RESEARCH ARTICLE

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Innovative Conversion Strategy for Wastewater with One-Pot Uranium Extraction and Valuable Chemical Production by a Smart COF Photocatalyst

*Liecheng Guo, Yuting Yang, Lele Gong, Qingyun Zhang, Yuxin Liao, Rong Hua, Zhiwu Yu, and Feng Luo**

L. Guo, Q. Zhang, Y. Liao, R. Hua, F. Luo
School of Chemistry and Materials Science
School of Nuclear Science and Engineering
East China University of Technology
Nanchang 330013, China
E-mail: luofengaiqi@ecut.edu.cn

Y. Yang
College of Chemistry and Environmental Science
Qujing Normal University
Qujing 655011, China

L. Gong
State Key Laboratory of NBC Protection for Civilian
Beijing 100191, China

Z. Yu
High Magnetic Field Laboratory
Chinese Academy of Sciences
Hefei 230031, China

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Background

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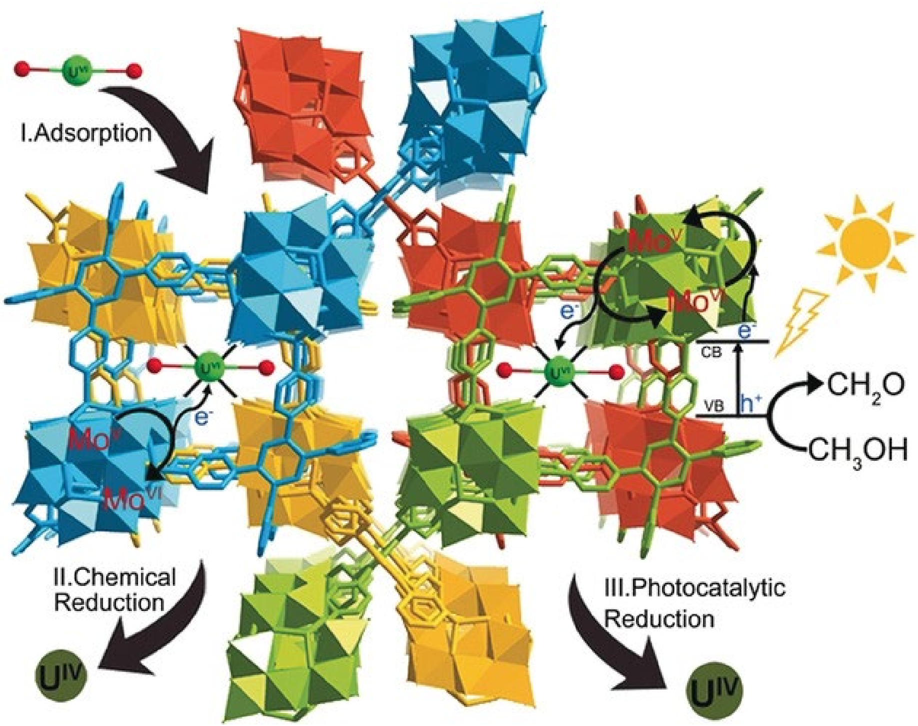
Three Mechanisms in One Material: Uranium Capture by a Polyoxometalate–Organic Framework through Combined Complexation, Chemical Reduction, and Photocatalytic Reduction

*Hailong Zhang, Wei Liu, Ao Li, Duo Zhang, Xiaoyan Li, Fuwan Zhai, Lanhua Chen, Long Chen, Yanlong Wang, and Shuao Wang**



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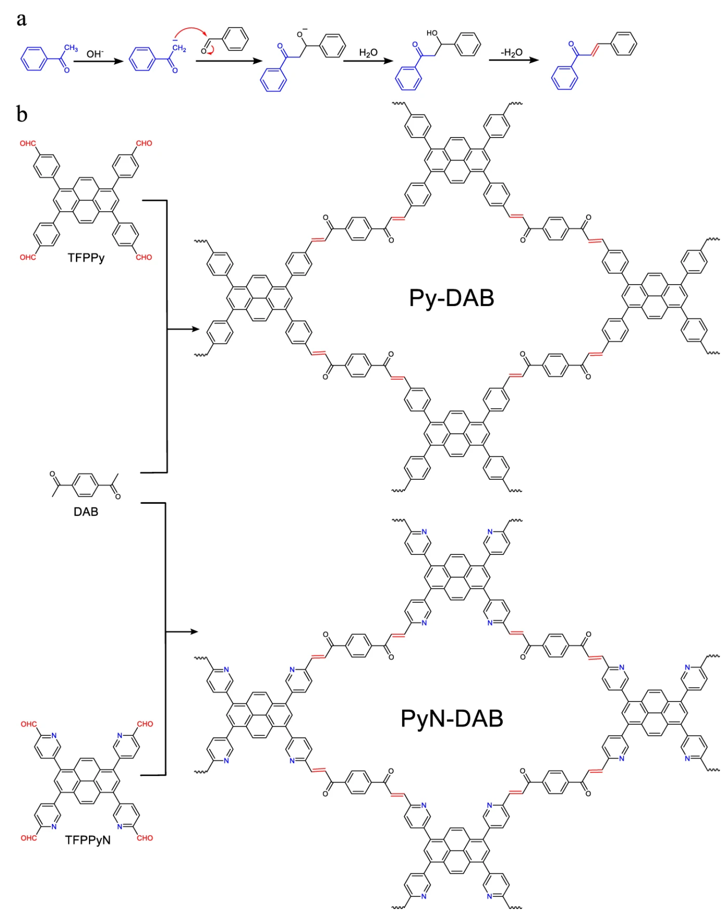
Synthesis of propenone-linked covalent organic frameworks via Claisen-Schmidt reaction for photocatalytic removal of uranium



Introduction

- This study presents an innovative conversion strategy using a **photoresponsive covalent organic framework (COF) photocatalyst** to achieve simultaneous uranium extraction and wastewater transformation into valuable hydrogen peroxide (H_2O_2).

