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Electrochemical transformation of trichloroethylene in aqueous solution by electrode polarity reversal

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ABSTRACT

Electrode polarity reversal is evaluated for electrochemical transformation of trichloroethylene (TCE) in aqueous solution using flow-through reactors with mixed metal oxide electrodes and Pd catalyst. The study tests the hypothesis that optimizing electrode polarity reversal will generate H_2O_2 in Pd presence in the system. The effect of polarity reversal frequency, duration of the polarity reversal intervals, current intensity and TCE concentration on TCE removal rate and removal mechanism were evaluated. TCE removal efficiencies under 6 cycles h^{-1} were similar in the presence of Pd catalyst (50.3%) and without Pd catalyst (49.8%), indicating that Pd has limited impact on TCE degradation under these conditions. The overall removal efficacies after 60 min treatment under polarity reversal frequencies of 6, 10, 15, 30 and 90 cycles h^{-1} were 50.3%, 56.3%, 69.3%, 34.7% and 23.4%, respectively. Increasing the frequency of polarity reversal increases TCE removal as long as sufficient charge is produced during each cycle for the reaction at the electrode. Electrode polarity reversal shifts oxidation/reduction and reduction/oxidation sequences in the system. The optimized polarity reversal frequency (15 cycles h^{-1} at 60 mA) enables two reaction zones formation where reduction/oxidation occurs at each electrode surface.

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1. Introduction

Trichloroethylene (TCE), a toxic chlorinated solvent, has been widely used as a degreasing agent, dry cleaning solvent and chemical extraction agent. Inappropriate disposal of chlorinated solvents have caused widespread groundwater TCE contamination. TCE is listed by the US Environmental Protection Agency (US EPA) as a priority pollutant and with a Maximum Contaminant Limit of 5 $\mu g\,L^{-1}$. Methods for removal of TCE from contaminated groundwater include biological transformation (e.g., Li et al., 2011; Semkiw and Barcelona, 2011; Tiehm and Schmidt, 2011), chemical oxidation (e.g., Teel et al., 2001; Yamazaki et al., 2001; Waldemer et al., 2007; Lee and Lee, 2010; Tsai et al., 2011; Yue-hua et al., 2011, Yuan et al., 2012a) reduction by zero valent iron (e.g., Li et al.,

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2003; Liu et al., 2006; Phillips et al., 2010; Petersen et al., 2012) and palladium-based materials (e.g., Lowry and Reinhard, 2001; Ma et al., 2012; Lin et al., 2009; He et al., 2007; Wu and Ritchie, 2006), photocatalytic degradation using TiO_2 catalyst (e.g., Che et al., 2011; Farooq et al., 2009) and ultrasonically enhanced oxidation (e.g., Rashid and Sato, 2012; Ayyildiz et al., 2007; Destaillats et al., 2001). Although these methods have advantages, there are limitations including high operating cost, long reaction time, and incomplete removal or accumulation of toxic byproducts.

Electrochemical transformation of dissolved organics has attracted a considerable interest due to the method's ability to control and manipulate groundwater redox conditions (e.g., Alshawabkeh and Sarahney, 2005; Petersen et al., 2007; Li and Farrell 2000, Mishra et al., 2008; Alshawabkeh, 2009; Gilbert et al., 2010; Scialdone et al., 2010; Mao et al., 2011; Lakshmipathiraj et al., 2012; Mao et al., 2012a, 2012b; Yuan et al., 2012a,b, Yuan et al. 2013). Anode reactions can induce direct electrolytic oxidation of the target compound and water oxidation. Cathode reactions can induce direct electrolytic reduction of the target compounds and water reduction. Aqueous species can be oxidized or reduced at the electrodes depending on the half-reaction potentials and influence target contaminant degradation pathway and removal rates. Given the highly oxidized nature of TCE, reduction has been considered the preferred degradation mechanism. Therefore, most of the literature has focused on cathodic conversion of chlorinated aliphatic hydrocarbons to the corresponding dehalogenated hydrocarbons (e.g., Petersen et al., 2007; Li and Farrell 2000, Mishra et al., 2008; Mao et al., 2011, 2012a, 2012b). Different cathode (e.g., Petersen et al., 2007; Mao et al., 2012b) and anode materials (Mao et al., 2011) were investigated to improve TCE reduction. Palladized electrodes are reported to significantly improve electrochemical TCE reduction (e.g., Roh et al., 2001; Chen et al., 2003; Li and Farrell 2000).

