



# Arsenite and arsenate removals from groundwater by electrocoagulation using iron ball anodes: Influence of operating parameters



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## ABSTRACT

Removals of arsenite (As(III)) and arsenate (As(V)) from groundwater by a cylindrical packed-bed electrocoagulation (EC) reactor using Fe ball anodes were investigated in this study. Effects of some operating parameters such as initial pH ( $pH_i$  of 6.5–8.5), applied current ( $i$  of 0.075–0.30 A), initial concentration ( $C_o$  of 30–200  $\mu\text{g/L}$ ), diameter of iron ball ( $d_p$  of 5.0–10.0 mm), height of anode balls in the reactor ( $h$  of 2–8 cm) and airflow rate ( $Q_{\text{air}}$  of 0.0–6.0 L/min) on the removal efficiency of arsenic were evaluated. The removal efficiency of arsenic decreased with increase in concentrations of arsenic from 30 to 200  $\mu\text{g/L}$  while its removal efficiency increased with increase in operating time, applied current, height of anode in the reactor, and airflow rate. The optimum operating conditions for effective As(III) and As(V) removals to meet the permissible level of arsenic effluent concentration of < 10  $\mu\text{g/L}$  were determined as 0.3 A, 14 min of EC time for As(III) and 12 min for As(V), a  $pH_i$  of 7.5,  $C_o$  of 200  $\mu\text{g/L}$ ,  $d_p$  of 7.5 mm,  $h$  of 7.5 cm and  $Q_{\text{air}}$  of 6 L/min, respectively. Arsenic removal efficiency, energy and electrode consumptions, operating cost, charge loading and arsenic removed capacity per amount of electrochemically generated Fe at the optimum conditions were also calculated as 96.0%, 1.442 kWh/ $\text{m}^3$ , 0.0752 kg/ $\text{m}^3$ , 0.612  $\$/\text{m}^3$ , 252 C and 2.55  $\mu\text{g}/\text{mg Fe}$  (0.762  $\mu\text{g}/\text{C}$ ) for As(III) removal and 95.8%, 1.386 kWh/ $\text{m}^3$ , 0.0628 kg/ $\text{m}^3$ , 0.546  $\$/\text{m}^3$ , 216 C and 3.05  $\mu\text{g}/\text{mg Fe}$  (0.887  $\mu\text{g}/\text{C}$ ) for As(V) removal, respectively.

## 1. Introduction

Elevated concentrations of arsenic in groundwater and other natural waters with geogenic sources occur in many areas around the world [1–3]. Processes of arsenic mobilization from sediments may vary depending on the hydro-geochemical characteristics of the aquifer, the presence of oxidized and/or reduced mineral phases and the cofactors associated with arsenic-rich solid phases [3]. Long term exposure to high levels of arsenic can cause a wide range of health effects including skin lesions such as hyperkeratosis and pigmentation changes, circulatory disorders, diabetes and cancers of bladder, lung, kidney and skin [4,5]. Thus, the World Health Organization (WHO) and the United States Environmental Protection Agency (US-EPA) have revised the guideline for arsenic maximum contaminant level (MCL) in drinking water from 50 to 10  $\mu\text{g/L}$  [6,7]. The arsenic threat also affects many countries including Argentina, Bangladesh, Cambodia, Chile, China, Hungary, India, Japan, Laos, Mexico, Myanmar, Nepal, Turkey, USA, Vietnam, etc. [2,3,8].

Naturally elevated arsenic concentrations are common in the West-Anatolia of Turkey, particularly in geothermal waters and groundwaters in borate mines areas. Recently, numerous cases of natural arsenic pollution in these waters have been reported namely, Igdeköy (10–10,700  $\mu\text{g/L}$ ) and Dulkadir (300–500  $\mu\text{g/L}$ ) villages of Kütahya-Emet [9–11], Kütahya-Hisarçik (10–3000  $\mu\text{g/L}$ ) [12], Balıkesir-Bigadiç (33–911  $\mu\text{g/L}$ ) [13], Kütahya-Simav Plain (0.5–562  $\mu\text{g/L}$ ) [14], İzmir-Balçova Plain (1–182  $\mu\text{g/L}$ ) [15], and Eskisehir-Kırka (7–150  $\mu\text{g/L}$ ) [16,17].