- This concept utilizes an **azobenzene-functionalized covalent organic framework (COF)** with **photoresponsive trans-to-cis isomerization**, enhancing uranium extraction and conversion efficiency under **UV irradiation** compared to visible light.

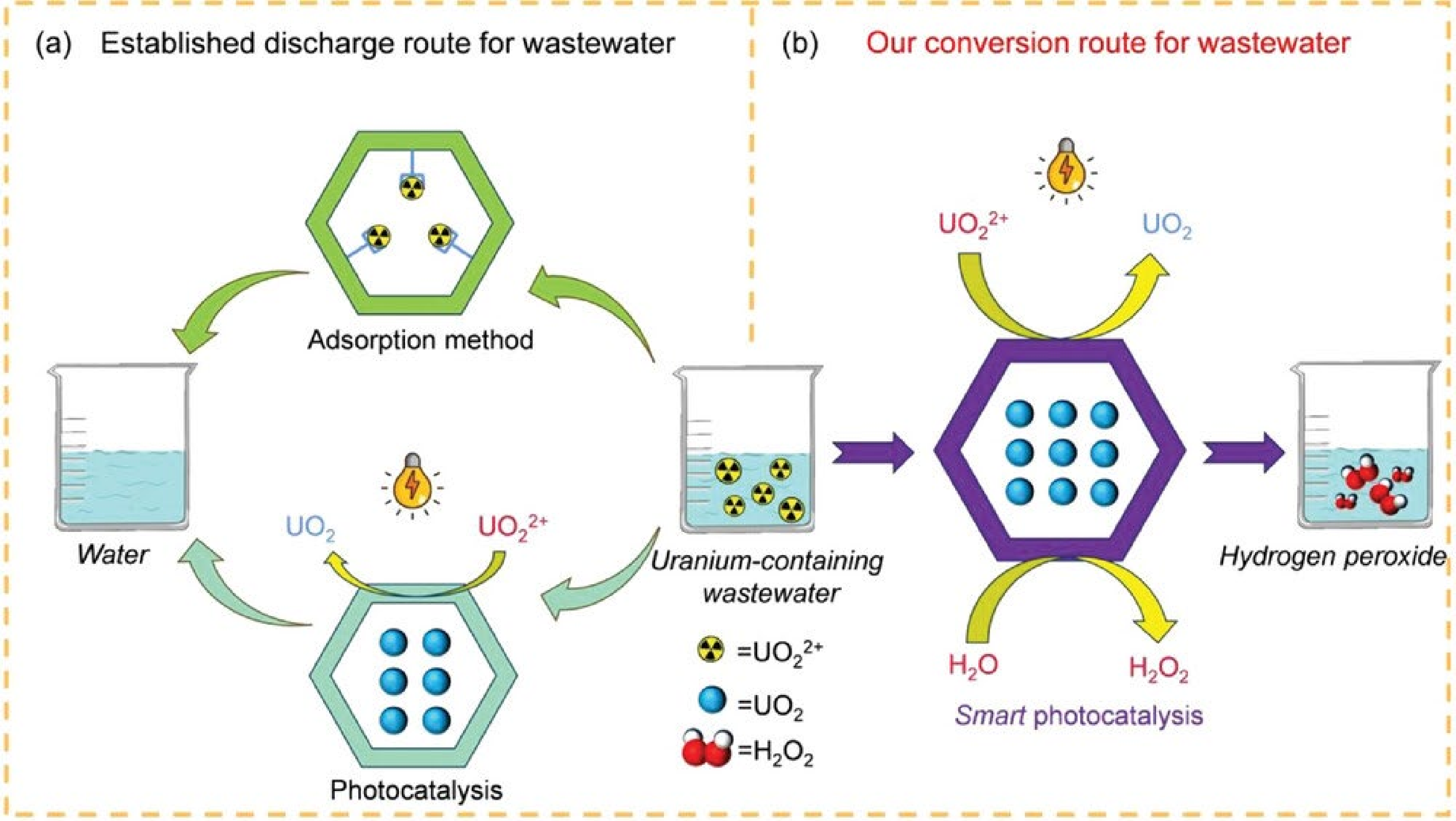
- In real wastewater, the material achieves **100% selective uranium extraction** and generates **H_2O_2 at 1872.3 μmol $g^{-1}h^{-1}$**.

- The mechanism involves a unique **photocatalytic coupling** of the **uranium reduction reaction (URR)** for uranium recovery and the **water oxidation reaction (WOR)** for converting wastewater into valuable **H_2O_2.**

Why this paper?

