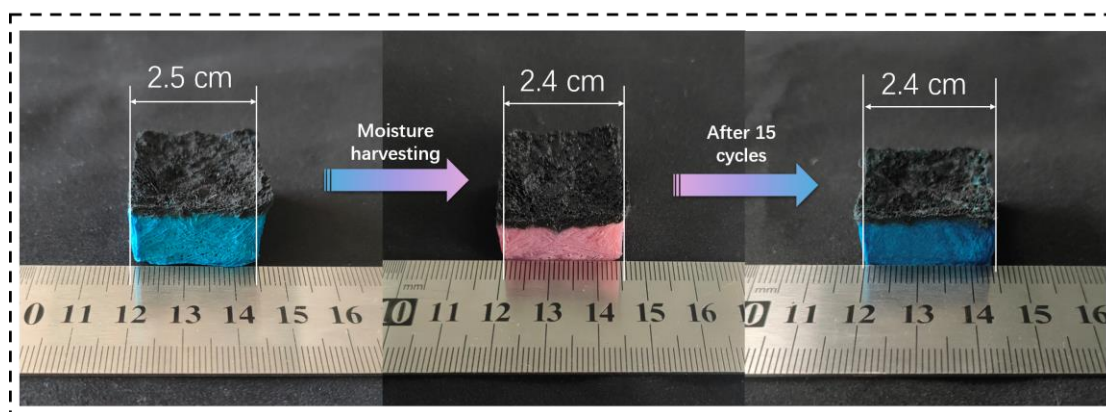
**Figure S6** FTIR spectra of ethanolamine,  $\text{CoCl}_2$  and  $\text{E-CoCl}_2$ .

It is observed that ethanolamine has two sharp bands of  $\nu(\text{C-O})$  at  $1074\text{ cm}^{-1}$  and  $\nu(\text{C-N})$  at  $1028\text{ cm}^{-1}$ , which have blue shifts compared to that of  $\text{N-CoCl}_2$ .<sup>[3]</sup> Besides, the  $\delta(\text{N-H})$  band at  $1598\text{ cm}^{-1}$ ,  $\delta(\text{C-H})$  band at  $1458\text{ cm}^{-1}$  and  $\delta(\text{O-H})$  band at  $1359\text{ cm}^{-1}$  of the free ethanolamine show red shifts (at  $1633\text{ cm}^{-1}$ ,  $1489\text{ cm}^{-1}$ , and  $1371\text{ cm}^{-1}$ , respectively) upon complexation.<sup>[2]</sup> These results confirm the coordination of cobalt with the hydroxy O atom and the amino N atom of the ethanolamine

