



Role of sol with iron oxyhydroxide/sodium dodecyl sulfate composites on Fenton oxidation of sorbed phenanthrene in sand



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ABSTRACT

In situ Fenton oxidation has been recently used to oxidize sorbed organic contaminants in soil. The objective of present contribution was to study the role of sodium dodecyl sulfate (SDS) as anionic surfactant and sol with iron oxyhydroxide/SDS for Fenton oxidation of sorbed phenanthrene in sand. The most effective experimental condition for phenanthrene oxidation was the Fenton-like reaction system with 0.35% H₂O₂, 30 mM SDS, and 4 mM FeCl₂. The Fenton-like reactions under these experimental conditions resulted in the production and sustenance of a stable sol with iron oxyhydroxide/SDS composites over 24 h. The formation of iron oxyhydroxide/SDS composites resulted in stabilization of H₂O₂, and then the Fenton-like reactions were sustained over 24 h. Furthermore, the sol of iron oxyhydroxide/SDS composites gave suitable sites to sustain oxidations of dissolved phenanthrene over a prolonged reaction span, which is required for in situ chemical oxidation.

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1. Introduction

Soils and subsurface systems contaminated with organic compounds such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) are significant concern for the environment. These chemicals not only result in ecotoxicity but also are carcinogens and endocrine disruptors (Sullivan and Krieger, 1992).

The chemical structure of PAHs and their low water solubilities make them particularly hazardous to the environment. There are several different ways to degrade organic contaminants to ameliorate environmental damage. Some remediation technologies that have been explored for the removal of PAHs from soils involve a physical, chemical, biological, or thermal process or a combination of these processes (Gan et al., 2009).

In situ chemical oxidation has been investigated for more than a decade for the treatment (Rivas, 2006; Forsey et al., 2010; Park and Kim, 2011; Lemaire et al., 2011; Laurent et al., 2012). Particularly, the Fenton oxidant as one of reagents for in situ chemical oxidation is one of the most widely used and studied methods that can be applied to soil contamination. In this reaction system, hydroxyl radicals are produced by the reaction of hydrogen peroxide and

ferrous iron. It is a very powerful, effective, and nonspecific oxidizing agent, having rate constants ranging between 10⁷ to 10¹⁰ M⁻¹ s⁻¹ for organic compounds (Dorfman and Adams, 1973). The inorganic reactions involved in Fenton mediated oxidations are now well established (see Table 1). Laboratory-scale research on its potential applications to soil remediation began in the 1990s (Tyre et al., 1991; Watts et al., 1991, 1993; Ravikumur and Gurol, 1994; Kakarla and Watts, 1997).

However, a major challenge in the Fenton treatment of PAH-contaminated sites is to overcome the limited mass transfer due to the strong sorption and low aqueous solubilities of PAHs. In this case, only limited quantities of PAH contaminants are available for degradation by OH• radicals that are generated in the aqueous phase. Martens and Frankenberger (1995) first conducted research on a Fenton oxidation with an anionic surfactant to remediate PAH-contaminated soil. Furthermore, incorporating a surfactant and a solvating agent such as vegetable oil as a co-solvent into Fenton oxidations is reported to be a more viable alternative (Lee et al., 2002; Bogan et al., 2003; Martens and Frankenberger, 1995; Kyin et al., 2009; Gryzenia et al., 2009). In addition, recent research proved the role of SDS as H₂O₂ stabilizer in in-situ chemical oxidation (Kim et al., 2007; Park and Kim, 2011).

Furthermore, organic compounds may act as iron-complexing agents, modifying the redox potential of ferrous and/or ferric iron and thereby affecting the reaction rates of the Fe with reactive oxygen species such as H₂O₂ and superoxide anion (O₂⁻) (Francis

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Table 1
Inorganic Fenton reactions.

Reactions	Rate constant ($M^{-1} s^{-1}$)	Ref.
(1) $Fe(II) + H_2O_2 \rightarrow Fe(III) + OH\cdot + OH^-$	55	a
(2) $Fe(III) + H_2O_2 \rightarrow Fe(II) + HO_2/O_2^- + H^+$	2.00×10^{-3}	b, c
(3) $H_2O_2 + OH\cdot \rightarrow HO_2/O_2^- + H_2O$	3.30×10^7	b, c
(4) $Fe(III) + HO_2/O_2^- \rightarrow Fe(II) + O_2 + H^+$	7.82×10^5	b, c
(5) $Fe(II) + OH\cdot \rightarrow Fe(III) + OH^-$	3.20×10^8	b, c
(6) $Fe(II) + HO_2/O_2^- \rightarrow Fe(III) + H_2O_2$	1.34×10^6	b, c
(7) $HO_2/O_2^- + HO_2/O_2^- \rightarrow H_2O_2$	2.33×10^6	b, c
(8) $OH\cdot + HO_2/O_2^- \rightarrow H_2O + O_2$	7.15×10^9	b, c
(9) $OH\cdot + OH\cdot \rightarrow H_2O_2$	5.20×10^9	b, c

a: Le Truong and Laa, 2004; b: Duesterberg et al., 2005; c: Kwan and Voelker, 2002.

et al., 1985). Some organic compounds have been observed to act as electron transfer agents with iron (Baxendale et al., 1951; Hynes and Coinceanainn, 2001; Mentasti et al., 1973; Powell and Taylor, 1982; Xu and Jordan, 1988).