- This study presents a novel dual-function strategy that not only achieves efficient uranium extraction from wastewater but also converts it into valuable hydrogen peroxide (H_2O_2).

- Such photo-switchable photocatalytic phenomenon is rare to observed in previous studies.



Scheme 1. a) Schematic description of the established discharge route for uranium extraction from wastewater, including both adsorption and photocatalysis
methods. b) Schematic description of our smart photocatalysis method for uranium extraction from wastewater in a distinct conversion route.

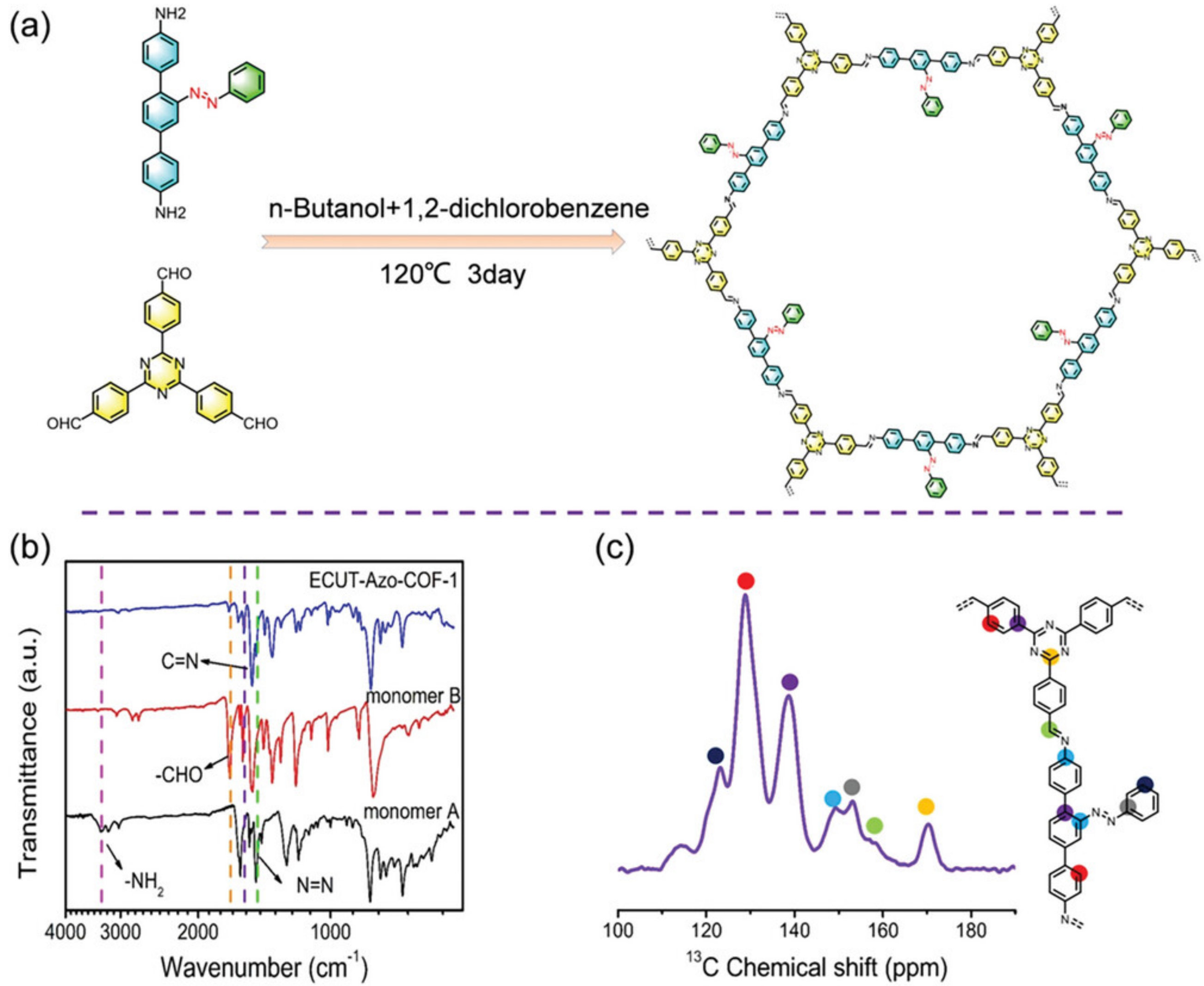


Figure 1. a) Synthesis route of ECUT-Azo-COF-1. c) IR of ECUT-Azo-COF-1. c) CP-MAS 13C NMR of ECUT-Azo-COF-1 with the assignment.