The USEPA proposed the best available technologies (BAT) for arsenic removal to achieve compliance with MCL from small water facilities such as ion exchange (95%), activated alumina (95%), reverse osmosis (> 95%), modified coagulation/filtration (95%), modified lime softening (90%), and electrodialysis (85%). In addition, iron oxide-coated sand, manganese green sand filtration, iron filings, and granular ferric hydroxide were classified as emerging technologies by USEPA [18,19]. Drawbacks of these arsenic removal technologies forced municipalities and various industries to search for effective alternative

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treatment technologies for the arsenic removal, ideally by electrochemical methods [20]. Electrocoagulation (EC) is one of the most efficient technologies for removal of both As(III) and As(V) from contaminated water [21–23]. Recently, Amrose et al. [24] reported that real groundwater samples with arsenic concentrations of 80–760 µg/L from Bangladesh and Cambodia in 100 and 600 L of EC reactors (named as ECAR) were reduced to < 10 µg/L. The operating cost was varied in the range of 0.22–1.04 \$/m<sup>3</sup>. The EC is a very promising treatment process for the arsenic removal because of some advantages such as no addition of chemicals or regeneration, a shortened reactive retention time, higher removal efficiency, no pH adjustment, no chemical requirement for pre-oxidation of As(III) to As(V), simplicity in operation, compact treatment facility, and relatively cost-effectiveness [20,21,25].

The most important parameters affecting removal of arsenic from waters were current density, reaction time, arsenic species, type and shape of electrodes, geometry of electrodes and airflow rate in the EC process. Plate and rod types of Al or Fe anode electrodes were generally used in the EC reactors and these had some disadvantages namely, its being time consuming (changing and maintenance) and accommodating a limited number of plate and rod types of electrodes with low surface areas. Therefore, an air-injected EC reactor was used to eliminate the above problems. The new EC reactor using Fe ball electrodes was reported in earlier studies [21,26]. The new EC reactor had specifications of compactness, ease of use, accommodating more anode electrodes with higher surface areas, and providing better removal efficiency.

In this study, As(III) and As(V) removals from the groundwater by a cylindrical packed-bed EC reactor using iron ball anodes were performed to determine the optimum operating conditions. For that reason, effects of operating parameters such as initial pH, applied current, initial arsenic concentration, diameter of Fe ball anode, Fe ball anode height in the EC reactor, airflow rate and operating time on the arsenic removal efficiencies were evaluated. Energy, electrode consumptions, and operating cost for the removal were also calculated.

## 2. Material and methods

### 2.1. Characterization of groundwater

Real groundwater was obtained from a well situated in the province of Kocaeli in Turkey and stored in five tones high-density polyethylene container. All chemical species present in groundwater were determined with standard methods [27]. The concentrations of cations, Ca, Mg, Mn, Na, and Si by ICP optical emission spectrometry (Perkin-Elmer ICP-OES Optima 7000 DV) and anions such as nitrate, sulphate, and chloride by ion chromatography (Shimadzu HIC-20A) were measured in the groundwater. The groundwater was characterized as a pH of 7.6, conductivity of 1055 µS/cm, dissolved organic carbon of 5 mg/L, total alkalinity of 260 mg CaCO<sub>3</sub>/L, total hardness of 418 mg CaCO<sub>3</sub>/L, total Mn of 0.006 mg/L, total silica of 10.18 mg/L, total sulphate of 94.2 mg/L and total nitrate of 24.0 mg/L. Fe, Al, P, and F in the real groundwater were not detected. The groundwater containing arsenic concentration of 30–200 µg/L was prepared daily using sodium arsenate (Na<sub>2</sub>HAsO<sub>4</sub> × 7H<sub>2</sub>O) or sodium arsenite (NaAsO<sub>2</sub>) salts.

### 2.2. Experimental setup

A batch mode of the cylindrical packed-bed EC reactor (acrylic electrochemical cell with a total volume of 2041 cm<sup>3</sup>; 100 mm in diameter and 260 mm in height) was used for removal of the groundwater containing arsenic in Fig. 1. An iron (Fe) ball as anode and cylindrically porous titanium (Ti) as cathode (70 mm in diameter, 255 mm in height and 3 mm in thickness) were used in the EC reactor. The groundwater sample (0.95 L) containing As(III) or As(V) was placed in the EC reactor (Fig. 1). The anode and cathode were connected to a digital DC power supply (Agilent 6675A; 120 V and 18 A). An air-fed diffuser was

attached underneath the reactor and the air was fed continuously at different rates in the reactor to maintain uniform shaking. The electrical current was adjusted to a desired value by the DC power supply and the experimental operation was started. The samples were periodically taken from the reactor during the EC process and then analysed.

