Supporting Information:

Electrostatic Self-Assembly Synthesized Fe-MOF/g-C_3N_4 S-Scheme Heterojunction for Enhanced Photocatalytic Pollutant Degradation

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Experimental Methods

Preparation of g-C_3N_4 (CN): Aluminum foil was used to cover a crucible that contained 10 g of urea. Bulk C_3N_4 was produced by heating to 550 °C at a rate of 5 °C/min, keeping it there for 4 hours in a muffle furnace, and then letting it cool to room temperature. To create g-C_3N_4, the powder was dissolved in 50 mL of pure ethanol, subjected to an ultrasonic treatment for 18 hours, centrifuged, and dried.

Preparation of Fe-MOF: The hydrothermal process was used to create Fe-MOF. Fumaric acid (1.16 g) and iron (III) chloride hexahydrate (1.62 g) were dissolved in 100 mL of N, N-dimethylformamide while being stirred and heated at 120 °C for two hours. After five cycles of centrifugal washing with deionized water and cooling to room temperature, MIL88A (Fe-MOF) was produced.

Preparation of Fe-MOF/g-C_3N_4: Powders of Fe-MOF and g-C_3N_4 were mixed and ground for 30 minutes. Obtained porous Fe-MOF/CN materials were calcined in a tube furnace under 350 °C for 2 h. On the basic of the different reactant mass ratio of Fe-MOF and g-C_3N_4 that is 1:10, 2:10, 3:10, 4:10, and 5:10, and the products are denoted MCN1-10, MCN2-10, MCN3-10, MCN4-10, and MCN5-10, respectively.

Microstructural Characterizations

The X-ray diffraction (XRD) patterns of the materials were obtained using Cu Kα radiation on a Bruker D8 Advance diffractometer. Fourier-transform infrared (FT-IR) spectra were recorded using an IN10+IZ10 spectrometer. The morphology and microstructure of the samples were characterized by transmission electron microscopy (TEM, JEM-2100F) and scanning electron microscopy (SEM, SU 8230). Elemental distribution was analyzed using an energy-dispersive X-ray (EDX) spectrophotometer attached to the TEM. Nitrogen adsorption-desorption isotherms were measured using a BELSORP-Max analyzer. The chemical composition and elemental states were determined by X-ray photoelectron spectroscopy (XPS, Thermo Fisher ESCALAB Xi+). UV-Vis diffuse reflectance spectra were acquired using a PELambda950 spectrophotometer. Photoluminescence (PL) spectra were recorded on a Perkin Elmer LS55 fluorophotometer. Electron paramagnetic resonance (EPR) signals were collected using an A300-9.5/12 spectrophotometer.

Photoelectrochemical Measurements

A 300 W xenon lamp was used as the simulated sunlight source. The photocatalytic performance of the materials was evaluated using Rhodamine B (RhB) as the target pollutant under visible light irradiation. In a typical experiment, 30 mg of the photocatalyst was dispersed in 30 mL of an aqueous RhB solution (0.1 g/L). The suspension was stirred in the dark for 0.5 h to achieve adsorption-desorption equilibrium, followed by irradiation under the xenon lamp. At 10-minute intervals, 1 mL of the solution was collected and filtered through a 0.22 μm membrane. The absorbance of the filtrate was measured using a UV-Vis spectrophotometer (Shimadzu UV3600) to determine the RhB concentration. The photocatalytic efficiency (η) was calculated using the following formula:

$\eta = C_t/C_0\#(1)$in which C_0 is the initial concentration of RhB (mg/L); C_t is the concentration of RhB after time t (mg/L). The reaction rate constant k_{obs} (min^{-1}) is calculated as:

$\ln C_t = \ln C_0 - k_{obs} \cdot t\#(2)$

Electrochemical measurements were performed on a CHI electrochemical workstation (Shanghai Chenhua Instrument Co., Ltd., China) using a three-electrode system. Ag/AgCl and platinum were used as the reference and counter electrodes, respectively. The working electrode was prepared by depositing the catalyst on an FTO glass substrate with an active area of 2 × 2 cm^2. The electrolyte used was 0.5 M sodium sulfate.

Free Radical Trapping Experiments

To identify the active species involved in the photocatalytic process, radical trapping experiments were conducted. Before illumination, 1 mL of isopropanol (IPA), 0.1 g of benzoquinone (BQ), and 0.3 g of ethylenediaminetetraacetic acid (EDTA) were added to the RhB solution to scavenge hydroxyl radicals (·OH), superoxide radicals ($\cdot O_2^-$), and holes (h^+), respectively. The remaining procedures were identical to the photocatalytic degradation experiments. Based on these experiments, the photocatalytic reaction mechanism was further elucidated.