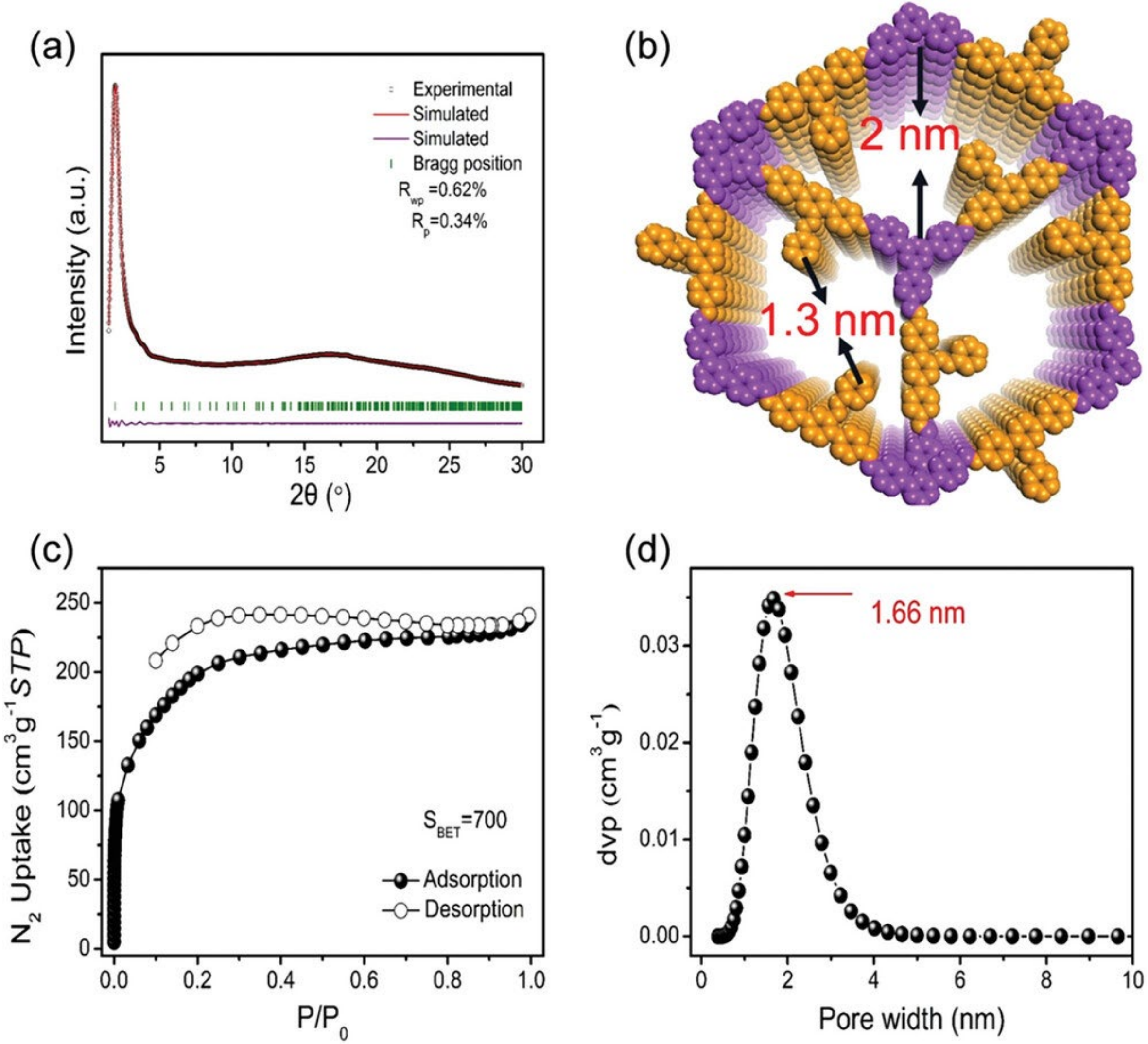


Figure 2. a) Experimental PXRD patterns of ECUT-Azo-COF-1 with corresponding Pawley refinement (red) and Bragg positions (green), showing good fit to the experimental data (black) with minimal differences (purple). b) View of the structure ECUT-Azo-COF-1 in a slipped stacking fashion. c) N_2 adsorption isotherm at 77 K. d) The pore size distribution.

