Supplementary Information

Acceptor Engineering Boosting Piezoelectricity in Conjugated Microporous Polymers for Piezo-photocatalytic Uranium Separation

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S1. Materials

All chemicals utilized in this study were of analytical grade and used as received without further purification. The monomers 3,6-dibromocarbazole, 1,4-benzenediboronic acid bis(pinacol) ester, 2,5-dibromobenzonitrile, and 2,5-dibromoterephthalonitrile were acquired from Jilin Chinese Academy of Sciences-Yanshen Technology Co., Ltd. All other reagents were supplied by Adamas-Beta. Deionized water (18 MΩ·cm) was employed for all aqueous solutions throughout the experiments.

S2. Characterization methods

Powder X-ray diffraction (PXRD) patterns were recorded on a Bruker D8 Advance diffractometer to analyze the crystallinity. Chemical structures were verified using Fourier-transform infrared spectroscopy (FT-IR, Bruker Tensor II) and solid-state ^{13}C NMR spectroscopy (Avance III HD 500 MHz). Surface elemental compositions were determined via X-ray photoelectron spectroscopy (XPS, AXIS Supra+). Porosity parameters, including specific surface area and pore size distribution, were evaluated using nitrogen adsorption-desorption isotherms at 77 K on a Brunauer-Emmett-Teller (BET) analyzer (BSD-660M) The Electron paramagnetic resonance spectroscopy spectra were measured on CIQTEK EPR200-Plus with continues-wave X band frequency. The 5,5-dimethyl-l-pyrroline N-oxide (DMPO) was employed as a probe under $\lambda > 420$ nm light irradiation. Piezoelectric properties were characterized using a commercial Piezoresponse Force Microscopy (PFM) system (Bruker Dimension Icon). Vertical PFM (V-PFM) measurements were conducted by applying an AC voltage to a conductive AFM tip, with the bottom electrode grounded. For domain imaging and switching spectroscopy, a drive amplitude of 10 V AC was employed.

S3 Fabrication of piezoelectric nanogenerator (PENG)

PENG devices were fabricated following established protocols [1]. Briefly, conductive electrodes were prepared by depositing silver layers (0.7×0.7 cm^2) onto two silicon substrates (1.2×1.2 cm^2). Copper wires were attached to these electrodes via soldering for electrical connection. To construct the device, Kapton tape was first applied to the perimeter of the bottom electrode to create a mold. A polydimethylsiloxane (PDMS) film (0.40 mm thick) with a central cutout matching the electrode area (0.5×0.5 cm^2) was then positioned on the substrate. The synthesized CMP powder (5 mg) was tightly packed into this cavity. The top electrode was subsequently aligned and secured with tape. Finally, the entire sandwich structure was encapsulated using Kapton tape to ensure mechanical stability. All electrical output measurements were performed under a constant applied force.

S4 DFT calculations

The calculations based on the density function theory (DFT) and time-dependent-density functional theory (TD-DFT) are carried out using the Gaussian package. In DFT calculations, the reaction mechanism, used to identify the energy paths, is performed on the B3LYP/Def2-SVP level. The vibrational frequency analyses were used to verify the minimum character of the configurations. In TD-DFT calculations, the excited states of optimized geometry are performed on the CAM-B3LYP/Def2-SVP level. The electron distributions at these exciting statuses were obtained with Multiwfn and Vesta. For all the calculations, the dispersion force contributed to energy is considered using the Grimme D3 method, and the solvent effect was described by a conductor-like polarizable continuum model (C-PCM).

S5. Figures

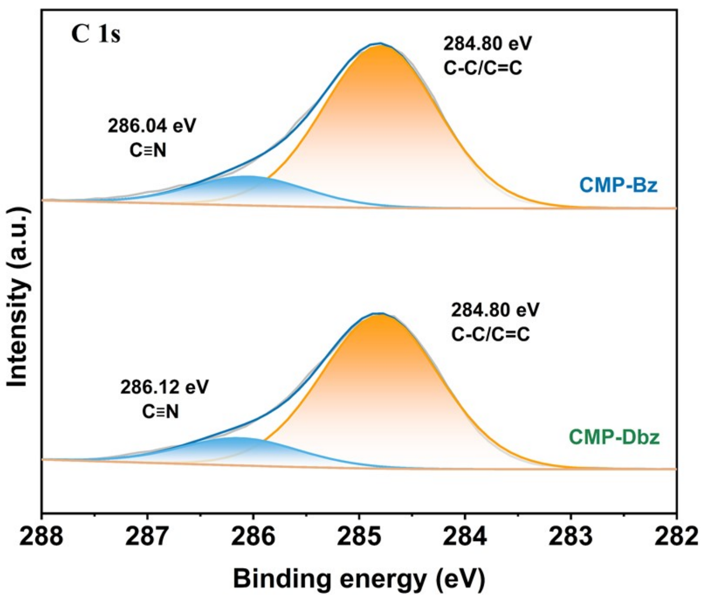

Fig.S1. High-resolution XPS spectra of C 1s for the synthesized CMPs.