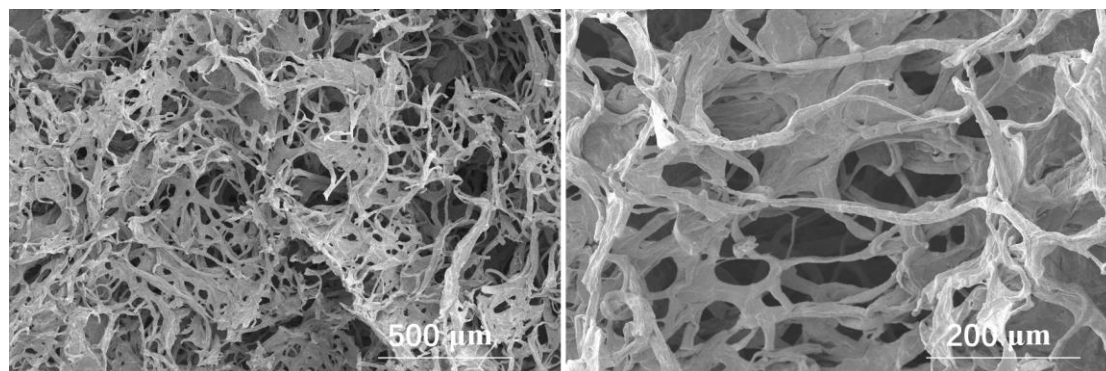




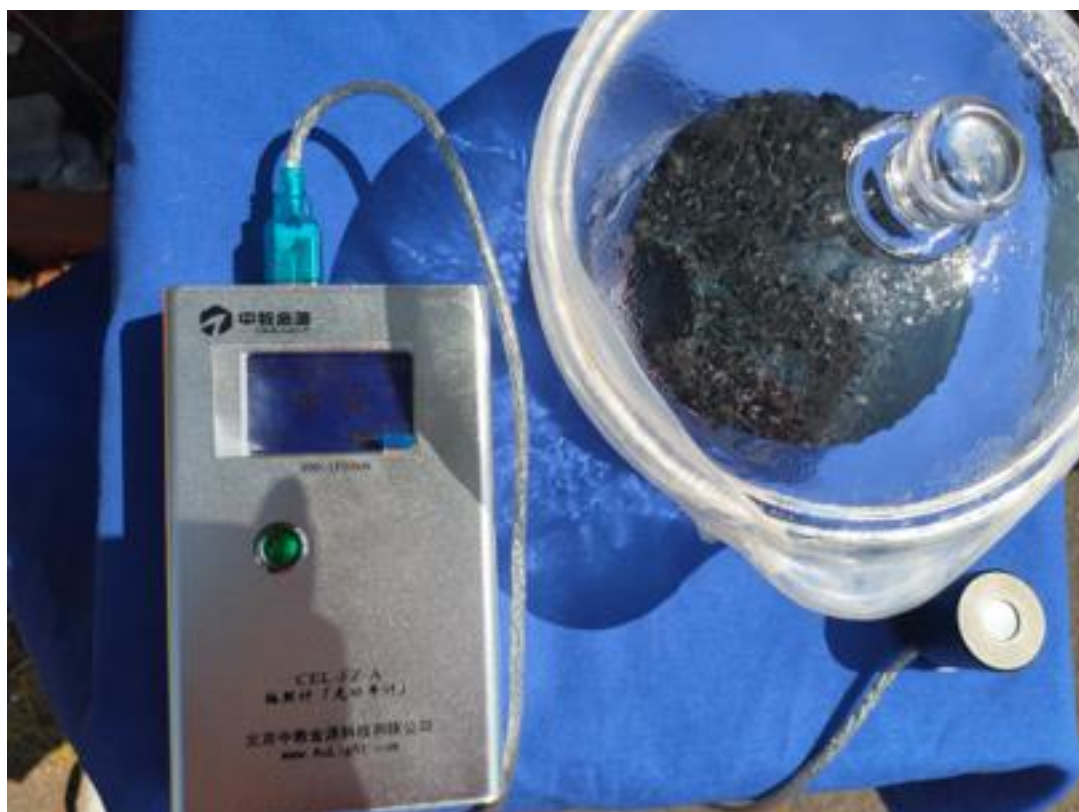
**Figure S7** Water sorption/desorption isotherms of (a) CA, (b) MCA and (c) E-CoCl<sub>2</sub> at 25 °C.



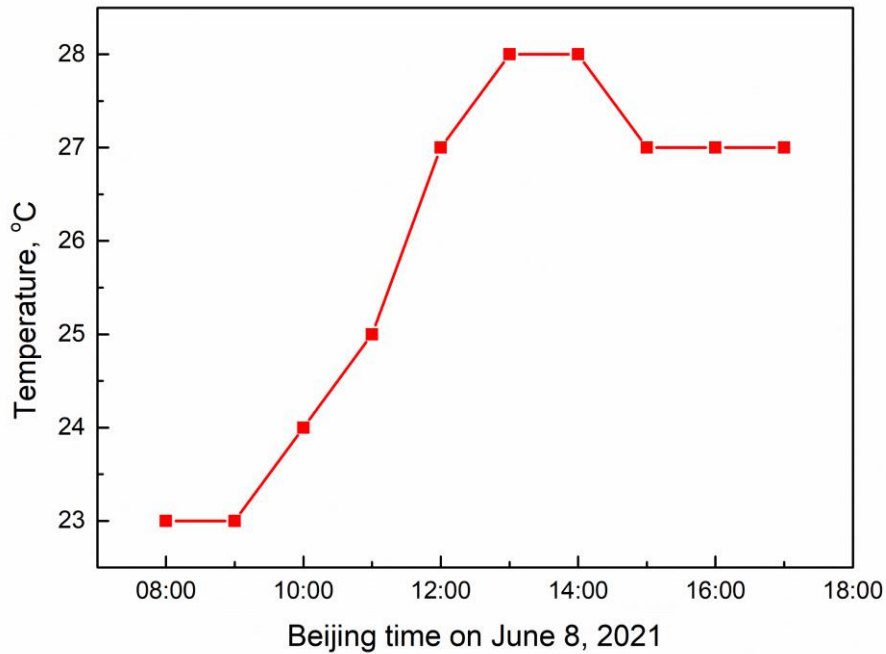
**Figure S8** Digital photos of size change of SMCA.



**Figure S9** SEM images of SMCA after 15 sorption/desorption cycles.



**Figure S10** Digital photos of device of outdoor experiments. (The above photo is the desorption process in the sun, and the bottom photo is the sorption process in the shade)



**Figure S11** Detailed temperature variation of outdoor experiment:

**Details of outdoor experiment:**

The first water-harvesting process was from 20:00 June 7, 2021 to 8:00, June 8, 2021. The first sunlight power desorption process was started on June 8, 2021 at 8:00, and around 10.88 g of water was collected within 106 min (8:00-9:46). The Second water-harvesting process was start from 9:46 June 8, 2021 to 11:50 June 8, 2021. The second sunlight power desorption process was started on June 8, 2021 at 11:55, and around 4.87 g of water was collected within 85 min (11:55-13:20). The third water-harvesting process was start from 13:25 June 8, 2021 to 15:25 June 8, 2021. The third sunlight power desorption process was started on June 8, 2021 at 15:25, and around 2.32 g of water was collected within 95 min (15:25-17:00).

## Reference

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