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Carbon-Functionalized Nanoscale Zerovalent Iron for Enhanced Removal and Transformation of Ciprofloxacin

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The reactivity of nanoscale zerovalent iron (nZVI) toward organic contaminants, including fluorinated pharmaceuticals such as ciprofloxacin, is influenced by iron corrosion dynamics and electron transfer efficiency. In oxic systems, rapid iron oxidation and uncontrolled Fe release can limit electron utilization for contaminant transformation. Carbon–iron composites have been proposed as a strategy to potentially regulate these processes; however, the extent to which carbon incorporation chemistry influences corrosion pathways and redox behavior remains insufficiently understood. In this study, carbon-functionalized nZVI composites were prepared via a facile approach in which polymer-coated pre-formed particles were subjected to pyrolysis, using poly(vinyl alcohol) (PVA) and carboxymethyl cellulose (CMC) as carbon precursors. Despite comparable initial carbon dosing, the two polymers produced distinct levels of carbon incorporation and surface textural properties, reflecting differences in functional group chemistry and thermal decomposition behavior. Spectroscopic investigations indicated the presence of mixed amorphous and graphitic carbon domains associated with the iron surface. Under oxic conditions, carbon functionalization altered ciprofloxacin transformation behavior relative to bare nZVI. Apparent pseudo-first-order rate constants were moderately higher for CMC-derived composites compared to bare nZVI, whereas PVA-derived composites exhibited approximately two-fold lower rate constants. Nevertheless, both carbon-functionalized composites demonstrated substantially higher equilibrium removal capacities on an iron-mass basis (approximately 30–50% greater than the ~50 mg g^{-1} Fe observed for bare nZVI). This enhancement occurred despite suppression of dissolved Fe release following carbon incorporation, even though ciprofloxacin degradation in oxic systems is influenced by iron dissolution and Fenton-type processes. Fluoride ion release was detected in solution for both carbon-functionalized nZVI particles, indicating partial defluorination of the fluoroquinolone structure during treatment. Mass spectrometric analysis revealed lower-mass transformation products in solution, while extracts of reacted composites confirmed sorption of both ciprofloxacin and intermediates to the carbon–iron surface. These observations indicate coupled adsorption and likely surface-mediated redox transformation occurring at the carbon–iron interface. Collectively, the results demonstrate that controlled carbon incorporation can modulate iron corrosion behavior and mechanistic pathways governing redox-active nanomaterials in oxic environments, with implications for transformation of organofluorine pharmaceuticals.