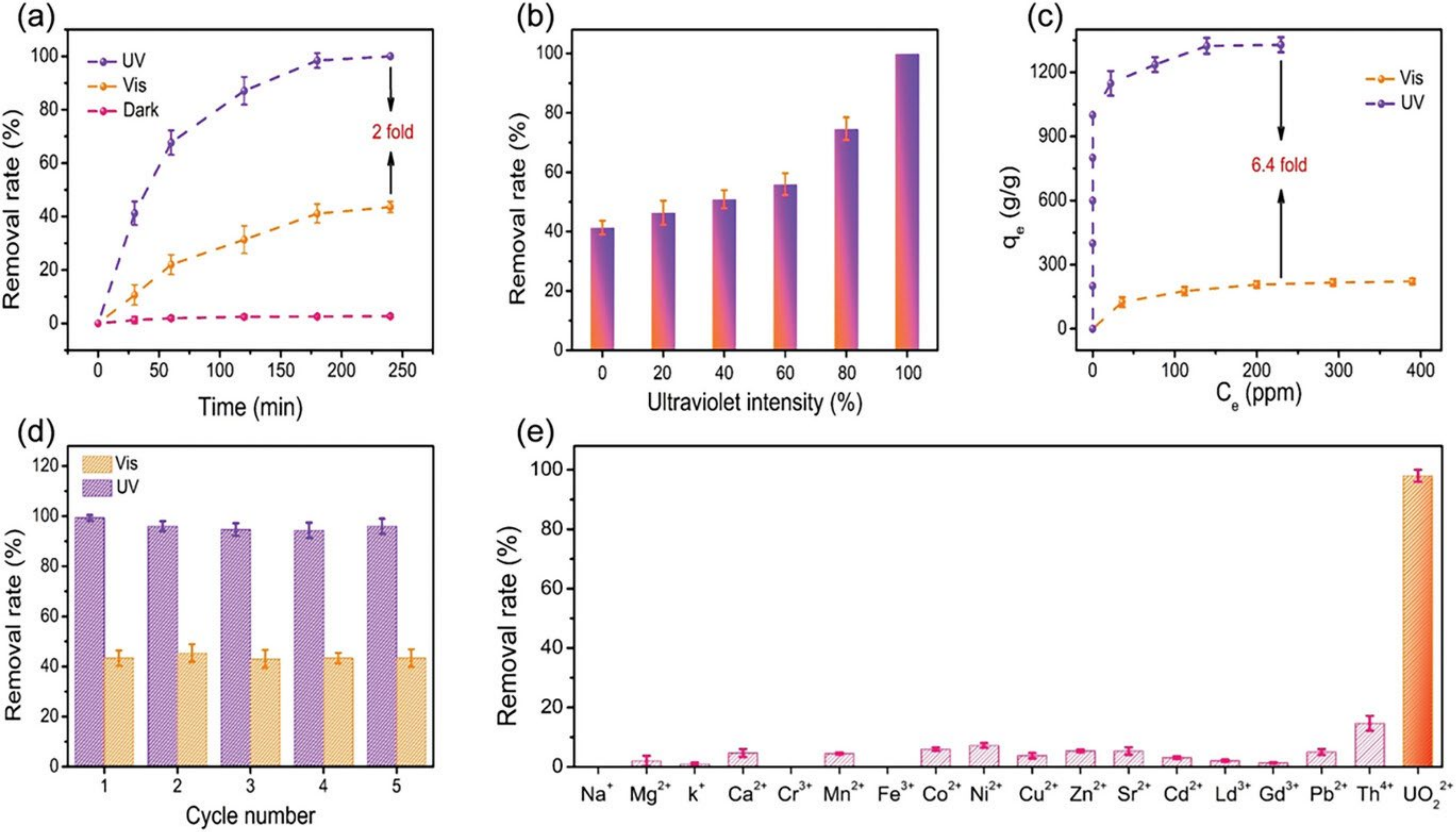


Figure 3. a) Uranium adsorption kinetics upon ECUT-Azo-COF-1 under various conditions such as visible light and UV irradiation and dark environment. b) A comparison of uranium removal efficiency under various ultraviolet intensity. c) Uranium adsorption isotherms upon ECUT-Azo-COF-1 under visible light and UV irradiation. d) Recycle tests under both visible light and UV irradiation. e) Selective adsorption toward uranium under UV irradiation from a 18-ions mixed solution.

