Development of Heterogeneous Fenton Catalyst System Capable of Regenerating for Advanced Degradation Treatment of Persistent Pollutants

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[Abstract]

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The Fenton reaction is well-known for its use in the oxidative degradation of organic pollutants using hydrogen peroxide (H_2O_2). Iron ions (Fe^{2+} and Fe^{3+}) have been traditionally utilized as homogeneous catalysts. The oxidative degradation of organic contaminants is catalytically achieved using hydroxyl radical (·OH), which possesses excellent oxidizability in the Fenton reaction using Fe^{2+} and H_2O_2 (Equations (1) and (2)).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \cdot OH + OH^- \quad (1) \qquad Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + \cdot O_2^- + 2H^+ \quad (2)$$

However, there are some significant issues with the Fenton reaction. For homogeneous catalysis, the separation and recovery of the iron ions catalyst after the reaction requires considerable energy, expense, and effort. The effectiveness of the complete degradation to carbon dioxide (CO_2) is insufficient. The regenerating reaction from Fe^{3+} to Fe^{2+} (Equation (2)) barely occurs compared with the reaction of Equation (1). The homogeneous reaction generates a large amount of industrial waste in the form of iron hydroxide ($Fe(OH)_3$) sludge.

In this study, we concentrated on composite-photocatalysts combined with semiconductor photocatalyst and layered double hydroxide (FeAl-LDH) composed of Fe^{2+} and Al^{3+} species as heterogeneous Fenton catalysts. Because it is known that some LDH species have photocatalytic properties due to metal-to-metal charge transfer (MMCT). Photo-self-regenerating reaction from Fe^{3+} to Fe^{2+} can be expected by the photo-induced MMCT effect. Furthermore, semiconductor photocatalysts, such as titanium oxide (TiO_2) and tungsten trioxide (WO_3), possess a high reductive potential for reducing Fe^{3+} to Fe^{2+} by light irradiation.

FeAl-LDH as Fe(II)-LDH consisting of Fe^{2+} and Al^{3+} species demonstrated relatively-high degradation performance even under dark conditions. It should be noted that the conversion to CO_2 from phenol as a model compound of persistent pollutants under visible light (> 420 nm) irradiation reached *ca.* 2 times compared with that in dark. Furthermore, FeAl-LDH/TiO_2 or WO_3 accomplished conversion to CO_2 of > 99% under irradiation of ultraviolet-visible light (> 300 nm) or visible light (> 420 nm). From the results of XPS measurement, the ratio of Fe(II)/Fe(III) in these catalysts after reaction under light irradiation significantly increased compared with that before reaction. These unique phenomena might be caused by the reduction/regeneration of Fe(III) to Fe(II) in the synergy effect of “photocatalytic reaction of TiO_2 or WO_3” and “photo-self-regenerating effect of FeAl-LDH.”

This research contributes to the development of promising design guidelines addressing global environmental issues through the environmentally friendly degradation and detoxification of persistent pollutants.

[References]

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