



The influence of cathode material on electrochemical degradation of trichloroethylene in aqueous solution



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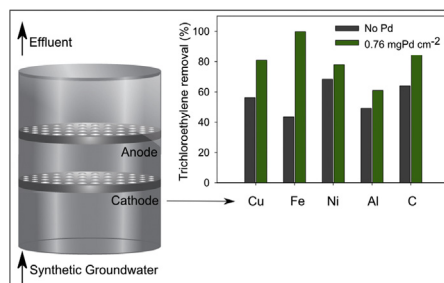
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HIGHLIGHTS

- The cathode material influences hydrodechlorination (HDC) rate of trichloroethylene.
- The effect of Pd coating is different for cathode materials tested.
- The Pd coating on the cathodes significantly enhances HDC of trichloroethylene.
- The presence of nitrates adversely influences the HDC of trichloroethylene.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 9 September 2015

Received in revised form

22 December 2015

Accepted 23 December 2015

Available online 4 January 2016

Handling Editor: E. Brillas

Keywords:

Electrochemical

Cathode

Palladium

Trichloroethylene

Groundwater

ABSTRACT

In this study, different cathode materials were evaluated for electrochemical degradation of aqueous phase trichloroethylene (TCE). A cathode followed by an anode electrode sequence was used to support reduction of TCE at the cathode via hydrodechlorination (HDC). The performance of iron (Fe), copper (Cu), nickel (Ni), aluminum (Al) and carbon (C) foam cathodes was evaluated. We tested commercially available foam materials, which provide large electrode surface area and important properties for field application of the technology. Ni foam cathode produced the highest TCE removal (68.4%) due to its high electrocatalytic activity for hydrogen generation and promotion of HDC. Different performances of the cathode materials originate from differences in the bond strength between atomic hydrogen and the material. With a higher electrocatalytic activity than Ni, Pd catalyst (used as cathode coating) increased TCE removal from 43.5% to 99.8% for Fe, from 56.2% to 79.6% for Cu, from 68.4% to 78.4% for Ni, from 42.0% to 63.6% for Al and from 64.9% to 86.2% for C cathode. The performance of the palladized Fe foam cathode was tested for degradation of TCE in the presence of nitrates, as another commonly found groundwater species. TCE removal decreased from 99% to 41.2% in presence of 100 mg L⁻¹ of nitrates due to the competition with TCE for HDC at the cathode.

The results indicate that the cathode material affects TCE removal rate while the Pd catalyst significantly enhances cathode activity to degrade TCE via HDC.

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1. Introduction

Trichloroethylene (TCE) has been widely used as an ingredient in industrial cleaning solutions and as a “universal” degreasing agent. Improper disposal of TCE coupled with its physical and

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chemical properties (low solubility and limited degradation) led to persistent TCE contamination at many hazardous waste sites (Moran et al., 2007). The USEPA has set Maximum Contaminant Levels (MCLs) for TCE in drinking water at very low concentrations of $5 \mu\text{g L}^{-1}$.

The presence of chlorine in the TCE structure contributes to its carcinogenic and mutagenic properties; thus, complete dechlorination is favorable for degradation of TCE dissolved in groundwater. So far, the efficient reductive dechlorination of chlorinated organic compounds (COCs) was achieved via zero-valent iron and palladium-based materials (Lowry and Reinhard, 2001; Lien and Zhang, 2005; Liu et al., 2006; Wu and Ritchie, 2006; Petersen et al., 2007a; Phillips et al., 2010; Ma et al., 2012; He et al., 2013).

Recently, electrochemically-induced reduction has gained interest for removal of COCs from groundwater (Li and Farrell, 2000; Petersen et al., 2007b; Mishra et al., 2008; Mao et al., 2011, 2012a, 2012b). Electrochemical reduction through hydrodechlorination (HDC) occurs at the cathode due to water electrolysis. The water electrolysis at the cathode leads to the hydrogen evolution via electrochemical hydrogen adsorption (Volmer reaction) where atomic hydrogen (H_a) is chemically adsorbed on active site of the electrode surface (M). The H_a further involves in electrochemical desorption (Heyrovsky reaction) or chemical desorption (Tafel reaction) to create hydrogen gas or interacts with the reducible chemicals like chlorinated substances, which leads to HDC. The dechlorination rate is affected by mass transfer, electron transfer and chemical reactions, as well as surface reactions, such as adsorption and desorption (Chaplin et al., 2012; He et al., 2013). Apart from electroreduction, electrochemically-induced oxidation of COCs can be achieved through reactions at the anode (Rodrigo et al., 2001) or via hydroxyl radicals generated in bulk solution (Azzam et al., 2000; Martinez-Huitile and Ferro, 2006; Panizza and Cerisola, 2009; Yuan et al., 2012, 2013). The oxidation rate at the anode highly depends on the anode material; with active anodes the oxygen production is promoted over formation of hydroxyl radicals and COCs oxidation is limited while non-active anodes (e.g. boron doped diamond) provide complete COCs transformation to CO_2 through reaction with hydroxyl radicals (Scialdone et al., 2010a; Randazzo et al., 2011). In mixed electrolyte systems, which are easier to implement and maintain in practice, the electrochemical degradation can be achieved by: a) reduction via HDC promoted by the use of iron anode (Mao et al., 2011, 2012a, 2012b), b) oxidation via Fenton reaction (Yuan et al., 2012, 2013) or b) simultaneous oxidation and reduction with the selection of appropriate electrode materials (Gilbert and Sale, 2005; Scialdone et al., 2010a). When using an active anode in mixed electrolyte system, the presence of oxygen competes with the target contaminant for the reaction sites at the cathode and limits HDC and it can involve in creation of hydrogen peroxide and promote Fenton reaction. A new system was designed to promote electrochemical reduction, which is the primary degradation mechanism of highly oxidized chemical compounds such as TCE. The design, introduced by our laboratory, adopts a flow-through cell with the cathode placed in front of the active anode with respect to the flow direction, to minimize the effect of oxygen on HDC of TCE (Rajic et al., 2014, 2015a, 2015b, 2015c).

