



Remediation of ammonia-contaminated groundwater in landfill sites with electrochemical reactive barriers: A bench scale study

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ABSTRACT

Leachate plumes originating from leaking landfills often cause the contamination of groundwater in subsurface. Ammonia nitrogen in the contaminated groundwater is usually hard to be attenuated due to the hypoxic condition in subsurface environment. In this study, an active chlorine mediated electrochemical reactive barrier (ACM-ERB) consisting of inert electrodes is proposed for the remediation of ammonia-contaminated groundwater because an elevated level of chloride ions was often found in the groundwater polluted by leachate plumes. Bench-scale experiments were conducted to evaluate the prototype of this remedial technology and to study the variables affecting the performance of ACM-ERB. The results showed that ammonia in the simulated groundwater can be effectively converted into nitrogen rather than undesirable nitrite/nitrate. RuO₂/Ti anode was better than PbO₂/Ti anode for the sake of ammonia removal. In the presence of naturally occurring level of bicarbonate, the electrode arrangement with an upstream cathode offered weak alkaline pH and therefore favored the removal of ammonia in the initial stage of experiment. Higher current densities and bicarbonate concentrations were favorable to the removal of ammonia. An ammonia removal efficiency up to 70% was achieved for 20 mg/L NH₄⁺-N influent, when the operating conditions were 250 mg/L chloride ions, 500 mA current, –80 mm water level and 6 mL/min flow rate. Polarity reversal could prevent the formation of scale on electrodes, thereby allowing the long-term operation of the ACM-ERB system in groundwater. Moreover, in the experiment using diluted leachate as influent solution, ammonia was preferentially removed relative to the organic contaminants. The present study demonstrates that ACM-ERB is a promising method to cope with the ammonia-contaminated groundwater in landfill sites.

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1. Introduction

Pollution of groundwater is probably the most severe environmental risk of landfill sites because landfill leachate may leak and cause the spread of various contaminants in subsurface. Generally, landfill leachate contains various pollutants, such as undesirable heavy metals (Fatta et al., 1999; Loizidou and Kapetanios, 1993; Mor et al., 2006; Murray et al., 1981), organic matter, ammonium and inorganic macrocomponents for groundwater (e.g., chloride anions and calcium cations). Based on the existing data regarding the groundwater quality of landfill sites, dissolved organic matter and ammonia were the typical contaminants due to their strong mobility in groundwater environment. Dissolved

organic matter, quantified as Chemical Oxygen Demand (COD) or Total Organic Carbon (TOC), generally includes volatile fatty acids, dissolved humic-like compounds, and xenobiotic organic compounds originating from household or industrial chemicals. In the suboxic or hypoxic environment of subsurface, COD can be naturally attenuated via anaerobic methanogenic process. However, the ammonia in groundwater is hard to be biologically transformed via reductive process because the nitrogen in ammonia has the lowest and most stable reductive valence of –3. As a result, many landfill sites showed elevated ammonia nitrogen in groundwater because of the leakage and transport of landfill leachate (Alsabahi et al., 2009; Husain et al., 1989; Mor et al., 2006).

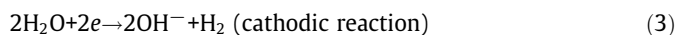
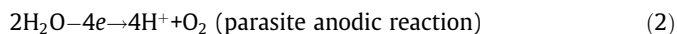
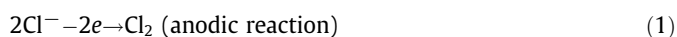
The most well-developed method for removing ammonia from wastewater or surface water is the simultaneous nitrification and denitrification (SND) process associated with the functioning of nitrifying and denitrifying bacteria. Normally, nitrification of

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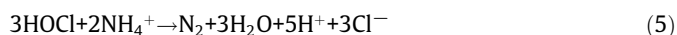
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ammonia is an autotrophic biological process that uses oxygen as electron acceptors; and the subsequent denitrification process normally needs extra carbon sources (or other reductive substances such as sulfur and hydrogen) as electron donors to achieve the conversion from nitrite/nitrate to nitrogen gas. However, for the remediation of ammonia-contaminated groundwater, it is not practical and cost-effective if both dissolved oxygen and electron donors in groundwater are required to be regulated in order to accomplish the transformation of ammonia nitrogen. In addition to the biological transformation, other processes including adsorption, ion-exchange (Li et al., 2009) and chemical oxidation (breakpoint chlorination method) (Chen et al., 2007) are also effective for the removal of ammonium nitrogen from aqueous solution. Nevertheless, these technologies are not suitable for the remediation of ammonia-contaminated groundwater in landfill sites, especially for long-term application of in-situ remediation.

