

Decomposition of Hydrogen Peroxide and Organic Compounds in the Presence of Iron and Iron Oxides

by

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Abstract

Most advanced oxidation processes use the hydroxyl radical ($\bullet\text{OH}$) to treat pollutants found in wastewater and contaminated aquifers because $\bullet\text{OH}$ reacts with numerous compounds at near diffusion-limited rates. $\bullet\text{OH}$ can be made by reacting hydrogen peroxide (H_2O_2) with either Fe(II) (the Fenton reaction), Fe(III), or iron oxide. This dissertation investigated the factors that influence the decomposition rates of H_2O_2 and organic compounds, as well as the generation rate of $\bullet\text{OH}$ (V_{OH}), in the presence of dissolved Fe(III) and iron oxide.

The Fe(III)-initiated chain reaction could be the dominant mechanism for the decomposition of H_2O_2 and organic compounds. The degradation rates of H^{14}COOH , an $\bullet\text{OH}$ probe, and H_2O_2 were measured in experiments at pH 4 containing either dissolved Fe(III) or ferrihydrite. Combined with the results from experiments using a radical chain terminator, we concluded that a solution chain reaction was important only in the Fe(III) system. In the ferrihydrite system the amount of dissolved Fe was insufficient to effectively propagate the chain reaction. In addition, a nonradical producing H_2O_2 loss pathway exists at the oxide surface.

The oxidation rate of any dissolved organic compound can be predicted from V_{OH} if the main sinks of $\bullet\text{OH}$ in the solution are known. Experiments using H^{14}COOH and ferrihydrite, goethite, or hematite showed that V_{OH} was proportional to the product of the concentrations of surface area and H_2O_2 . Based on these results, a model was created for predicting the pseudo-first-order oxidation rate coefficients of dissolved organic compounds (k_{org}) in systems containing iron oxide and H_2O_2 . While our model successfully predicted k_{org} in aquifer sand experiments, it yielded mixed results when compared to measurements from previously published studies.

Some factors that could have caused the disagreements between model predictions and measurements were examined to refine our model. Results from experiments containing goethite, H^{14}COOH , and 2-Chlorophenol showed that H^{14}COOH detected more $\bullet\text{OH}$, which is produced at the oxide surface, than did 2-Chlorophenol. This was attributed to electrostatic attraction between the formate anions and the positively charged oxide surface, and could explain why our model, based on H^{14}COOH , overpredicted the k_{org} values of many neutral compounds.

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