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Ph.D. DISSERTATION DEFENSE

Candidate: Meng Ji
Degree: Doctor of Philosophy
School/Department: Charles V. Schaefer, Jr. School of Engineering and Science/Civil, Environmental & Ocean Engineering
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Title: Treatment of Perfluorooctane Sulfonic Acid (PFOS) and Nitrate using Micro-Scale Zero-Valent Iron (mZVI) from Water
Chairperson: Dr. Xiaoguang Meng, Department of Civil, Environmental & Ocean Engineering
Committee Members: Dr. Christos Christodoulatos, Department of Civil, Environment & Ocean Engineering
Dr. George Korfiatis, Department of Civil, Environmental & Ocean Engineering
Dr. Hongjun Wang, Department of Biomedical Engineering
Dr. Tsan-Liang Su, Department of Civil, Environmental & Ocean Engineering

ABSTRACT

The widespread use of Aqueous Film Forming Foam (AFFF) has led to perfluorooctane sulfonic acid (PFOS, $C_8F_{17}SO_3H$) pollution in water, soil, and air, which is directly attributable to adverse effects on human health due to its high water solubility and high thermal and chemical stability. Nitrate (NO_3^-) is a common contaminant in groundwater and surface water globally, and the United States Environmental Protection Agency (EPA) has set the maximum contaminant level (MCL) of 10 mg-N/L for NO_3^- in drinking water to protect human health. Microscale zero valent iron (mZVI) has been widely used to treat organic and inorganic contaminants for its high chemical reactivity and environmental safety. This dissertation investigated the kinetics and mechanisms of PFOS and nitrate removal using mZVI.

For PFOS, the dissertation compares the removal of PFOS by mZVI and activated carbon (AC) and investigates the contribution of both the magnetic and non-magnetic portions of reacted mZVI to the removal of PFOS. Results showed that the mZVI had a significantly higher areal adsorption capacity (21 mg/m^2) compared to AC (0.813 mg/m^2). 10 g/L of mZVI at neutral pH reduced PFOS concentrations from 50 mg/L to 6 mg/L within 8 hours. Magnetic solids showed a significantly higher removal capacity than iron oxides and hydroxides. Low F^- concentration (~1 mg/L) suggested that defluorination was not the main removal mechanism. Acid-washed ZVI showed only minor improvements in removal efficiency, indicating the iron oxide layer does not significantly affect PFOS adsorption. The in-situ ATR-FTIR spectra revealed that the removal of PFOS by mZVI was not due to the formation of covalent bonds between PFOS and mZVI. The negatively charged mZVI significantly removed PFOS anions at pH>7, indicating that the removal mechanism was not due to the electrostatic attraction. Furthermore, the presence of NaCl enhanced PFOS removal, proving the hydrophobic effect as a key mechanism.

For nitrate, this dissertation investigated the synergistic use of mZVI combined with anaerobic nitrate-reducing bacteria to enhance the removal of high nitrate concentrations (~400 mg/L NO_3-N) from wastewater. Experimental results showed that the combined mZVI with bacteria (mZVI-B) system achieved complete NO_3^- reduction within 25 hours, compared to approximately 30 hours for the B-only system, with reduced nitrite (NO_2^-) accumulation and negligible NH_4^+ production. After five consecutive treatments, the reduction time further decreased to 15 hours. Optimal NO_3^- removal occurred at a neutral pH (7-8), carbon-to-nitrogen (C/N) ratios (1.8-3.6), and with higher mZVI dosages (9 g/L). The 3-(4,5-Dimethylthiazol-2-yl)-2,5-Diphenyltetrazolium Bromide (MTT) and 16S metagenomic results indicated that the addition of mZVI enriches the abundance of Pseudomonas, Paracoccus, and Thauera at the genus level. Scanning electron

microscopy (SEM) images revealed biofilm formation on mZVI. These findings underscore the potential of the mZVI-B system as an effective and sustainable solution for treating water affected by high concentrations of NO_3^-.