### 2.3. Analytical methods

The arsenic concentration in the groundwater samples was determined by an atomic absorption spectrometer (Perkin-Elmer SIMAA 6000 AAS) equipped with a manual hydride generator (MSH-10, PerkinElmer) at 188.9 nm wavelength. The detection limit of this study was 0.10 µg/L and analysis of the duplicates was within 2% of errors. pH of the solutions was adjusted by 0.10 N NaOH or 0.10 N H<sub>2</sub>SO<sub>4</sub>. pH and conductivity of solutions before and after the EC process were measured by a pH meter (Mettler Toledo Seven Compact) and a conductivity meter (Mettler Toledo Seven Go), respectively.

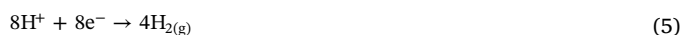
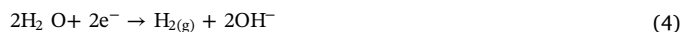
## 3. Mechanism of arsenic removal in the EC process

When a charge is applied through an external power source, the electrolytic dissolution of sacrificial anode produces the cationic monomeric species according to the following Eqs. ((1)–(3)):

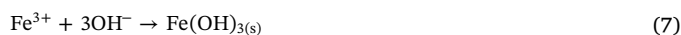
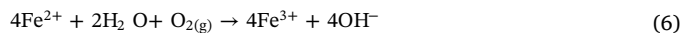


Fe(II) forms highly reactive oxidizing species [Fe(IV)] during oxidation which can oxidize As(III) to As(IV) [28]. This affects oxidation of Fe(II) and effective Fe(IV) utilization such as pH, dissolved oxygen for removal of arsenic.

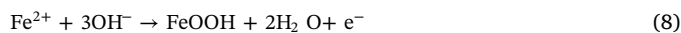
The increase in pH during the EC is primarily attributed to the increase in hydroxyl ion concentration in solution resulting from reduction of water at the cathode (Eqs. (4) and (5)).



The rate of the oxidation depends on the availability of dissolved oxygen (Eq. (6)). Typically at the cathode, the solution becomes alkaline with time. The applied current forced hydroxyl ion migration towards the anode favors ferric hydroxide formation (Eq. (7)). Fe<sup>2+</sup> ions released from anode are gradually hydrolyzed and form Fe(OH)<sub>3(s)</sub>. Thus, the removal of arsenic species from solution is sorption onto the freshly precipitated Fe(OH)<sub>3(s)</sub> particles or flocs



Iron is dissolved giving rise to ferrous ions and its oxidation occurs in the following reaction.



Ferric ions generated by electrochemical oxidation of iron electrode may form monomeric species with respect to pH of the medium, Fe(OH), and polymeric hydroxyl complexes such as Fe(OH)<sup>2+</sup>, Fe(OH)<sub>2</sub><sup>+</sup>, Fe<sub>2</sub>(OH)<sub>4</sub><sup>2+</sup>, Fe(OH)<sub>4</sub><sup>-</sup>, Fe(H<sub>2</sub>O)<sub>2</sub><sup>+</sup>, Fe(H<sub>2</sub>O)<sub>5</sub>(OH)<sup>2+</sup>, Fe(H<sub>2</sub>O)<sub>4</sub>(OH)<sub>2</sub><sup>+</sup>, Fe(H<sub>2</sub>O)<sub>8</sub>(OH)<sub>2</sub><sup>4+</sup>, and Fe<sub>2</sub>(H<sub>2</sub>O)<sub>5</sub>(OH)<sub>4</sub><sup>2+</sup>. These iron hydroxides/polyhydroxides compounds (HFO) have strong affinity for arsenic species. Generally FeOOH produced in the EC has an isoelectric pH of about 7.0. Above the isoelectric point, both arsenic species and FeOOH surface are negatively charged and adsorption is less favorable [21,26]. Hydrous ferric oxides such as amorphous ferric hydroxide, ferrihydrite, lepidocrocite, and goethite strongly sorb both As(V) and As(III) [29–31]. Iron(III) oxyhydroxides can also participate in the

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