



Degradation of phthalate esters and acetaminophen in river sediments using the electrokinetic process integrated with a novel Fenton-like process catalyzed by nanoscale schwertmannite



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HIGHLIGHTS

- Novel oxidant injection was effective oxidation contaminants in river sediment.
- The electrode polarity reversal would maintain neutral pH for sediment.
- An extension of treatment time is beneficial to the removal efficiency.

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ABSTRACT

The main objective of this study was to develop and establish an *in situ* remediation technology coupling nano-schwertmannite/H₂O₂ process and electrokinetic (EK) process for the removal of phthalates (PAEs) and acetaminophen in river sediments. Test results are given as follows: (1) injection of nano-schwertmannite slurry and H₂O₂ (collectively, “novel oxidant”) into the anode reservoir would yield ·OH radicals that then will be diffused into the sediment compartment and further transported by the electroosmotic flow and/or electrophoresis from the anode end toward the cathode to degrade PAEs and pharmaceuticals in the sediment if any; (2) an electric potential gradient of 1.5 V cm⁻¹ would help the removal of PAEs and acetaminophen in the blank test, which no “novel oxidants” was added to the remediation system; (3) the practice of electrode polarity reversal would maintain neutral pH for sediment after remediation; (4) injection of equally divided dose of 10 mL novel oxidant into the anode reservoir and four injection ports on the top of sediment chamber would further enhance the removal efficiency; and (5) an extension of treatment time from 14 d to 28 d is beneficial to the removal efficiency as expected. In comparison, the remediation performance obtained by the EK-assisted nano-SHM/H₂O₂ oxidation process is superior to that of the batch degradation test, but is comparable with other EK integrated technologies for the treatment of same contaminants. Thus, it is expected that the EK-assisted nano-SHM/H₂O₂ oxidation process is a viable technology for the removal of phthalate esters and pharmaceuticals from river sediments in large-scale operations.

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1. Introduction

In the past decades, emerging contaminants (ECs) have received much attention by researches over the world. ECs include endocrine disruption compounds (e.g., bisphenol A), phthalate esters (PAEs), pharmaceuticals and personal care products (PPCPs), nanoscale contaminants, some microbes, etc. Nowadays, plastic

products are widely used in our daily life. PAEs are not only served as important additives which impart flexibility in plastic resins, but also used as solvents in paint, personal hygienic products, cleaning products, and cosmetics (ECB, 2003, 2004, 2006). Several PAEs are considered as hazardous pollutants due to their mutagenicity and carcinogenicity. These chemicals may pose risks to human health due to their xeno-estrogenic effects, and carcinogenic and mutagenic effects (ATSDR, 1995, 1997, 2001, 2002). PPCPs describe a large class of chemical contaminants that can originate from human usage and excretions, and veterinary applications of a variety of

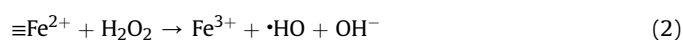
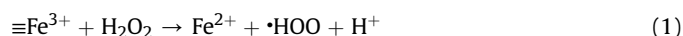
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products, such as over-the-counter and prescription medications, and fungicides and disinfectants used for industrial, domestic, agricultural and livestock practices (Richardson and Ternes, 2011; Richardson, 2012). PAEs and PPCPs have been detected in sediments around the world including Dianbao River in Taiwan (Yang et al., 2015a), and their concentrations were in the range of ND–881.18 mg kg⁻¹ dw and ND–0.51 mg kg⁻¹ dw, respectively (Table SM-1). Thus, it is necessary to develop appropriate technologies for the remediation of PAEs and PPCPs contaminated sediment.

Among others, advanced oxidation processes (AOPs) are commonly used remediation technologies for hazardous, refractory and non-biodegradable organic pollutants. Theoretically, AOPs can generate active oxygen species and further destroy the structure of organic compounds and mineralize them to carbon dioxide and water (Herney-Ramirez et al., 2010). Typical AOPs include catalytic ozonation (Qi et al., 2012), photodegradation (Kambur et al., 2012), ultraviolet irradiation (Xu et al., 2010), and Fenton process (Pinto et al., 2012). Of which, Fenton process is commonly employed because of its low toxicity of reagents, relatively low cost, high performance, and simplicity of technology (Homem et al., 2010). Thus, Fenton process [Fe(II)/H₂O₂] is a promising method for the removal of ECs in aqueous solutions and other environmental media such as soils, sediments or sludges (Neyens and Baeyens, 2003).

