

Arsenate Removal from Groundwater by Air-injected EC with Al Ball Anodes: Effects of operational parameters

Hava akışlı elektrokoagülasyon reaktörü ile yeraltı sularından arsenik (V) giderimine işletme parametrelerinin etkisi

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Abstract

Treatment of arsenate from groundwater by electrocoagulation (EC) reactor with air supply unit using Al ball electrodes were studied in this paper. Influence of some operating variables, for instance, applied current (0.075-0.3 A), initial pH (5.5-8.5), air flow rate (0-6 L/min), size of Al ball electrodes (5-10 mm), and height of electrode in EC reactor (2-8 cm) on the As(V) removal efficiency were evaluated. The As(V) removal efficiency increased with the increment of applied current, air flow, electrode altitude in EC reactor, and EC time while its removal efficiency decreased with the increment of size of Al ball electrodes. The maximum As(V) removal percentage, minimum operating cost and energy consumption were found as 98.68 %, 0.609 \$/m³ and 3.7694 kWh/m³ at pH of 7.5, current density of 0.30 A, size of Al balls of 7.5 mm, height of electrode in EC of 5 cm, and air flow rate of 6 L/min, respectively.

Keywords: Air-injected EC reactor, Al ball electrodes, As(V)

Öz

Bu çalışmada hava akışlı elektrokoagülasyon (HA-EK) reaktörü kullanılarak yeraltı sularından arsenik (V) giderimi çalışılmıştır. Arsenik (V) giderimi üzerine, uygulana akım (0,075-0,3 A), başlangıç pH sı (5,5-8,5), hava akış debisi (0-6 L/dakika), Al bilye elektrot boyutu (5-10 mm) ve EK reaktöründe elektrot yüksekliği (2-8 cm) gibi işletme parametrelerinin etkisi incelenmiştir. Arsenik giderimi veriminde artan akım yoğunluğu, hava debisi ve elektrot yüksekliği ile birlikte ciddi bir artış gözlenmiştir. Ancak, arsenik giderim verimi artan Al bilye boyutu ile birlikte azalmıştır. Optimum şartlarda (pH:7,5, akım yoğunluğu: 0,3 A, Al bilye boyutu: 7,5 mm, elektrot yüksekliği: 5cm, hava akış debisi: 6 L/dakika) en yüksek arsenik giderim verimi %98,68, en düşük işletme maliyeti 0.609 \$/m³ ve en düşük enerji tüketimi 3,7694 kWh/m³ olarak hesaplanmıştır.

Anahtar kelimeler: HA- EK reaktörü, Al bilye elektrotlar, As(V)

1 Introduction

Groundwater contaminated with arsenic became a major concern around the worldwide especially in undeveloped or developing countries. Arsenic contamination has been reported from a several countries in the world for instance, Argentina, Cambodia, Chile, , Finland, Hungary, India, Japan, Nepal, Turkey, United States, and Vietnam [1]-[3]. Turkey is also one of the countries under threat of the arsenic pollution. Especially in the western parts of the country, water sources contain higher levels of arsenic than the maximum contaminated level (MCL) set as 10 µg As/L which are as follows: 7.1-834 µg/L for Kütahya, 10-10,700 µg/L in surface and ground waters of Igdekey near the Hisarcık colemanite mining open pits, 30-911 µg/L for Balıkesir in located near boron mines open pits, 10.3-201 µg/L for Aksaray, 1-170 µg/L for Izmir, 50-120 µg/L for Giresun, 0.5-345 µg/L for Sivas, respectively [4]-[10]. Exposure to arsenic lead chronic health problems including pigmentation changes, hyperkeratosis, and cancers of bladder, kidney, liver, skin, lung, and prostate [11]-[12]. Therefore, the WHO and the US-EPA have established 10 µg As/L as a guideline value for arsenic contaminant level in drinking water [13]-[14].