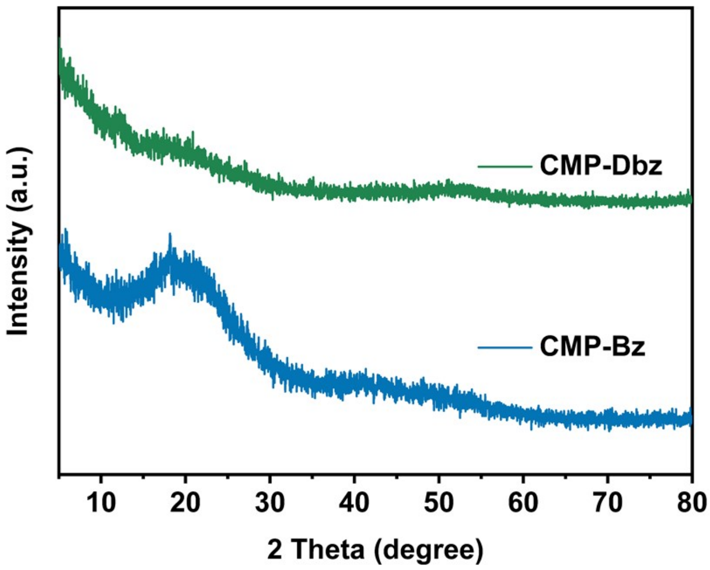

Fig.S2. The PXRD of CMP-Bz and CMP-Dbz.

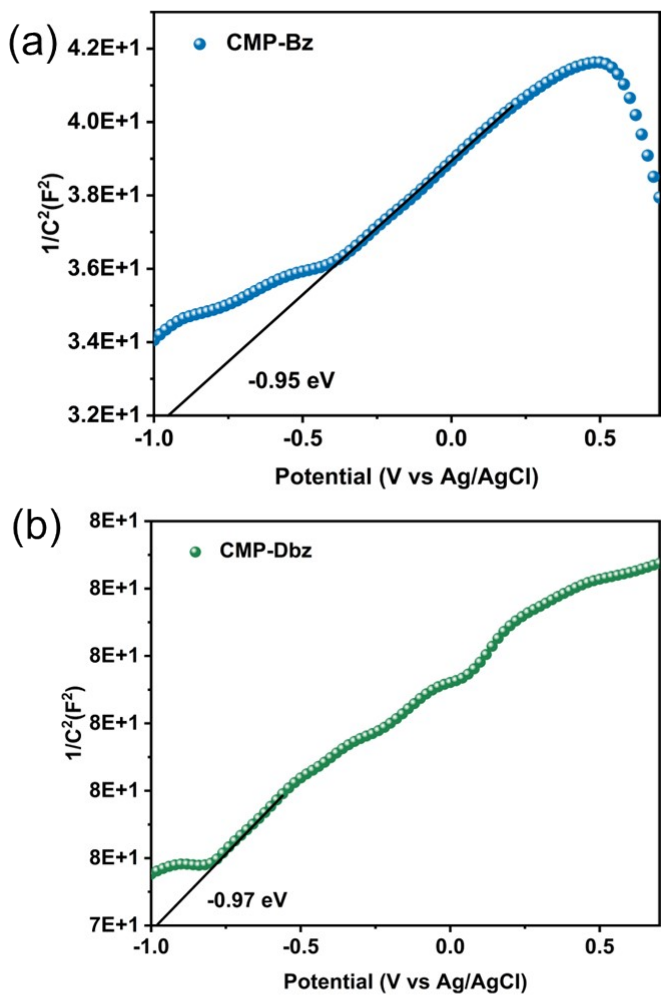

Fig.S3. Mott-Schottky curves for CMP-Bz and CMP-Dbz.

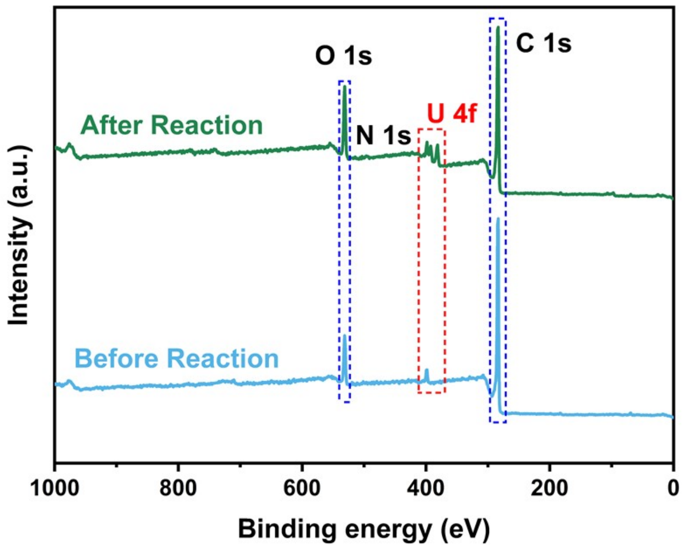

Fig.S4. XPS survey of CMP-Dbz before and after reaction.

Reference

[1] Z. Li, Z. Dong, Z. Zhang, B. Wei, C. Meng, W. Zhai, Y. Wang, X. Cao, B. Han, Y. Liu. Covalent Organic Frameworks for Boosting H_2O_2 Photosynthesis via the Synergy of Multiple Charge Transfer Channels and Polarized Field. *Angew. Chem. Int. Ed.*, 2025. 64: e202420218.