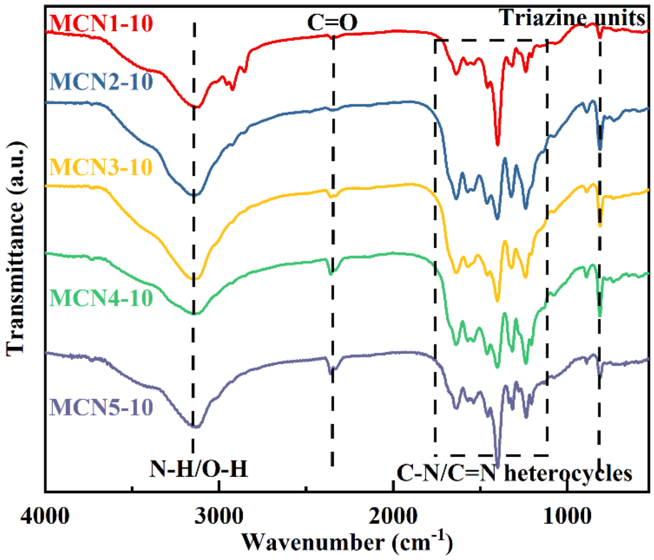

Fig. S1. FT-IR patterns of MCNx.

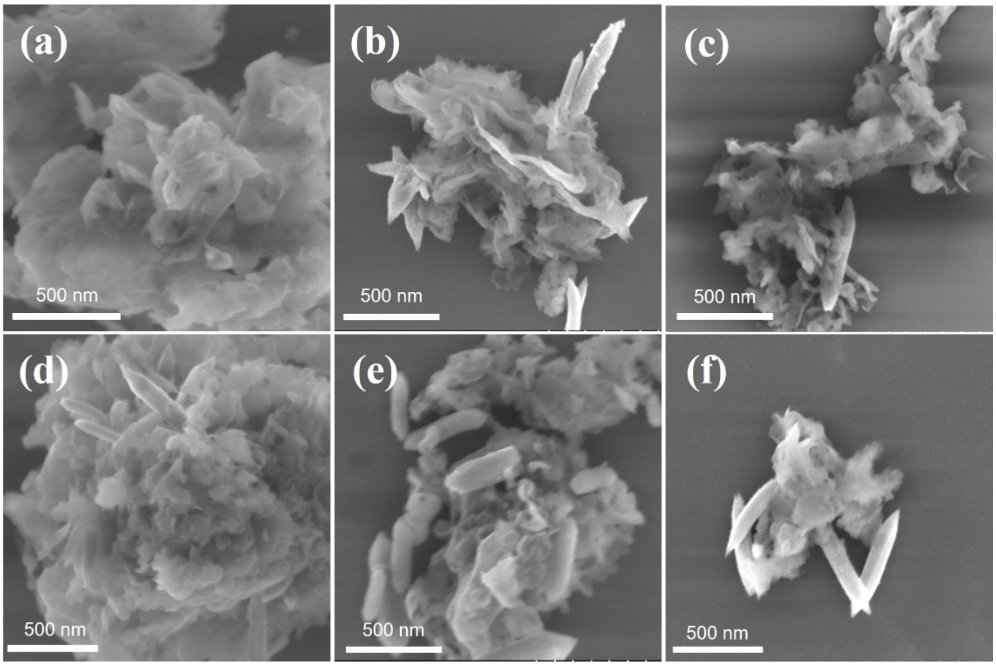

Fig. S2. SEM image of (a) g-C_3N_4, (b)MCN1-10, (c) MCN2-10, (d) MCN3-10, (e) MCN4-10, (f) MCN5-10.

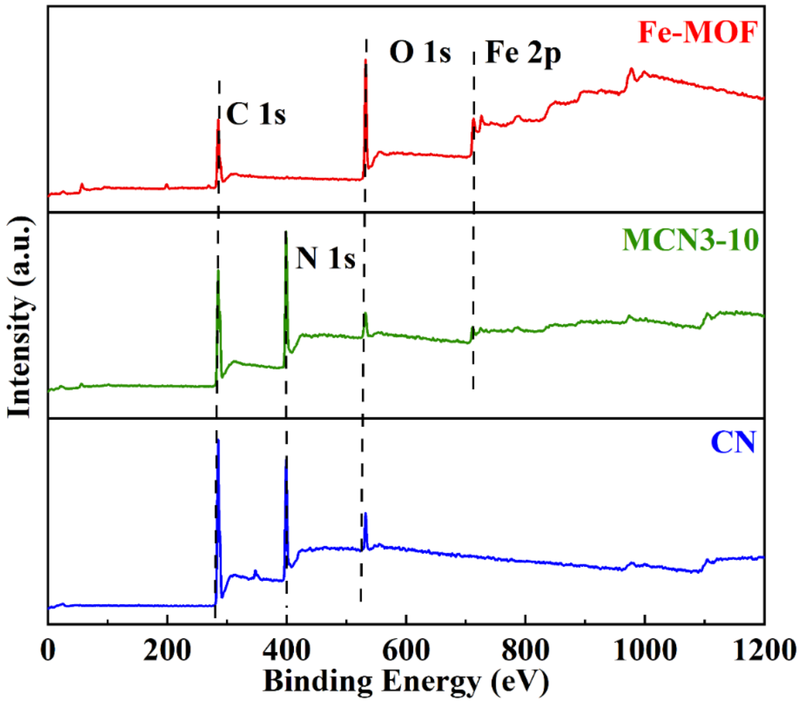

Fig. S3. XPS spectroscopy of survey for Fe-MOF, CN and MCN3-10.

