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Chloramphenicol removal by zero valent iron activated peroxymonosulfate system: Kinetics and mechanism of radical generation

Chaoqun Tan^{a,b*}, Yujie Dong^{a,b}, Dafang Fu^{a,b}, Naiyun Gao^c, Jinxia Ma^{a,b}, Xiaoyu Liu^{a,b}

a School of Civil Engineering, Southeast University, Nanjing 210096, China

b Key Laboratory of Concrete and Prestressed Concrete Structures of the Ministry of Education, Southeast University, Nanjing, 210096, China

c Skate Key Laboratory of Pollution Control and Resource Reuse, Tongji University, Shanghai City 200092, China

Abstract: Zero valent iron (Fe^0) was used as a catalyst to activate peroxymonosulfate (PMS) for removal of chloramphenicol (CAP). About 95.2% of 10 mg/L CAP was removed by 0.2 mM peroxymonosulfate and 0.5 g/L Fe^0 at neutral pH, with little Fe^{3+} leaching. The degradation reactions well followed a pseudo-first-order kinetics pattern ($R^2 > 0.95$). The production of hydroxyl radicals ($\text{OH}\cdot$) and sulfate radicals ($\text{SO}_4\cdot^-$) was directly identified in in-situ Electron paramagnetic resonance (EPR) tests with 0.1 M 5,5-dimethyl-1-pyrrolidine N-oxide (DMPO) and indirectly identified through quenching tests. The second order rate constant between CAP and $\text{SO}_4\cdot^-$ was ascertained to be $6.18 \times 10^{10} \text{ M}^{-1} \text{ s}^{-1}$ in competition reaction. According to SEM and XRD analysis, the average diameter of catalyst was elevated to be about 88 nm after use. Possible mechanisms on the radical generation were proposed based on the results of EPR and XPS analyses. It emerged that the cycle of Fe^{2+} - Fe^{3+} on the Fe^0 was answerable for the $\text{OH}\cdot$ and $\text{SO}_4\cdot^-$ generation, and iron sulfate layer was assembled on the Fe^0 following exposure to peroxymonosulfate compared to the FeOOH layer present on the fresh Fe^0 . While the presence of common anions including Cl^- , CO_3^{2-} , Fe^{2+} , Cu^{2+} and Mn^{2+} will inhibit the degradation of CAP in Fe^0 /PMS system. Besides, Fe^0 /PMS pre-oxidation before chlorine or chloramine engendered in a sharp increment in the concentration of disinfection by-products (DBPs) precursors. The results manifested that Fe^0 /PMS was effective in CAP removal at neutral pH, whereas it should be more cautiously investigated when combined with chlorine disinfection.

Corresponding author. Tel.: +86 025 83790750

Fax.: +86 025 83790750

E-mail address: tancq@seu.edu.cn (CQ. Tan)

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