Electrochemical redox systems consisting of anode and cathode have been investigated as an option to treat different contaminants in groundwater, such as chlorinated solvents, energetic compounds (Gent et al., 2009; Gilbert and Sale, 2005) and heavy metal ions. Electrochemical processes have the advantages of simple operation, easy-automation, fast response and versatility, being a promising technique for the in-situ remediation of contaminated groundwater (Chen, 2004; Ding et al., 2014; Janssen and Koene, 2002; Li et al., 2017). Previous studies have demonstrated the effectiveness of an electrically induced redox barrier for the in situ remediation of sites contaminated with trichloroethylene and trichloromethane (Sale, 2005). Our studies also showed that a solar cell driven electrochemical system consisting of iron anode could be used for the removal of various contaminants, such as TCE, Cr(VI) and nitrate, from simulated groundwater (Mao et al., 2012). Generally, the electrolytic ferrous ions and the direct electrochemical reduction on cathode offered the decontaminating capability for the electrochemical system. For the remediation of ammonia-contaminated groundwater, however, a different electrochemical system has to be redesigned to achieve the transformation of ammonia. According to the existing data, the polluted groundwater in landfill sites basically showed an elevated level of chloride ions, ranging from several hundred ppm to a thousand ppm (Fatta et al., 1999; Han et al., 2014; Mor et al., 2006). This important information inspires us that an electrochemical system that produces active chlorine (chlorine/hypochlorous acid/hypochlorite) can be constructed to remediate the ammonia-contaminated groundwater in the subsurface beneath a landfill site. For this system, the reactions at electrodes are shown as below (Kim et al., 2005; Lin and Wu, 1996; Trasatti, 1984):



As such, the following chemical oxidation reactions can occur in the solution, resulting in the transformation of ammonia nitrogen.



The previous studies revealed that the oxidation of ammonia by electrogenerated active chlorine was affected by a series of factors, such as electrode materials (Ma et al., 2012; Szpyrkowicz et al., 2005), current density (Chiang et al., 1995; Vanlangendonck et al., 2005), initial pH (Chiang et al., 1995; Lin and Wu, 1996; Vanlangendonck et al., 2005), Cl^- concentration and coexisting ions (Li et al., 2010; Vanlangendonck et al., 2005). However, the

oxidative transformation of ammonia nitrogen by active chlorine has not been investigated under the circumstance of groundwater remediation. In this study, a bench-scale reactor is designed to mimic the condition of a shallow aquifer contaminated by ammonia nitrogen, and an active chlorine mediated electrochemical reactive barrier (ACM-ERB) is set up in the reactor to investigate the effectiveness of the electrochemical oxidation on the remediation of ammonia nitrogen. We hope that the present study could give us clues about the applicability of the electrochemical active-chlorine process on the in-situ remediation of ammonia-contaminated groundwater.

2. Experimental

2.1. Chemicals and materials

The chemical reagents including sodium sulfate, sodium chloride, ammonia sulfate, sodium bicarbonate, calcium sulfate, sodium hyposulfite, potassium sodium tartrate, potassium iodide, mercuric iodide and sodium hydroxide, were purchased from the Shanghai Chemical Reagent Corporation, China. The packed reagents for the measurement of active chlorine, total chlorine, nitrite, and COD were provided by the Hach Company. The electrode materials including RuO_2/Ti and PbO_2/Ti (titanium mesh substrate with a size of 170 mm long and 50 mm wide) were purchased from Zhiming Special Metals Company, Shanxi, China. The cathodes of all trials were RuO_2/Ti , and the anodes were RuO_2/Ti or PbO_2/Ti , respectively.

2.2. Experimental reactor and methods

The electrochemical oxidation of ammonia by electrochemical reactive barrier was carried out in a tank reactor, as shown in Fig. 1. The tank reactor, filled with quartz sands (4–8 mm particle size), was made of plexiglass with a dimension of 600 mm in length, 50 mm in width, 185 mm in height. A power source supplied constant current for the built-in electrode pair. The anode and cathode were fixed by plastic screws and nuts, and their distance was 5 mm. The electrode pair was perpendicular to the direction of the flow. Reaction area of electrode depended on the water level: 50 cm^2 for –80 mm water level and 80 cm^2 for –50 mm water level. The influent solution containing ammonia was pumped from the holding bottle to the inlet via a peristaltic pump (BT100-2J, Longerpump, China), and flowed through the electrode pair and the tank reactor. The water level of the simulated water was expressed by the depth from the surface of sand, and it can be regulated by selecting the outlets of the tank. pH and oxidation-reduction potential (ORP) probes (Fondriest Company, USA) were used to monitor the changes of the pH and ORP of the effluents. Water samples within the tank could be collected by a glass syringe through the sampling ports. All sampling ports were sealed with nuts and septa.

Three kinds of influent solutions were prepared for experiments. First, simulated groundwater containing ammonia was prepared by dissolving $(\text{NH}_4)_2\text{SO}_4$, NaCl and Na_2SO_4 in deionized water. The concentration of ammonia nitrogen ($\text{NH}_4^+\text{-N}$) was 20 mg/L; Na_2SO_4 was constantly 10 mM for all trials; and the amount of NaCl was adjusted to achieve different chloride ion concentrations. In addition to the simulated groundwater solution, real landfill leachate after an anaerobic fermentation process was collected from the Chenjiachong landfill site of Wuhan City for two additional experiments. The real landfill leachate was filtered by filter papers, and was used as the stock solution for preparing influent solution. Before dilution, the leachate contained 1790 mg/L $\text{NH}_4^+\text{-N}$, 4000 mg/L COD and 3900 mg/L chloride ions. In the experiments

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