

Supporting Information

Proton Conducting Metal-Organic Frameworks with Light Response for the Multistate Logic Gates

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1. Synthesis of Sulfated Spiropyran

The SSP was synthesized according to the already reported literature.¹ Briefly, 2,3,3-trimethylindolenine (1.6 mL, 10 mmol) was mixed with 20 mL CH₃I (12 mmol) and stirred at 60 °C for 2 h. White solid precipitated were obtained from the solution after cooling to the room temperature. 1,2,3,3-Tetramethyl-3H-indolium iodide (602.3 mg, 2 mmol) was obtained by washing the residue with ethanol and diethyl ether for several times. The precipitates were dissolved in 10 mL methanol, and reflux at 80 °C. Then piperidine (200 µL, 2 mmol) and another solution of sodium salicylaldehyde-5-sulfonate (448.3 mg, 2 mmol) in 10 mL methanol were mixed into the reaction vessel successively and refluxed for 4 h with stirring. After cooling to room temperature, the solvent was evaporated and the the residue was dissolved in chloroform and recrystallized from benzene. Sulfated spiropyran (458 mg) was obtained after washed with benzene several times.

Table S1. Detailed Composition of the Prepared Membranes.

Sample Codes	CuHNs (mL)	PSS (mL)	SSP (mL)	TCPP (mL)
C ₀₋₀	30	0	0	4
C ₁₋₀	30	1	0	4
C ₁₋₁	30	1	1	4
C ₁₋₂	30	1	2	4
C ₂₋₀	30	2	0	4
C ₂₋₁	30	2	1	4
C ₂₋₂	30	2	2	4

