



Short communication

Electrochemical removal and recovery of iron from groundwater using non-corrosive electrodes

Van Khanh Nguyen, Yeonghee Ahn*

Department of Environmental Engineering, Dong-A University, Busan 49315, Republic of Korea

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ABSTRACT

Iron contamination in groundwater has attracted much attention from environmentalists and government agencies because it can cause many problems in human life and in industrial and agricultural activities when groundwater is directly used without any treatment. This study aims to investigate the electrochemical oxidation of Fe(II) to Fe(III) and recovery of insoluble Fe(III) using non-corrosive graphite electrode which serves as a controllable, low-cost, low maintenance and virtually unlimited electron acceptor for Fe(II) oxidation. The lab-scale results indicated that Fe(II) removal up to 100% was obtained at an applied voltage higher than 2 V. The Fe(II) removal efficiency was linearly increased with the increase of potential supply in the range of 1–4 V in the salinity 0.5%. The Fe(II) removal rate could no longer be enhanced at the applied potential higher than 8 V in the condition without salinity. The results from SEM-EDS and XRD revealed that Fe was recovered as FeOOH by conventional filtration with a recovery efficiency of 82.7–92.1%. The electrochemical Fe(II) removal might be an alternative for the conventional method of the in situ Fe removal from groundwater. Besides, the recovered FeOOH can be used as a raw material for environmental remediation and pigment industry.

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1. Introduction

Iron is an essential element for human body but its presence in groundwater at elevated concentrations can cause many inconvenient problems when groundwater is directly used as the water source without any treatment. Iron contamination in groundwater and its environmental problems were investigated widely in many regions (Achary, 2014; Hossain et al., 2015; Ngah and Nwankwoala, 2013; Raju, 2006). Iron occurs in groundwater mainly due to the dissolution of naturally-existing iron-containing minerals in aquifers and bedrocks and partially from industrial discharges (Ityel, 2011; Ngah and Nwankwoala, 2013). Aqueous ferrous iron [Fe(II)] and insoluble ferric iron [Fe(III)] are two common forms of Fe found in water sources (Hossain et al., 2015; Palmucci et al., 2016). As a result of anaerobic dissolution, aqueous Fe(II) is more dominant in groundwater. Also, the microbial Fe(III)-reduction significantly contributed to the release of Fe(II) in the anaerobic aquifer (Ko et al., 2016). The microbial Fe(III) oxyhydroxide reductive dissolution is dependent on both the redox potential of the anaerobic

environment and electron donor sources (organic matter) (Christensen et al., 2000; Di Curzio et al., 2016). Based on Gibb's free energy of contaminant reduction with organic matter mineralization, Fe(II) was only released into groundwater when the denitrification and Mn(IV) reduction were completed (Christensen et al., 2000). When the water containing aqueous invisible Fe(II) was exposed to the air, Fe(II) was oxidized to insoluble reddish Fe(III). This is the cause of all problems related to iron contamination in water.

Iron contamination in groundwater can result in many environmental problems in both industrial and agricultural activities (Ityel, 2011). Water quality obtained from the aquifer storage transfer and recovery (ASTR) was much deteriorated due to the presence of Fe(II) in the recovering well. Unpleasant taste and turbidity can be developed at the Fe(II) concentration higher than 0.3 mg L⁻¹ (WHO, 2003). Laundry and sanitary equipment can also be stained by water with a Fe(II) concentration above 0.3 mg L⁻¹ (WHO, 2003). Color and turbidity can be developed in the pipe system with the water containing Fe(II) higher than 0.05 mg L⁻¹ (WHO, 2003). Water source contaminated with high Fe(II) concentration can block pump system and the precipitation of Fe can reduce the light transmission in crop greenhouse resulting a decrease in crop productivity. Therefore, most of the countries have

* Corresponding author.

E-mail address: yahn@dau.ac.kr (Y. Ahn).

adopted the guideline value from World Health Organization (WHO) on the maximum acceptable concentration of Fe(II) in drinking water at 0.3 mg L^{-1} (Ityel, 2011).

The effective methods for iron removal from water such as membrane, adsorption, and softening with lime often suffer from high cost of materials and operation. Furthermore, due to the nature of these technologies, the in situ application is adventurous because the transportation of absorbent materials in the subsurface environment is non-controllable (Van Halem et al., 2010). The common technologies for Fe removal are the oxidation of soluble Fe(II) followed by the filtration of insoluble Fe(III) (Ityel, 2011). Various oxidizing agents could be used for Fe(II) oxidation such as ClO_2 , O_3 , and KMnO_4 . However, the most cost-effective and eco-friendly method for Fe(II) oxidation has been based on aeration which utilized atmosphere oxygen as the oxidizing agent. The injection of oxygenated water into aquifer could effectively mediate Fe(II) oxidation in groundwater but this technology is limited by the uncontrollable transportation of injected oxygen in the underground environment. The injected oxygen can be consumed for other side reactions and mediate some unwanted underground processes.

This study investigated electrochemical Fe(II) removal and recovery in a reactor with graphite rod as the low-cost and non-corrosive electrode. Electrochemical Fe(II) oxidation was carried out under both conditions with and without salinity at various applied voltages to achieve the desired removal efficiency. The insoluble Fe(III) was recovered through filtration and introduced to characterization. The basic mechanism for electrochemical Fe(II) removal by graphite electrode was also clarified.