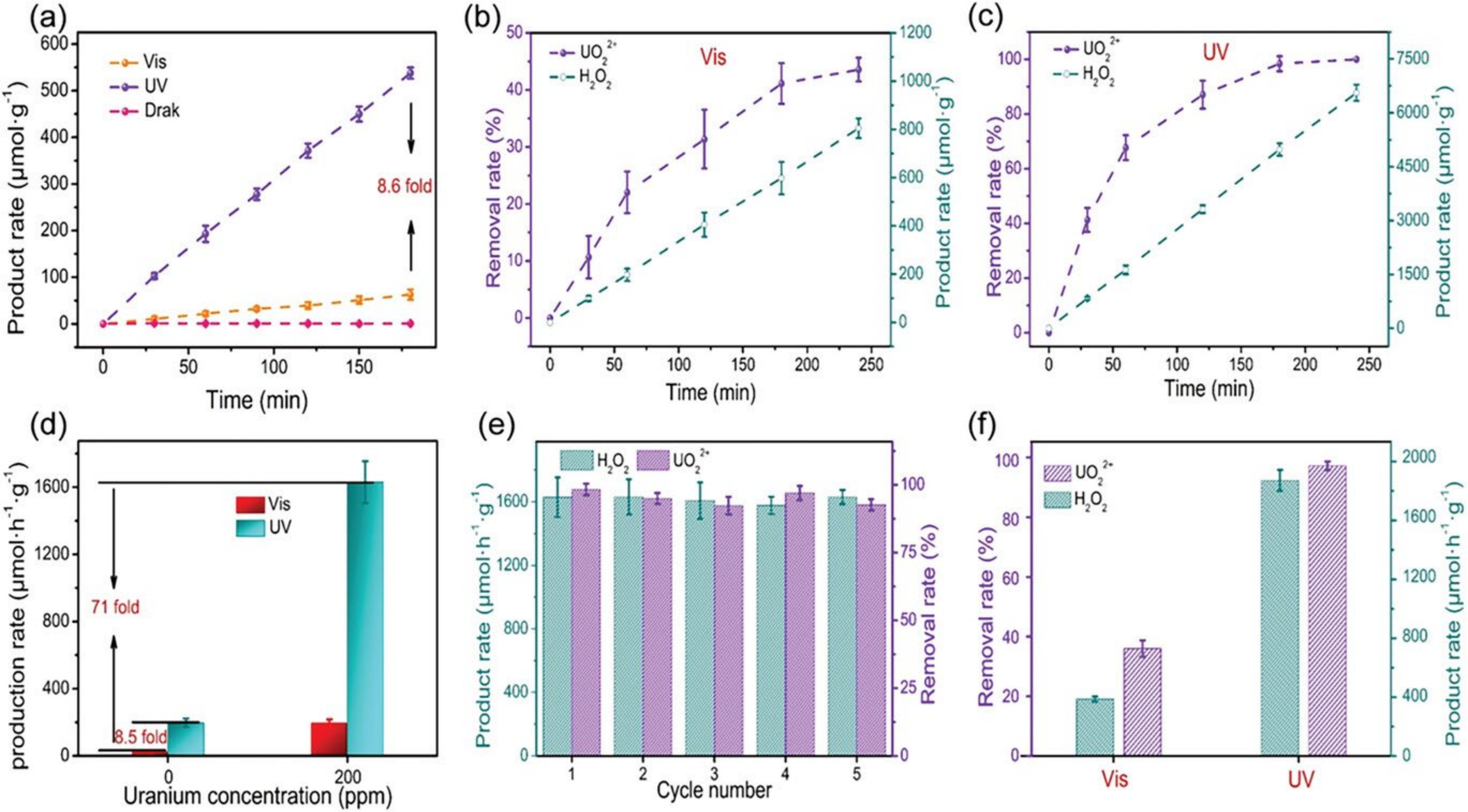


Figure 4. a) Photosynthesis of H_2O_2 upon ECUT-Azo-COF-1photocatalyst under visible light, UV irradiation, and dark environment. b) Photocatalysis coupling upon ECUT-Azo-COF-1photocatalyst under visible light irradiation in the presence of 200 ppm UO_2^{2+} ions. c) Photocatalysis coupling upon ECUT-Azo-COF-1 photocatalyst under UV irradiation in the presence of 200 ppm UO_2^{2+} ions. d) A comparison of photosynthesis of H_2O_2 under visible light and UV irradiation with the presence of 200 ppm UO_2^{2+} ions or not. e) Photocatalysis coupling recycle tests. f) Photocatalysis coupling for the real uranium-containing wastewater under visible light and UV irradiation.

