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Oxalate Enhanced Aniline Degradation by Goethite: Structural Dependent Activity, Hydroxyl Radicals Generation and Toxicity Evaluation

TIANYUAN XU

China University of Mining and Technology

Presenting Author: xutianyuan@cumt.edu.cn

A photochemical system combining iron (hydr)oxides and oxalate (Ox) shows application prospects in wastewater treatment due to abundance reactive oxygen species (ROS) generation. Nevertheless, it is a challenge to the investigate photochemical activity of iron (hydr)oxides/Ox system with varying structural properties. Herein, the photochemical behaviors of Ox on goethite (Gt) surface from the view of structural dependent activity, containment degradation, ROS generation were explored in detail. Results confirmed that bidentate mononuclear complex was formed on Gt surface after complexing Ox. With the assistance of visible light, the Fe(III)-Ox complexes formed on Gt or detached in solution underwent photolysis, resulting in superior efficiency of aniline degradation. After irradiating 120 min visible light, 96.5% aniline was degraded by 1.0 mM Ox and 0.2 g/L Gt. Combined with density functional theory calculation and pH time evolution during aniline degradation, the photochemical activity of the Gt/Ox system fell in between that of ferrihydrite/Ox and hematite/Ox systems. As proved by the experiments of radical quenching and electron paramagnetic resonance, •OH and $O_2^{\bullet-}$/•OOH played a key role in aniline degradation, which generated during the process of dissolved oxygen activation by $CO_2^{\bullet-}$ radicals and Fe(II). The amount of •OH in vis/Gt/Ox system could be up to 309.3 μM, and its generation was closely associated with Fe(II) while slightly affected by the generated H_2O_2. Moreover, as revealed by high-performance liquid chromatography with mass spectrometric and ECOSAR, the toxicity of the intermediates of aniline degradation in the vis/Gt/Ox system towards fish and green algae increased first but then declined accompanied with the generation of non-toxic ring-opening products at the end of reaction. According to the findings in the presented study, it could be concluded that vis/Gt/Ox is an encouraging approach to wiping out aniline wastewater.