Metal-organic Framework-derived Iron Nitride Composites for Rapid Light-Driven Diclofenac Degradation via Persulfate Activation

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Abstract

The prevalence of pharmaceuticals and other organic micropollutants in surface and groundwater poses a serious environmental consequence. Here, metal–organic framework (MOF)–derived iron nitrides (FeN) carbon composites were designed as an efficient photochemical oxidation catalyst to degrade diclofenac (DCF) via persulfate (PS) activation. The carbon-supported FeN materials (FeN(w%)@C_x, with w and x representing the weight percentage and the pyrolysis temperature, respectively) were obtained by preparing iron-doped MOF precursors and subsequently subjecting them to high-temperature pyrolysis. The prepared FeN(w%)@C_xs were applied to remove DCF from aqueous media after thorough characterization. The FeN(w%)@C_xs efficiently activated PS under light condition (intensity 950 W m^{-2}), enabling rapid DCF degradation with high turnover frequencies. DCF removal efficiency reached nearly ~93% in 40 min using FeN(10%)@C_{950}/PS system under optimized parameters such as catalyst (0.05 g L^{-1}), PS dose (1 mM), and initial DCF concentration (10 mg L^{-1}), etc. The Fe–N–C structure of FeN(10%)@C_{950} enables the light–driven activation process involving radical species ($SO_4^{\bullet-}$ and $^{\bullet}OH$), as well as non-radical reactive oxygen species ($^1O_2$), proceeding via an electron-transfer mechanism, as confirmed by scavenger tests, ESR spectroscopy, and electrochemistry. DCF degradation rate was influenced with the addition of different anions, cations, and organic acids in FeN(10%)C_{950}/PS system under light. The optimum pH (5-9) showed higher degradation rate. Additionally, the reusability and stability of the FeN(10%)@C_{950} were confirmed over multiple cycles and DCF degradation products using LC-MS. Finally, the obtained results emphasize the potential of these catalysts in wastewater remediation.

Keywords

Iron Nitride Composites, Persulfate, Pharmaceuticals, Photochemical Oxidation, Wastewater Treatment