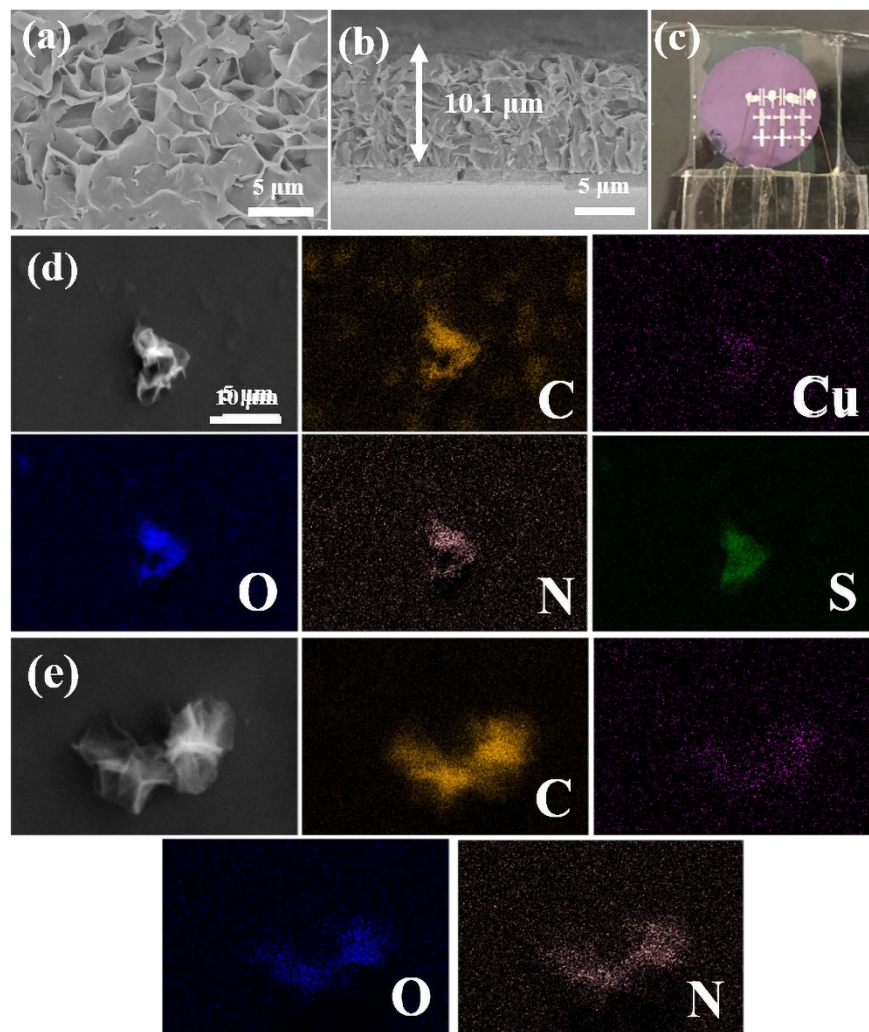


Fig. S1. SEM surface and cross-sectional images of (a, b) C₁₋₀; (c) Photograph of PSS-SSP@Cu-TCPP membrane with attached silver electrodes connected with copper wires; EDX elemental mapping of (d) C₂₋₁ and (e) C₀₋₀.

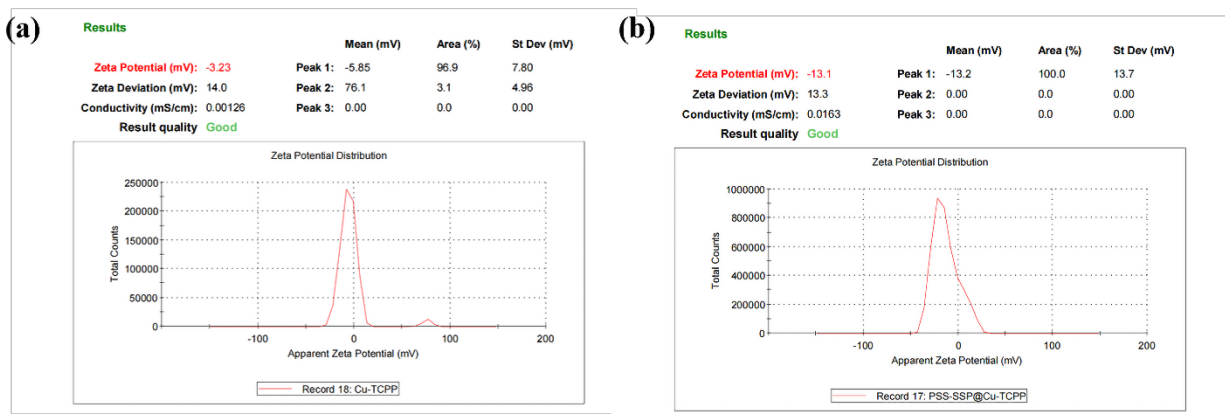


Fig. S2. Zeta potential of (a) Cu-TCPP (b) PSS-SSP@Cu-TCPP nanosheets ethanolic dispersion.