A few studies focus on electrochemical oxidation for TCE removal (e.g., Yuan et al., 2012b; Lakshmipathiraj et al., 2012; Yuan et al., 2013). Several anodic materials have been tested for chlorinated hydrocarbons electrooxidation, such as Pt, Au, Ebonex®, PbO_2, boron-doped diamond, graphite, SnO_2, and mixed metal oxide (MMO) electrodes (e.g., Scialdone et al., 2008, 2010). Among the MMO electrodes, IrO_2-based anodes, and in particular the binary system IrO_2–Ta_2O_5, are reported to exhibit good performance in anodic stability and electrocatalytic activity (e.g., Scialdone et al., 2008, 2009). TCE is oxidized on the MMO anode surface (above 0.84 V vs. SCE – saturated calomel electrode) according to the following reactions (Lakshmipathiraj et al., 2012).

$$IrO_2 + H_2O \leftrightarrow IrO_2(OH\cdot) + H^+ + e^- \quad (1)$$

$$IrO_2(OH\cdot) \leftrightarrow IrO_3 + H^+ + e^- \quad (2)$$

$$IrO_3 + TCE_{ads} \rightarrow IrO_2 + TCE - O_{ads} \quad (3)$$

However, oxygen evolution competes with TCE oxidation at the Ti/IrO_2–Ta_2O_5 electrode surface as given by Panizza and Cerisola (2009).

$$IrO_3 + H_2O \rightarrow IrO_2 + O_2 + 2H^+ + 2e^- \quad (4)$$

Pd can catalyze the decomposition of H_2O_2 in weak acidic conditions to a strong oxidizing •OH radical (Reaction (6)) (e.g., Yuan et al., 2012b, 2013). Furthermore, Pd catalyst can be used to enable electro-Fenton process for TCE oxidation when pH < 4. This is achieved in a flow-through reactor by employing three electrodes with current splitting. Oxidation supported by Pd catalyst is given by.

$$H_2 + O_2 \overset{Pd}{\longleftrightarrow} H_2O_2 \quad (5)$$

$$H_2O_2 \overset{Pd}{\longleftrightarrow} 2\cdot OH \quad (6)$$

$$H_2O_2 + Fe^{2+} + H^+ \rightarrow Fe^{3+} + \cdot OH + H_2O \quad (7)$$

$$TCE + \cdot OH \rightarrow \text{oxidation products} \quad (8)$$

Electrode polarity reversal is a technique that is based on defined time intervals of operation under reversed electrode polarity (e.g., Pazos et al., 2006; Luo et al., 2005). It has been investigated to control electrokinetic remediation of heterogeneous media. In groundwater treatment, the technique was used (in the presence of an additional electrode) to control electrode fouling due to CaO and $CaCO_3$ formation (e.g., Gilbert et al., 2010; Duarte et al., 1998). pH profile can be controlled by optimizing the electrode sequence and reversing polarity duration (Pazos et al., 2006). Both H_2 and O_2 could be generated in the Pd catalyst vicinity when changing the electrode polarity. While this process was used to control pH and chemical conditions, it was never evaluated for TCE removal from groundwater.

The objective of this study is to investigate and optimize TCE removal from groundwater using electrode polarity reversal. Two electrodes are used to support TCE removal in the presence of Pd catalysts (Equations (5), (6) and (8)). H_2O_2 formation and acidic condition evolution in Pd vicinity were measured to test if polarity reversal could support Fenton reaction in Fe^{2+} presence. The effect of specific parameters including polarity reversal frequency, duration of intervals during polarity reversal, groundwater chemistry, current intensity and TCE concentration is evaluated.

2. Materials and methods

2.1. Chemicals and materials

All chemicals used in this study were analytical grade. TCE (99.5%) and cis-dichloroethylene (cis-DCE, 97%) were purchased from Sigma–Aldrich. H_2O_2 (30%) was purchased from Fisher Sci. Calcium sulfate was purchased from JT Baker, oxalic acid (anhydrous, 98%) from Acros, sodium chloride and sodium bicarbonate from Fisher Scientific and hydrocarbon gas standard (analytical standard, 1% (w/w) methane, ethene, acetylene in nitrogen) from Supelco. Two mesh Ti/MMO electrodes (Ti/IrO_2–Ta_2O_5-3N International) were used as cathode and anode. The electrodes consist of IrO_2 and Ta_2O_5 coating on titanium mesh with dimensions of 1.8 cm diameter and 1.8 mm thickness. Deionized water (18.0 MΩ cm) obtained

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