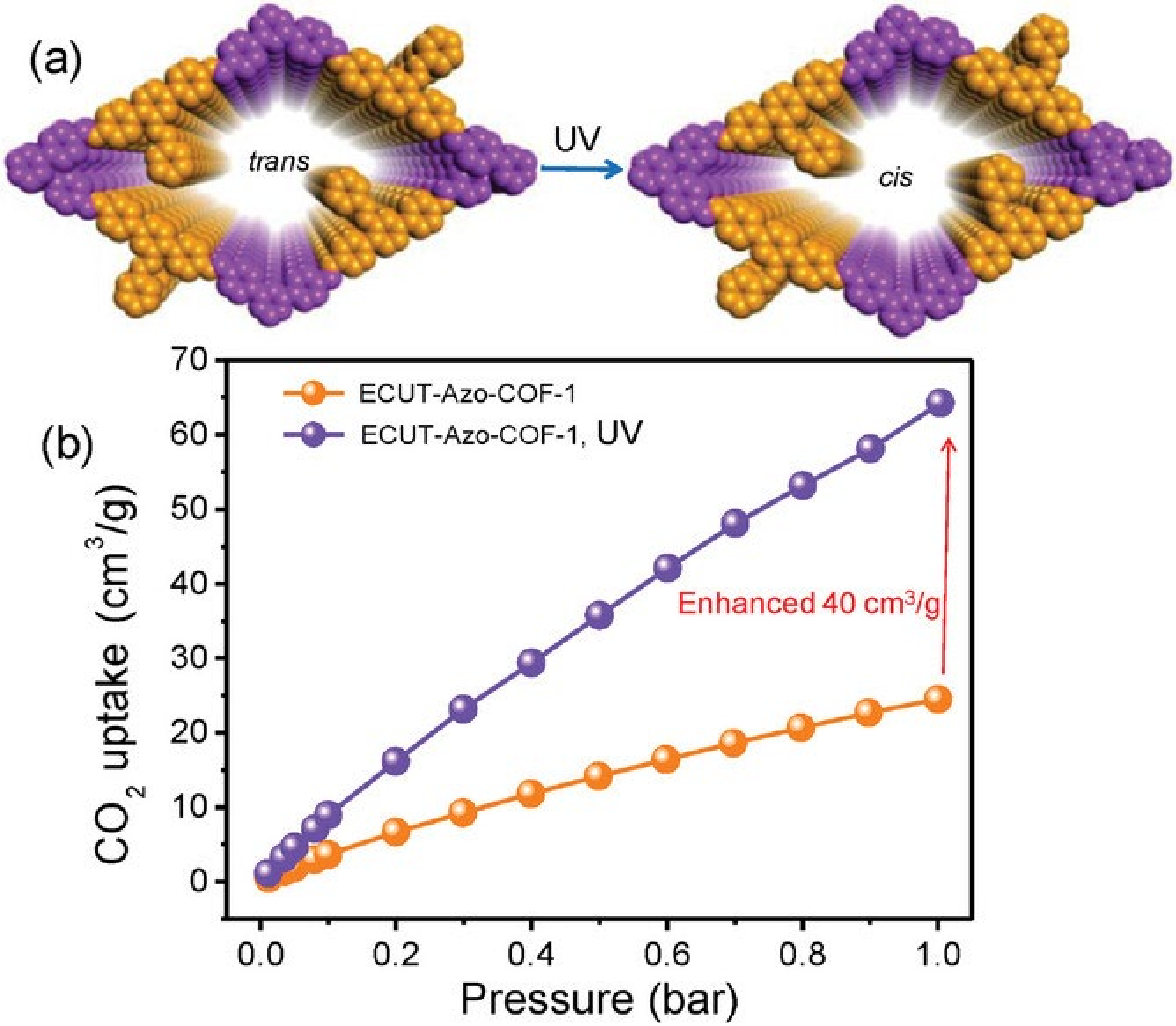


Figure 5. a) View of the *trans*-to-*cis* isomerization of azobenzene under UV irradiation. b) The CO2 adsorption isotherms at 298 K under visible light and UV irradiation.

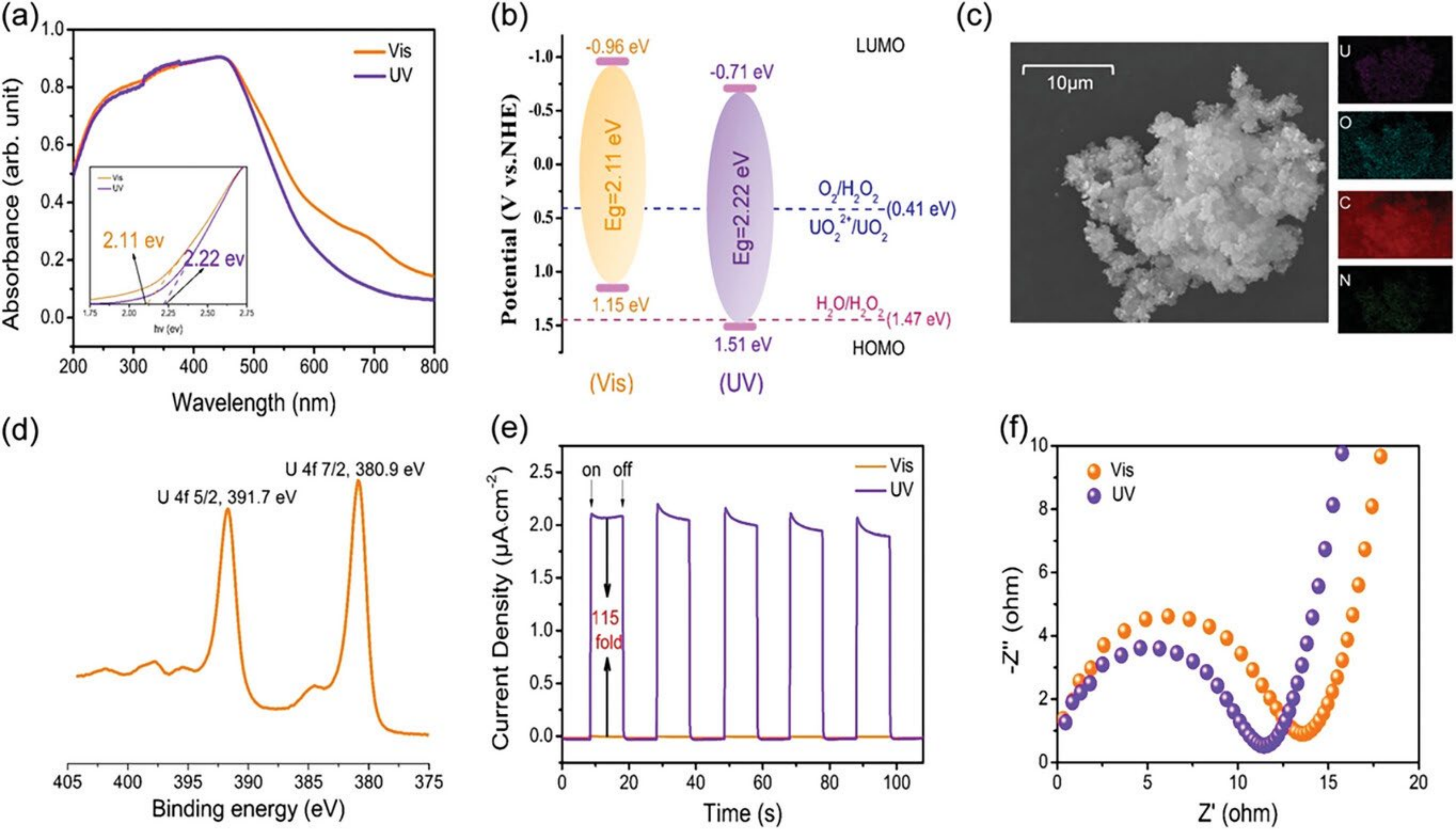


Figure 6. a) Solid-state UV–vis spectra of ECUT-Azo-COF-1 and the counterpart under UV irradiation and corresponding Kubelka–Munk plots. b) The HOMO and LUMO level of ECUT-Azo-COF-1 and the counterpart under UV irradiation. c) The SEM-EDS of samples after photocatalysis coupling. d) XPS spectra of U element for the samples after photocatalysis coupling. e) Transient current density of ECUT-Azo-COF-1 under visible light and UV irradiation. f) Electrochemical impedance spectra (EIS) of ECUT-Azo-COF-1 and the counterpart under UV irradiation.