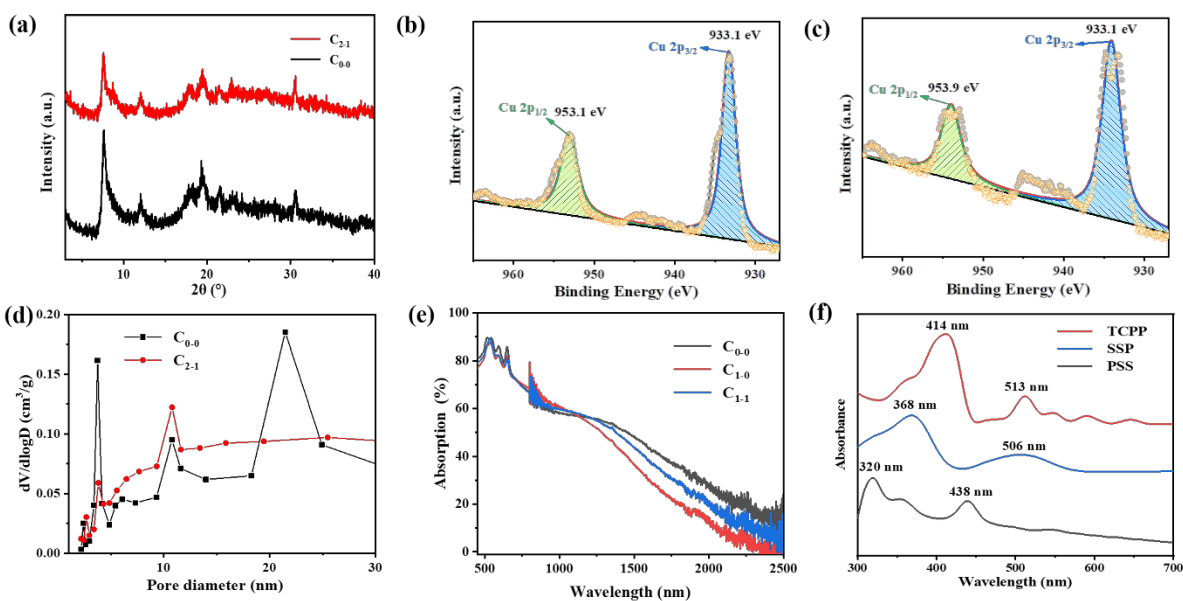


Fig. S3. (a) PXRD patterns of C_{0-0} and C_{2-1} ; XPS spectra with fitted Cu peaks of (b) C_{0-0} ; (c) C_{1-1} ; (d) pore size distribution of C_{0-0} and C_{2-1} ; (e) UV-Vis-NIR absorption spectra of C_{0-0} , C_{1-0} , C_{1-1} and (f) UV-Vis absorption spectra of TCPP, PSS and SSP.