In this research, the effects of SDS as an anionic surfactant were investigated in the degradation of sorbed phenanthrene in sand by a Fenton-like reaction. SDS added during Fenton oxidation reacted with dissolved iron to precipitate a new composite. These samples were characterized by transmission electron microscopy (TEM), powder X-ray diffraction (PXRD), and Fourier-transform infrared (FT-IR) spectroscopy. Concentrations of H_2O_2 , Fe^{2+} , Fe^{3+} , and phenanthrene were measured to investigate the effect of SDS on the reaction.

2. Materials and methods

2.1. Materials

White silica sand (50–70 mesh) was obtained from Aldrich and was used without further treatment. 35% H_2O_2 was obtained from SHOWA, and all other chemical reagents were purchased from Aldrich.

2.2. Experimental procedures and analytical methods

2.2.1. Procedure for oxidation experiments

To determine the distribution of phenanthrene in sand, a stock solution of phenanthrene (500 mg) in hexane (1 L) was first prepared. Sand (2 g) was spiked with 0.5 mL of the stock solution. The hexane from this mixture was evaporated over one day to give an initial phenanthrene concentration of 125 mg/kg.

The Fenton type reactions were initiated by adding 0.5 mL of a 0.7% H_2O_2 solution after the addition of 0.5 mL of a $FeCl_3/FeCl_2$ and SDS solution. Initial concentrations of surfactant and Fe were determined to be 2–10 mM $FeCl_2/FeCl_3$ and 10–30 mM SDS. Most experiments were performed without buffers or ionic strength compensation to avoid potential complications due to reactions between a third additive and radical species. All tests were conducted in triplicate.

2.2.2. Analysis

2.2.2.1. Total phenanthrene concentration. After the oxidation experiments, the remaining phenanthrene was extracted by sonication in acetone. The experiment vials was added 20 mL of acetone and the contents were vortexed for 2 min, then immersed in an ultrasonic bath for 1 h. After sonication, the residual water was removed by the addition of anhydrous sodium sulfate, and the extracted phenanthrene was analyzed by HPLC. This method could extract more than 80% of residual phenanthrene (Kanel et al., 2003).

2.2.2.2. Dissolved phenanthrene concentration. Control experiments were conducted without the addition of H_2O_2 to investigate the possibility that phenanthrene could simply be dissolved by SDS. The procedure for these experiments was equivalent to that of the oxidation experiments. Samples were collected 3 h after starting the experiments. 0.5 mL samples were withdrawn through the septa of the serum bottles using a gastight glass syringe. A 0.22 μm disposable Luer-lock filter was placed between the needle and the syringe, and 0.05 mL of the filtrate was immediately transferred from the syringe into the 5 mL vials containing 1 mL of a solution consisting of hexane for phenanthrene extraction. An 85% acetonitrile/15% water solution was used as the mobile phase for HPLC analysis. Extracts were analyzed by HPLC using a fluorescence detector. The excitation level for the fluorescence detector was set at 254 nm and emission level at 390 nm.

2.2.2.3. H_2O_2 . H_2O_2 was measured using a modified peroxytitanic acid colorimetric procedure. A titanium sulfate solution was prepared by adding 0.6 g of titanium sulfate to 100 mL of concentrated sulfate solution. Here, a 0.9 mL of sample (0–3 mM) was added to 0.1 mL of titanium sulfate ($TiSO_4$) solution and allowed to react for 1 h. The absorbance of the H_2O_2 – $TiSO_4$ mixture was measured at 405 nm with a spectrophotometer. The samples were filtered from slurries (Gelman 0.2 μm) and prepared in triplicate.

2.2.2.4. Fe^{2+} and Fe^{3+} . After the oxidation tests, the filtered 0.5 mL samples (from the phenanthrene concentration analysis) were diluted by the addition of 4.5 mL of DI water. These diluted samples were used for analysis by UV-spectrophotometry. The Fe^{2+} and Fe^{3+} concentrations in the filtered supernatants were obtained by measuring the light absorption of its colored complex produced by the ferrozine method (Viollier et al., 2000).

2.2.2.5. Characterizations of precipitated Fe/surfactant composite. The analysis method and results were described in supplementary data.

3. Results and discussion

3.1. Effects of SDS

The increase in phenanthrene solubility with 20–50 mM SDS (Fig. 1) obviously improved degradation of phenanthrene

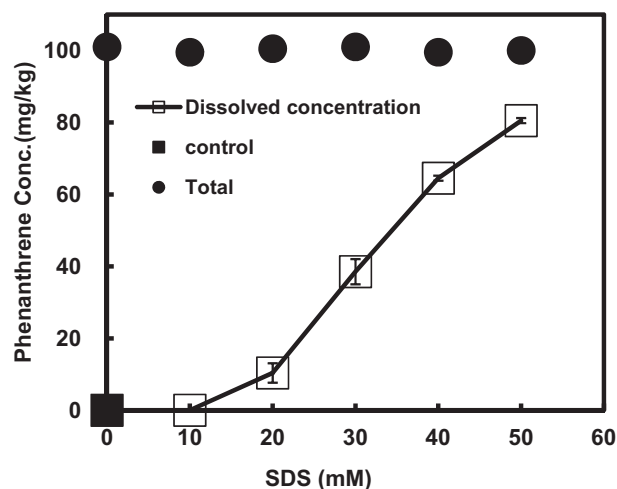


Fig. 1. Possible dissolved concentration of phenanthrene from sand in experiments of SDS dose with 4 mM $FeCl_2$ after 3 h (Values are the means of 3 replicates with standard deviation).

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