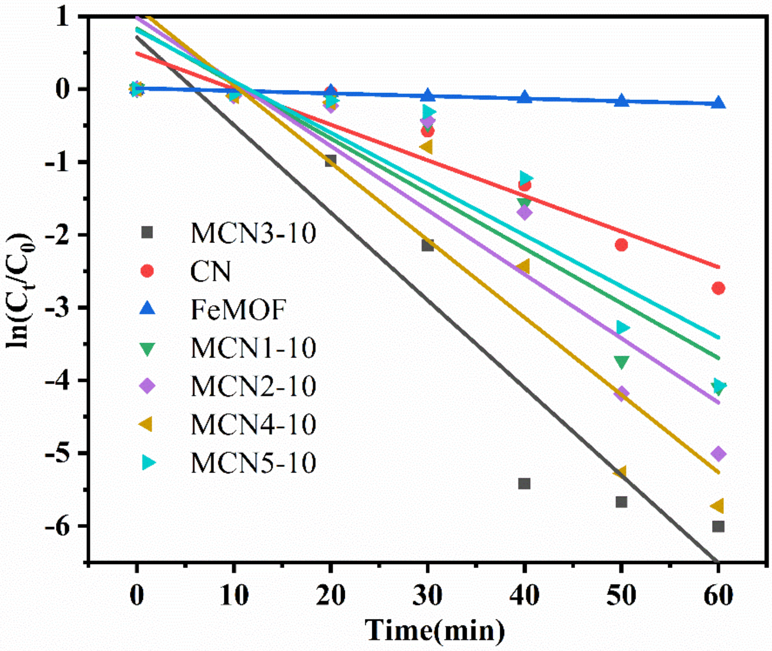

Fig.S4. Linear plots of ln(Ct/C0) vs. time for for all samples.

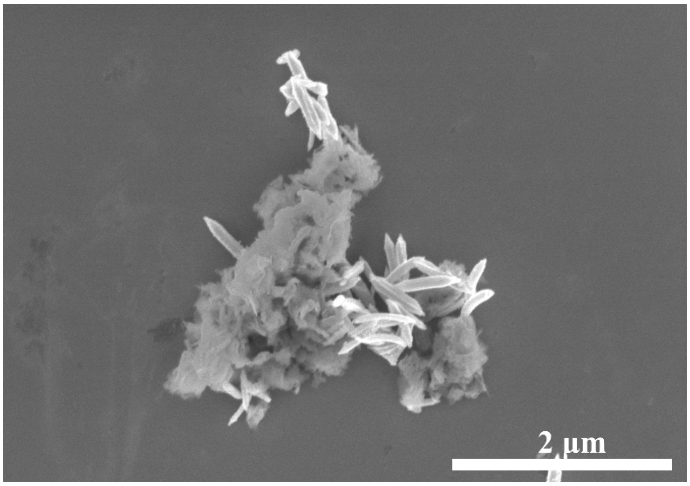

Fig.S5. The SEM image of MCN3-10 after five consecutive cycles.

Table S1. MOF/g-C_3N_4-based photocatalysts for RhB degradation in comparison with recently reported studies.

Photocatalysts	**synthesis method**	**degradation time**	**Stability**	**Reference**
MOF/g-C_3N_4	electrostatic self-assembly	40min	90%	This work
MOF-5/g-C_3N_4	mechanical grinding method	90min	90%	[S1] *Environmental Science and Pollution Research, 2024, **31**, 60298-60313.*
g-C_3N_4@MOF	the phase inversion method	180min	90%	[S2] *Surfaces and Interfaces*, 2024, **55**, 105399.
Ni-MOF/g-C_3N_4	integrated	-	92%	[S3] *ACS ES&T Water*, 2024, **4**, 4454-4463.
Fe_2O_3/g-C_3N_4	thermal polymerization and hydrothermal methods	150min	-	[S4] *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, 2024, **310**, 123972.
graphene quantum dots/g-C_3N_4	sonication-assisted mechanochemical approach	120min	-	[S5] *Scientific Reports*, 2025, **15**, 27276.
ZnO/g-C_3N_4	single-step calcination	-	-	[S6] *Journal of Saudi Chemical Society, 2024, **28**, 101821.*

References

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