

Supporting Materials

Table S1 R1, R2, R3 and R4 experimental conditions.

Name of system	Fe dosage form	Whether or not H ₂ O ₂ is added	Whether or not to add electricity
R1	SI and GAC	Do not add	electric powerless
R2	SI and GAC	add	electric powerless
R3	Fe ²⁺	add	add electricity
R4 (3DEF)	SI and GAC	add	add electricity

Notes: Except for the differences in the conditions mentioned above in this table, the other conditions remain the same: pH of 3–3.5, particle electrode dosage of 100 g/L, SI:GAC mass ratio of 3:1, 1.45 mmol/L Fe²⁺ current density of 30 mA/cm², H₂O₂ dosage of 50 mmol/L, cathodic aeration of 0.8 L/min, and hydraulic retention time of 120 min.

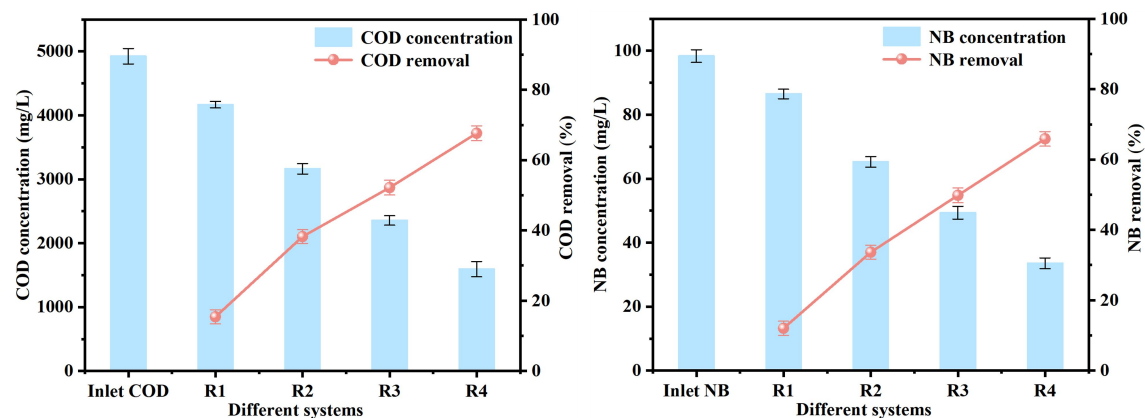


Fig. S1 Removal efficiency of COD and NB in different systems.

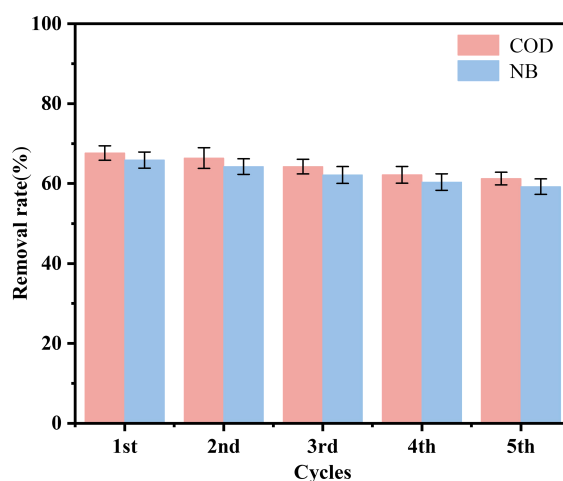


Fig. S2 Stability of iron-carbon filler in 3DEF system. Condition: COD = 4926.20 mg/L, NB = 98.35 mg/L, other conditions are the most conditions explored.

Table S2 D1, D2, and D3 experimental conditions (Supplementary data for stability test experiments).

Name of system	Fe dosage form	Fe ²⁺ dosing rate	Whether or not H ₂ O ₂ is added/electricity
D1 (3DEF)	SI and GAC	/	add
D2	Fe ²⁺	1.45mmol/L	add
D3	Fe ²⁺	3.75mmol/L	add

Notes: Except for the differences in the conditions mentioned above in the table, the other conditions remain the same: pH of 3–3.5, particle electrode dosage of 100 g/L, SI:GAC mass ratio of 3:1, current density of 30 mA/cm², H₂O₂ dosage of 50 mmol/L, cathodic aeration of 0.8 L/min, and hydraulic retention time of 120 min.

To provide a clearer comparison than in the literature between homogeneous and heterogeneous reactions, we evaluated the COD and NB removal rates and sludge production across the D1 (3DEF), D2, and D3 systems. The results showed that the COD and NB removal rates in the D1 system were 67.65% and 65.88%, respectively; in the D2 system, they were 52.17% and 49.82%, respectively; and in the D3 system, they were 48.57% and 45.26%, respectively. In terms of sludge production, the amounts in the D1 and D3 systems were not significantly different. These experimental results indicate that under the optimized reaction conditions of the D1 (3DEF) system, the advantages of the iron-carbon filler in stabilizing iron ion availability, catalyst regeneration capability, and porous structure lead to a significant increase in COD and NB

removal rates. This phenomenon results in a decrease in the sludge being produced per kg of COD removed. The experimental results of the D2 and D3 systems demonstrate that an excess of Fe^{2+} rapidly consumes H_2O_2 , reducing the production of $\bullet\text{OH}$; excess Fe^{2+} reacts with $\bullet\text{OH}$, leading to radical quenching and decreased oxidation efficiency; and the resulting $\text{Fe}(\text{OH})_3$ precipitate encapsulates organic pollutants, reducing their contact with $\bullet\text{OH}$.