



Transformation and removal of arsenic in groundwater by sequential anodic oxidation and electrocoagulation



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ABSTRACT

Oxidation of As(III) to As(V) is generally essential for the efficient remediation of As(III)-contaminated groundwater. The performance and mechanisms of As(III) oxidation by an as-synthesized active anode, SnO₂ loaded onto Ti-based TiO₂ nanotubes (Ti/TiO₂NTs/Sb–SnO₂), were investigated. The subsequent removal of total arsenic by electrocoagulation (EC) was further tested. The Ti/TiO₂NTs/Sb–SnO₂ anode showed a high and lasting electrochemical activity for As(III) oxidation. 6.67 μM As(III) in synthetic groundwater was completely oxidized to As(V) within 60 min at 50 mA. Direct electron transfer was mainly responsible at the current below 30 mA, while hydroxyl radicals contributed increasingly with the increase in the current above 30 mA. As(III) oxidation was moderately inhibited by the presence of bicarbonate (20 mM), while was dramatically increased with increasing the concentration of chloride (0–10 mM). After the complete oxidation of As(III) to As(V), total arsenic was efficiently removed by EC in the same reactor by reversing electrode polarity. The removal efficiency increased with increasing the current but decreased by the presence of phosphate and silica. Anodic oxidation represents an effective pretreatment approach to increasing EC removal of As(III) in groundwater under O₂-limited conditions.

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1. Introduction

Contamination of groundwater by arsenic has been a global environmental problem due to geological and anthropogenic reasons (Fendorf et al., 2010; Neumann et al., 2010; Nordstrom, 2002; Rodríguez-Lado et al., 2013). Long-term consumption of groundwater contaminated with above 10 μg/L arsenic can cause irreversible damage to a variety of organs and systems of the body's functions (Smedley and Kinniburgh, 2002). The health of over 100 million people is threatened by arsenic contamination in the world (Fendorf et al., 2010). Due to the high toxicity of arsenic, the World Health Organization (WHO) sets the maximum acceptable level in drinking water at 10 μg/L (Sharma and Sohn, 2009; van Geen, 2008). It is therefore

urgent to develop cost-effective processes for removing arsenic in contaminated groundwater.

In anoxic groundwater, As(III) is the predominant As species. As(III) is much more toxic than As(V) (Sharma and Sohn, 2009), but it is generally more difficult to remove from waters than As(V) by commonly available treatment processes (Rahman et al., 2003). Therefore, preoxidation of As(III) to As(V) is necessary to enhance the removal (Lee et al., 2003). Chlorine, ozone, KMnO₄, photocatalysis and so on have been used for oxidizing As(III) (Mondal et al., 2013). As an environmentally friendly advanced oxidation process, anodic oxidation has been intensively used for treating refractory organic pollutants in the past decades (Bunce et al., 1997; Vaghela et al., 2005; Vlyssides et al., 2004; Zhao et al., 2009). Nevertheless, for the practical application of anodic oxidation for in situ transforming As(III) in groundwater, it is prerequisite that the anode has a high mechanical stability and electrochemical activity. Dimensional stable anodes (DSAs) are the

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most commonly used anode in chemical industrial processes. DSAs have been widely used in treating organics contaminated wastewaters (Scialdone et al., 2009; Wei et al., 2011), but are seldom tested for As(III) oxidation. According to our knowledge, Zhao et al. (2010, 2011) reported that As(III) removal by electrocoagulation (EC) could be enhanced by the simultaneous oxidation of As(III) on a commercial DSA. However, the mechanism of enhancement was not elucidated because of the complicated influence of O₂ (Hug and Leupin, 2003; Li et al., 2012; Pang et al., 2011). Lacasa et al. (2012) found that a DSA could oxidize As(III) completely at large current densities. Direct and indirect oxidation was proposed as the transformation mechanism, but the relative contributions were not quantified (Lacasa et al., 2012).

In the previous investigations (Lacasa et al., 2012; Zhao et al., 2010, 2011), the mechanisms of As(III) oxidation are not well understood. In recent years, a new type of DSAs, active components loaded onto Ti-based TiO₂ nanotubes (Ti/TiO₂NTs) instead of Ti plate, shows better performance than commercial DSAs in wastewater treatment (Li et al., 2009; Quan et al., 2005; Zhao et al., 2009). The active components are tightly bound to the nanotubes, implying a lasting electrochemical activity. This offers the possibility of application in groundwater remediation. However, the performance and mechanisms of this new anode for oxidizing As(III) have not been investigated.

In this study, we will load the active components of SnO₂ and Sb₂O₅ onto Ti/TiO₂NTs arrays (Ti/TiO₂NTs/Sb–SnO₂) and use it as an active anode for oxidizing As(III) in groundwater. Upon the complete oxidation of As(III) to As(V), EC is tested for the subsequent removal of total arsenic because of the high efficiency (Kumar et al., 2004; Li et al., 2012; Wan et al., 2011). Although As(III) can be oxidized in EC when exposed to O₂ or air (Kumar et al., 2004; Li et al., 2012; Wan et al., 2011), the oxidation is minimal when operated in a deep tank or in O₂-limited subsurface. The focus of this study is paid to the oxidation of As(III) by the Ti/TiO₂NTs/Sb–SnO₂ anode as EC has been extensively investigated for As removal. The main objectives are (1) to test the performance of the Ti/TiO₂NTs/Sb–SnO₂ anode on As(III) oxidation under different conditions, (2) to quantify the relative contributions of direct electron transfer and hydroxyl radicals to As(III) oxidation under different currents, and (3) to evaluate the efficiency of sequential anodic oxidation and EC for As(III) removal in groundwater.