The type of cathode material has a major effect on the HDC reaction. The good HDC catalyst should have strong bond with H_a to allow proton-electron transfer process but weak enough to ensure the bond breaking and the product release (Zheng et al., 2015). If the hydrogen-metal surface (H_a -M) binding energy is too high, adsorption is slow and limits the overall rate but if it is too low, desorption is slow. The standard free energy for H_a adsorption on solid electrode can be used to evaluate the adsorption of H_a and the product release. Good catalytic activity of noble metals (e.g., Ag, Pt

and Pd) arises from the fact that the free energy of hydrogen adsorption is close to zero (Zheng et al., 2015). It has been demonstrated that noble metals can be effectively used for the reductive dechlorination of chlorinated hydrocarbons when used as working electrodes, and as coatings on different electrode materials (Sonoyama and Sakata, 1999; Hoshi et al., 2004; Xu et al., 2007; Scialdone et al., 2008; Rondinini and Vertova, 2010; Scialdone et al., 2010b).

Specifically, palladized electrodes were widely investigated for removal of COCs (Li and Farrell, 2000; Roh et al., 2001; Jovanovic et al., 2005; Yang et al., 2006, 2007; He et al., 2013; Li et al., 2013). In this case, the deposition of small amounts of Pd catalyst on the cathode substrate enhances its ability to absorb hydrogen into the lattice and increases the rate of HDC. It was found that the hydrogen absorption is higher in thin Pd films than in the bulk metal; thin Pd films crystallites size is much smaller and more hydrogen may be located in the grain boundaries (Bucur and Bota, 1982; Stuhr et al., 1997; Gabrielli et al., 2004). Various permeable materials were used as cathode substrates for palladium coating, such as activated carbon fiber/cloth/felt, reticulated vitreous carbon, carbon nanotubes meshed Ti, Ti/TiO₂ nanotubes, silver, Ni foam, Fe foam and Ebonex[®] (Cheng et al., 1997, 2003; Yang et al., 2006, 2007; Durante et al., 2009; Scialdone et al., 2010b; Sun et al., 2010; Durante et al., 2013; Xie et al., 2013). Ni foam is known to be an ideal cathode substrate for hydrogenation catalyst; Cu is potentially a good catalyst for the hydrogenation for treatment of selected chlorinated aliphatic hydrocarbon (Zhu et al., 2010). However, slightly stronger adsorption of hydrogen by Cu and weaker adsorption by Ni and Fe leads to less electroactivity than noble metals (Zheng et al., 2015). It was found that in the bimetallic Pd/Ni foam system, the Pd particles at the cathode substrate influence on HDC rate by increasing the surface area and allowing the diffusion of H_a among particle, which maintain and regenerate electrocatalytic activity (Yang et al., 2007).

The influence of commonly found chemicals in groundwater, such as nitrates, is critically important to implement remediation technology. Nitrates are commonly found species in groundwater at high levels due to the intensive use of nitrogen fertilizers, industrial wastes, animal wastes and septic systems and present a significant environmental problem (Wakida and Lerner, 2005; Burkart and Stoner, 2007). Electrochemical reduction by palladized electrodes is found to be an efficient mechanism for the nitrates removal from water (de Vooys et al., 2000; Peel et al., 2003; Wang and Qu, 2006; Abdallah et al., 2014). The nitrates reduction mechanism initially involves interaction with the atomic hydrogen (Reactions 1–2) and is a competing reaction with HDC.



The main goal of this study is to evaluate the performance of different cathode materials for the electrochemical dechlorination of TCE in a mixed, undivided electrolyte system. The cathode followed by anode setup was used since it promotes HDC in the undivided, flow-through electrochemical cell. We tested the performance of Fe, Cu, Ni, Al and C foams and their palladized forms for degradation of TCE by a cathode upstream of an anode electrode sequence. These materials show good electrocatalytic properties towards dechlorination and are cost-effective and commercially available materials – important properties for possible filed application of the technology. In addition, the influence of nitrates on electroreduction of TCE in aqueous solution was also investigated.

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