2. Materials and methods

2.1. Electrochemical reactor configuration and operation

The experiment was carried out using 4 glass reactors with the working volume of 700 mL for each reactor. No membranes or separators were used between anode and cathode chambers. Both anode and cathode electrodes were graphite rods with 15 cm in length and 0.5 cm in diameter. The contacting part of the electrode with the solution is 8 cm in length and 12.8 cm^2 in surface area. Schematic diagram of the reactor was described in Fig. 1. Two reactors were connected to a programmable direct current (DC) power supply (OPE-303QI, ODA Technologies, Incheon, Korea) whereas two other reactors were not connected to any power supply to maintain as a control.

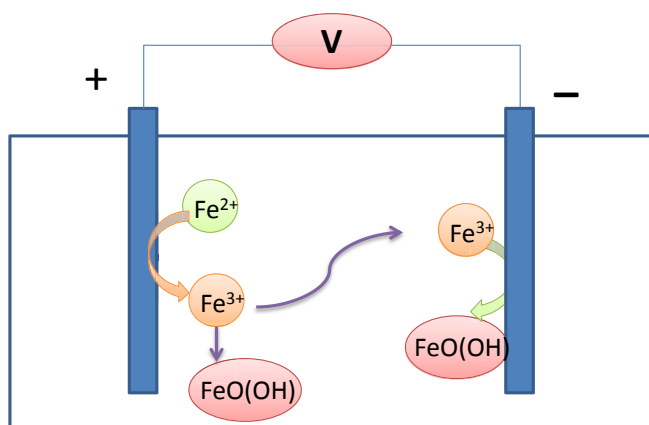


Fig. 1. Schematic diagram of electrochemical system used for iron removal from groundwater.

According to the previous study (Langdon and Nath, 2010), electrochemical oxidation of Fe would be accelerated at high salinity. Therefore, the reactor was firstly operated at salinity of 0.5% (5 g L^{-1} of NaCl) with the voltage applied in the range from 1 to 4 V. The salinity was selected according to the salinity value of the groundwater measured at the testbed site of a river delta which is the target site for the in-situ application (Ko et al., 2016). Then, the experiment was carried out at a salinity of 0% with the voltage applied in the range from 4 to 8 V. The synthetic groundwater was prepared by adding 0.1 g L^{-1} NaNO_3 , 0.01 g L^{-1} KH_2PO_4 , and 0.06 g L^{-1} CH_3COONa into distilled water (Ciardelli et al., 2008). The pH of synthetic groundwater was adjusted at 7.0. Fe(II) was added at an initial concentration of 100 mg L^{-1} by dissolving 0.5 g L^{-1} of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ in the synthetic groundwater. At the end of each experimental run, the iron precipitates from both control and working reactors were recovered by filtering the solution through the $0.45\text{-}\mu\text{m}$ filter membrane. The obtained precipitates were dried in the oven at 105°C for 2 h and then kept in a desiccator for further investigations. Especially, the precipitates attached on the surface of anodes and cathodes were scraped and air-dried in a desiccator for scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) analyses.

2.2. Iron precipitates characterization

All iron precipitate samples collected from both working and control reactors was first coated with Pt and then introduced to an analytical high-resolution SEM (Hitachi SU-70, Hitachi High-Technologies, Japan) for morphology observation. The morphology of iron precipitates was captured at three different magnifications ($\times 10\text{k}$, $\times 50\text{k}$, and $\times 100\text{k}$). Elemental components of precipitates were determined by the EDS analysis using an EDAX instrument (EDAX Inc., Mahwah, NJ, USA) which was attached to the Hitachi SEM SU-70 microscope. Iron precipitates recovered from control and working reactors were introduced to the X-ray powder diffraction (XRD) analysis using a multi-purpose X-ray diffractometer system (Empyrean series 2, PANanalytical B.V., Almelo, the Netherlands) with Cu cathode (CuK_α) and the 2θ in the range from 10 to 80° . The obtained XRD pattern data were compared with the pattern of standard compounds in the database using the Match! program version 3.5 (Crystal Impact GbR, Bonn, Germany).

2.3. Analytical methods and calculations

Ferrous iron concentration was analyzed using Stookey's method (Stookey, 1970). For this, ferrozine solution was separately prepared by adding 0.1% (w/v) of the 3-(2-Pyridyl)-5,6-diphenyl-1,2,4-triazine-4',4''-disulfonic acid sodium salt ($\text{C}_{20}\text{H}_{13}\text{N}_4\text{NaO}_6\text{S}_2$) into a 0.5% (w/v) ammonium acetate ($\text{CH}_3\text{COONH}_4$) buffer solution of pH 7.0. A sample volume of 0.1 mL was thoroughly mixed with 4.9 mL of newly prepared ferrozine solution. The mixture was kept for 5 min at room temperature to allow for a complete complexation of Fe(II) and ferrozine. The mixture was then poured into a cuvette to measure the optical absorption at 562 nm using a multi-purpose UV-Visible spectrophotometer (UV-1280, Shimadzu, Japan). A linear calibration curve ($R^2 > 0.99$) was obtained for the absorbances and Fe(II) concentration in the range from 5 to 100 mg L^{-1} .

Fe recovery efficiency was calculated according to the following equation:

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