2. Materials and methods

2.1. Chemicals

As₂O₃ (99.8%) was purchased from Shanghai General Reagent Factory, China. Na₂HAsO₄·7H₂O (99.99%) was obtained from Sigma-Aldrich. Ti plates (2 cm length, 1 cm width and 1 mm thickness), graphite (2 cm length, 1 cm width and 1 cm thickness), and mixed metal oxides (MMO, IrO₂ mixed Ta₂O₅ coating on titanium diamond mesh, Shanxi Kaida Chemical Ltd.) with dimensions of 58 mm length, 50 mm width and 1.7 mm thickness, Pt plates (2 cm length, 2 cm width and 0.1 mm thickness, Wuhan Corrtest, China), Fe plates (6 cm length, 2 cm width and 2 mm thickness, S45C type, Wuhan Steel Processing Co., Ltd.) were used as

electrodes. Twelve holes (4.1 mm in diameter) are evenly distributed in one iron plate. Prior to the experiments, the iron plate was polished with coarse emery cloth, etched by diluted HCl solution (5 wt.%), and washed with deionized (DI) water. DI water (18.2 MΩ·cm) obtained from a Heal Force NW ultra-pure water system was used in all the experiments. All the other chemicals are above analytical grade.

2.2. Synthesis and characterization of Ti/TiO₂NTs/Sb–SnO₂ electrode

The highly ordered TiO₂NTs were synthesized by anodization of a Ti plate in an NH₄F electrolyte according to the literature (Prakasam et al., 2007). The Ti plate was mechanically polished with different abrasive papers and cleaned with DI water in an ultrasonic bath (200 W, 40 KHz) for 10 min, followed by air drying. The polished plate was immersed in a mixed acids solution (V_{HF} = 10 mL, V_{HNO₃} = 40 mL, V_{H₂O} = 50 mL) for 25 min and washed sequentially in DI water, acetone and DI water for 10 min each in the ultrasonic bath. Then the Ti plate was used as the anode and an MMO was used as the cathode with 4 cm spacing in parallel. The electrolyte consisted of 300 mL of glycerine, 2 g of NH₄F and 10 mL of DI water. A constant voltage of 35 V was supplied by a DC power source (GPC-6030D, Taiwan Guwei Electronic Ltd., Inc., Taiwan). The anodization lasted for 6 h in the ultrasonic bath. After washing with DI water, the as-formed Ti/TiO₂NTs were calcined in a muffle burner at 500 °C for 2 h.

The active component of SnO₂ nanoparticles was loaded onto the as-formed Ti/TiO₂NTs by sol–gel method (Kotz et al., 1991; Zhao et al., 2009). 0.723 g of SbCl₃ and 10 g of SnCl₄·5H₂O were added to the mixed solution of 100-mL ethanol and 10-mL hydrochloric acid. The mixtures were mechanically stirred at 80 °C for 2 h in a three-neck flask. The formed sol–gel was aged for 9 days at room temperature to increase concentration. The as-formed Ti/TiO₂NTs were slowly dipped into the sol–gel 5 times. The Ti/TiO₂NTs loading sol–gel were dried at 100 °C for 10 min and calcined at 500 °C for 10 min. This procedure was repeated 10 times. After annealing at 500 °C for 1 h, the Ti/TiO₂NTs/Sb–SnO₂ anode was obtained.

2.3. Performance of Ti/TiO₂NTs/Sb–SnO₂ anode in oxidizing As(III)

The oxidation of As(III) by the as-synthesized Ti/TiO₂NTs/Sb–SnO₂ anode was first tested by CV on a CS150 electrochemical workstation (Wuhan CorrTest Instrument, China). A Pt plate (2 × 2 cm) was used as the counter electrode and a saturated calomel electrode (SCE) was used as the reference electrode. The electrolytes consisted of 133 μM As(III) and 0.1 M Na₂SO₄ at pH 7. The solution was stirred at 200 rpm. The CVs were scanned at different rates (20, 50, 100, 200 and 400 mV/s). Ti/TiO₂NTs/Sb–SnO₂ was replaced by different electrodes for comparison. For the measurement of oxygen and chlorine evolution potentials, the CVs were scanned at 100 mV/s in 0.1 M Na₂SO₄ and 1 M NaCl, respectively.

The anodic oxidation of As(III) in synthetic groundwater was conducted in an undivided electrolytic cell (Fig. S1). The as-synthesized Ti/TiO₂NTs/Sb–SnO₂ was used as the anode, and an Fe plate was used as the cathode. For each trial,

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