However, Fenton process also has some limitations such as difficulty in reuse of Fe²⁺ and maintenance of low pHs (≤3), and generation of a large amount of iron sludge needed to be properly treated (Yuan et al., 2013). As compared with conventional Fenton process, Fenton-like process can solve these problems by using heterogeneous catalysts like goethite (Watts et al., 1990). Generally, Fenton-like system uses a solid catalyst, which can maintain its effectiveness over a wide pH range for the oxidative treatment of organic pollutants. •OH radicals generated as a result of H₂O₂ catalysis by Fe²⁺/Fe³⁺ in Fenton/Fenton-like processes are capable of non-selectively degrading and mineralizing organic pollutants to carbon dioxide and water (Sirés et al., 2007; Son et al., 2009). The decomposition of hydrogen peroxide (H₂O₂) in heterogeneous Fenton-like reaction was suggested according to the Eq. (1) and Eq. (2) where ≡ represents the iron species bound to the surface of catalyst support (Hassan and Hameed, 2011):



In the past, many heterogeneous catalysts such as zero-valent iron (Son et al., 2010), iron oxides (Xu and Wang, 2012) and other iron-containing materials have been studied in Fenton-like process (Gao et al., 2013). Zhang et al. (2014) studied the degradation of 4-chlorophenol (4-CP) using a novel Fe(III)-containing polyoxometalate as the catalyst. It was found that the complete removal of total organic carbon (TOC) for 100 mg l⁻¹ 4-CP solution could be achieved. Other than the aforementioned iron-containing materials, Wang et al. (2013) used schwertmannite (SHM) as a new Fenton-like catalyst in the oxidation of phenol by H₂O₂. It was found that 98% phenol removal could still be obtained even after SHM was successively used for 12 cycles. Very recently, Duan et al. (2016) reported using SHM as a heterogeneous Fenton-like catalyst for the degradation of nitrobenzene (NB) in a closed batch system. The test results showed that the NB removal was significantly increased in the presence of SHM and the catalyst retained its high catalytic activity even after 5 consecutive runs. Apparently, SHM/

H₂O₂ oxidation process is a promising technology for the treatment of various organic contaminants.

Schwertmannite {SHM; [Fe₈O₈(OH)_{8-2x}(SO₄)_x, where x typically spans 1–1.75]} is a nanoscale and poorly-ordered Fe(III)-oxyhydroxy-sulfate mineral commonly found in acid sulfate soil (ASS) and acid mine drainage (AMD) environments (Bigham and Nordstrom, 2000; Sullivan and Bush, 2004). SHM readily undergoes phase transformation to more crystalline Fe(III) (hydr) oxides such as goethite under some pHs and oxidizing conditions (Eq. (3)) (Antelo et al., 2012). Therefore, SHM has a longer effective duration than goethite for Fenton-like process (Wang et al., 2013). This mineral not only has an amorphous structure with a high specific surface area, but also possesses a high surface reactivity rendering the mobility hindrance of trace metal and metalloid (Bigham et al., 1990; Burgos et al., 2011).



SHM can be synthesized by several techniques: dialysis by adding ferric chloride/nitrate to sodium/potassium sulfate solutions at 60 °C, oxidation of FeSO₄ solutions by H₂O₂, and bio-oxidation of FeSO₄ solutions by acidithiobacillus ferrooxidans (Bigham et al., 1990; Regenspurg et al., 2004; Liao et al., 2009). Wang et al. (2013) reported that SHM had a good catalytic activity in the oxidation of phenol via an •OH radical mechanism. The free radicals could be generated on the SHM surface by Fe(III) species and in bulk solution by dissolved Fe(III) over a wide pH range (Wang et al., 2013).

Electrokinetic remediation (i.e., EK process) is a promising technology to remediate fine-grained soil contaminated by inorganic, organic, and mixed contaminants. The EK process involves passing a low electric current between electrode pairs embedded in the ground for the removal of subsurface contaminants via electroosmosis, electromigration, and electrophoresis. Moreover, the EK process can be used in combination with many other cleanup techniques (Yang and Long, 1999; Yang and Liu, 2001; Virkutyte and Silanpää, 2007; Li et al., 2010; Colacicco et al., 2010).

To date many lab-scale and field studies employing chemical oxidation and EK process for remediation of different contaminants (e.g., heavy metals, polycyclic aromatic hydrocarbons, and polychlorinated biphenyls) in sediments can be found in the literature (De Giannis et al., 2009; Wittle et al., 2009; Mulligan et al., 2010). To the best knowledge of the present authors, however, to date no other research groups except the present authors' group has reported the remediation of phthalates in sediment using the EK integrated technologies. EK coupled with nano-Fe₃O₄/S₂O₈²⁻ oxidation has been employed for remediation of phthalate esters in river sediment (Yang et al., 2015b). Very recently, the same research group has further reported the remediation of phthalates in river sediment by integrated enhanced bioremediation and electrokinetic process (Yang et al., 2016).

As indicated above, SHM has been used as a new catalyst in Fenton-like process for the oxidation of phenol (Wang et al., 2013) and nitrobenzene (Duan et al., 2016) in batch experiments using some types of continuous stirred tank reactor (CSTR). To date, no study on coupling SHM/H₂O₂ oxidation with a column reactor for degradation of any organic compounds has been reported. In fact, the electrokinetic remediation system is involved in multiphase transport in a porous media under an external electric field that is much more complex than an ordinary plug flow reactor. To meet this need, researches in this regard are of significance. The objectives of this study are two-fold: (1) to develop and establish a feasible *in situ* remediation technology coupling nano-SHM/H₂O₂

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