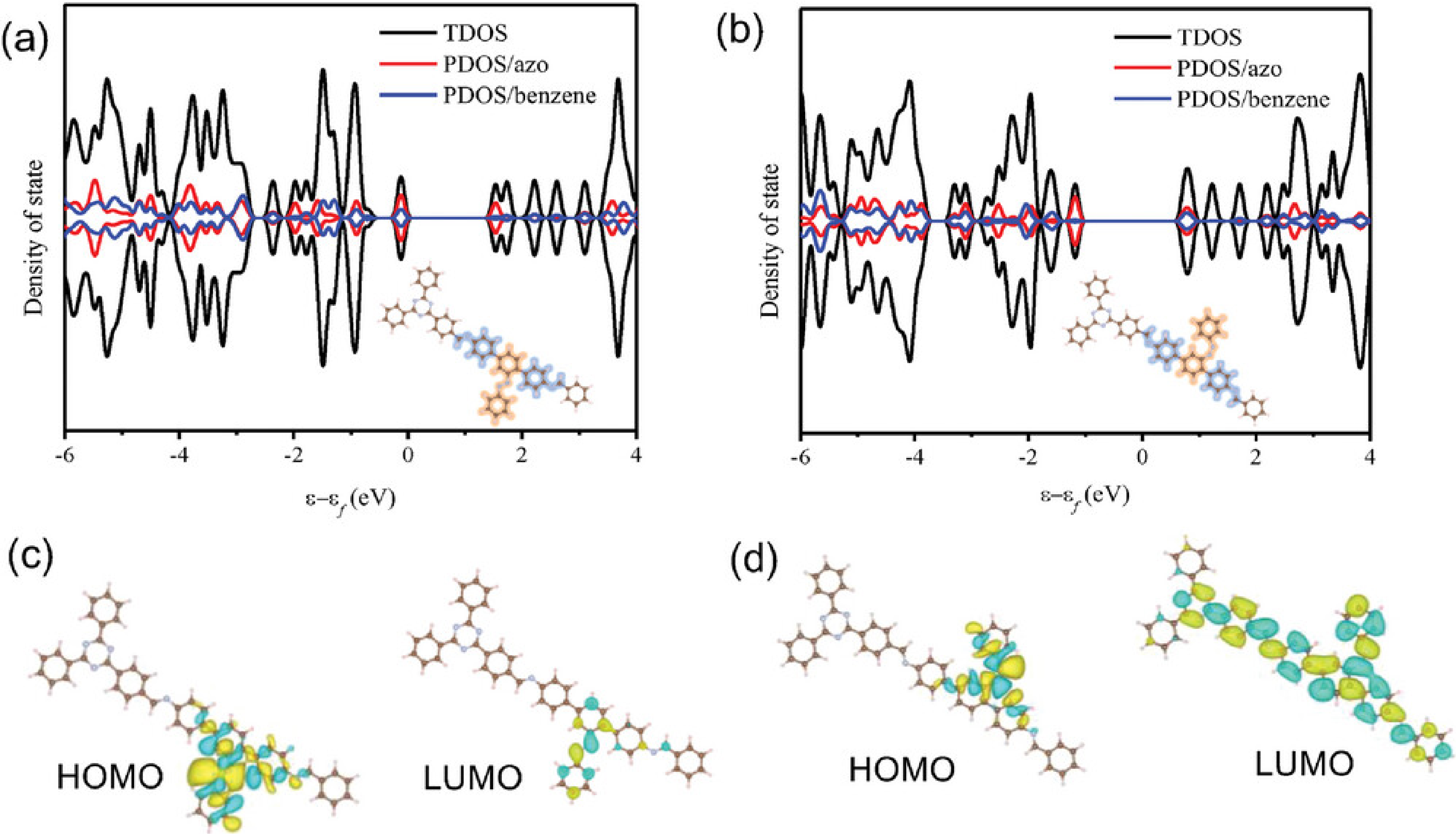


Figure 7. a,b) The density of states (DOS) and partial density of states (PDOS) of *trans*- and *cis*-isomers. c,d) The HOMO and LUMO of *trans*-isomer and *cis*-isomers. The cyan and yellow colors indicated the spin-up or spin-down, respectively. The isosurface value is set to 3.6 × 10−8 e/ Bohr−3. The brown, silver, and pink balls represent carbon, nitrogen, and hydrogen atoms.

Conclusion

- They successfully demonstrated an innovative conversion strategy using a novel photosensitive azobenzene-pendent COF and photocatalytic technology for treating uranium-containing wastewater.
- The results revealed that cooperative photocatalysis under UV irradiation enabled a highly efficient conversion of wastewater into hydrogen peroxide while achieving complete uranium removal.
- The effectiveness of conversion concept was further verified through its application in real uranium-contaminated wastewater.

Thank you