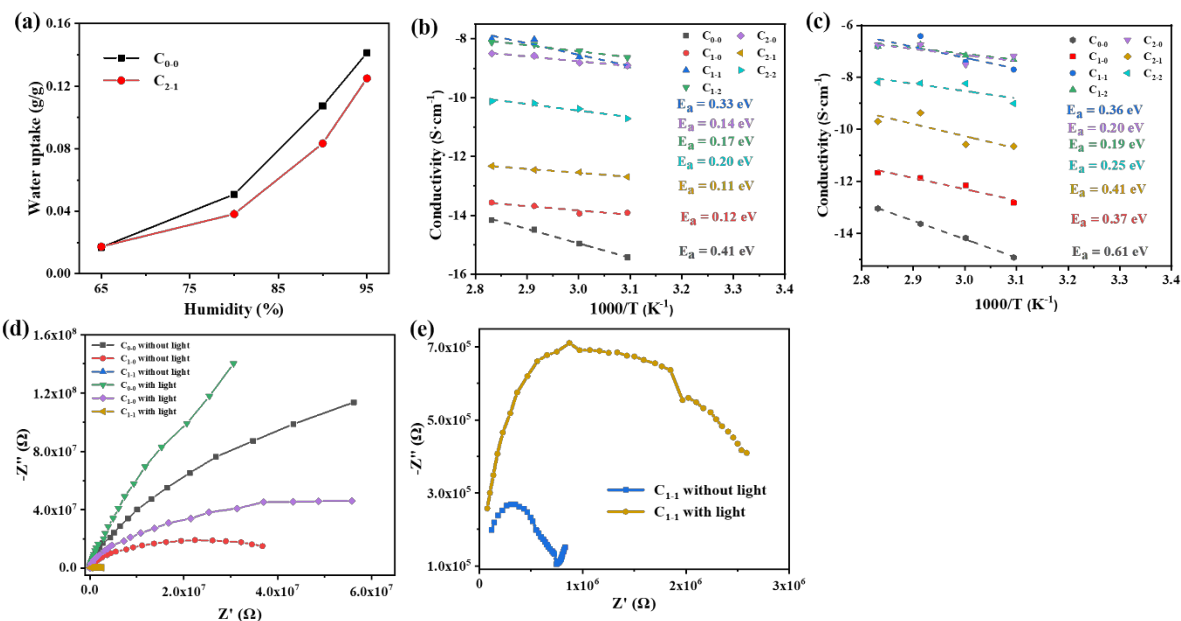


Fig. S4. (a) Water uptake curve with different humidity of C_{0-0} and C_{2-1} ; Arrhenius plots and corresponding activation energies of Cu-TCPP composite membranes at 95% RH (b) With light using $100 \text{ mW} \cdot \text{cm}^{-2}$ Xe lamp; (c) Without light; (d, e) Nyquist plots of C_{0-0} , C_{1-0} , and C_{1-1} in the dark and $100 \text{ mW} \cdot \text{cm}^{-2}$ light at $60 \text{ }^\circ\text{C}$ and 95% RH.

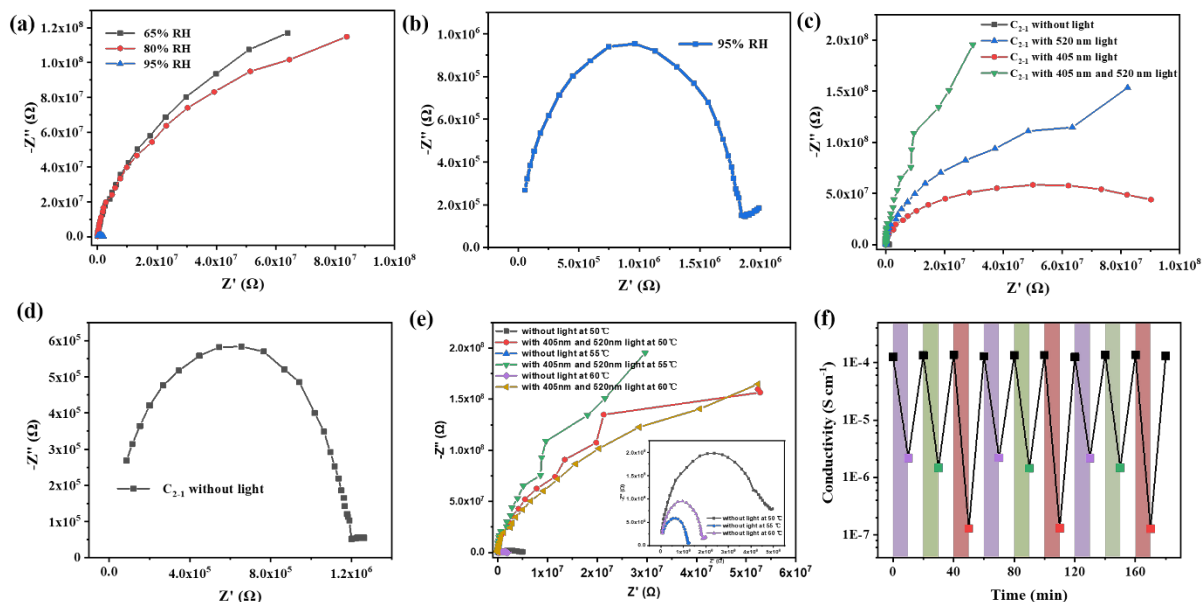


Fig. S5. Nyquist plots of (a, b) C_{2-1} under different relative humidity in the dark at 80 °C; (c, d) C_{2-1} under different light condition at 60 °C; (e) C_{2-1} under different temperature in the dark and light at 95% RH; (f) Proton conductivity of C_{2-1} in the dark (black point) and under different light condition (black point: 405 nm light with $400\ mW\cdot cm^{-2}$; green point: 520 nm light with $400\ mW\cdot cm^{-2}$; red point: 405 nm light with $400\ mW\cdot cm^{-2}$ and 520 nm light with $200\ mW\cdot cm^{-2}$ simultaneously) at 55 °C and 95% RH.

References:

1. H. Liang, Y. Guo, Y. Shi, X. Peng, B. Liang and B. Chen, *Angew. Chem.-Int. Edit.*, 2020, **59**, 7732.