Number of treatment processes have been conducted to remove arsenic in the wastewater/drinking water. These technologies can be listed as follows: adsorption, precipitation, oxidation-filtration, ion exchange, membrane process such as reverse osmosis, biological process [15]-[19]. However, these treatment technologies required several pH adjustments, high maintenance and operating costs, need a long operating time,

and generate a large amount of sludge and secondary pollutants. Therefore, the alternative treatment methods were needed to overcome or/and enhance some of these problems. Electrocoagulation (EC) technique has been receiving greater attention for the removal of arsenic due to its compact treatment facility, affordable maintenance and operating cost, high treatment performance, low sludge formation, and no addition of reagents for oxidation of arsenite (As(III)) to arsenate (As(V)) [20]-[22]. In EC process, rod and plate Al or Fe electrodes commonly used for arsenic removal [23]-[24]. However, these types of electrodes have some disadvantages because of low surface areas and difficulties of operation. Thus, EC reactor using Fe ball type anodes for removal of arsenic was designed and optimized for investigating the operating variables to eliminate the above problems in our previous work [25].

In this study, As (V) removal from groundwater by EC reactor with air injection unit using Al ball anodes was investigated to evaluate the optimum operating variables. Therefore, influence of operating parameters such as applied current, initial pH, EC time, air flow rate, size of Al ball electrodes, and electrode level in the EC reactor. The amount of sludge formation, operating cost, energy and electrode consumptions were also evaluated.

2 Materials and method

2.1 Groundwater Characterization

The groundwater used in EC process (silica 10.18 mg/L, sulphate 94.2 mg/L, nitrate 24.0 mg/L, manganese 0.006 mg/L,

total hardness 418 mg CaCO₃/L, total alkalinity 260 mg CaCO₃/L, dissolved organic carbon 5 mg/L, pH 7, conductivity 1055 μS/cm, and arsenic, aluminum, phosphate, fluoride, iron ions were not detected) was collected from the province of Kocaeli, Turkey. The concentration of ions were measured by IC (Shimadzu HIC-20A) and ICP-OES (Perkin Elmer ICP-OES Optima 7000 DV), respectively. A groundwater containing specified arsenate concentration (200 μg/L) was prepared freshly dissolving sodium arsenate salt (Na₂HAsO₄×7H₂O) to simulate arsenic contaminated groundwater. The arsenate concentration of samples was measured by ICP-OES. pH of the groundwater samples was set by 0.1 M H₂SO₄ and 0.1 M NaOH.

2.2 EC reactor set-up and procedure

Fig. 1. shows a diagram of the air-injected EC reactor. A detailed design of the air-injected EC reactor is described in our previous study [25]. At the beginning of each experimental run, the air-injected EC reactor was fed with 0.8 L of the groundwater. All experiments were conducted in a batch mode and effect of operating conditions on arsenate removal such as operating time, pH, current density, air flow rate, column height in reactor, and Al ball size were investigated. During the experiments, current density applied using DC power supply and voltage values were recorded to calculate the energy consumptions. The weight of Al ball electrodes were also measured before and after the experiments to calculate the electrode consumption. The samples were collected at specified time intervals and filtered using 0.45 μm filter before analyses.

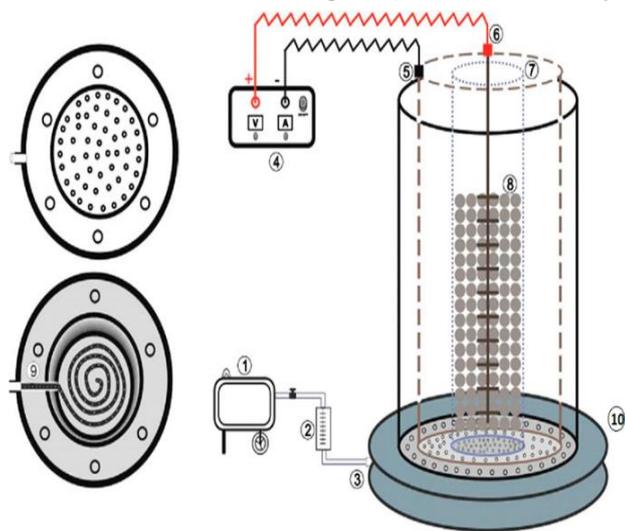


Figure1: The diagram of air-injected EC reactor 1. Air supplier, 2. Flow meter, 3. Pipe line for air supply, 4. Power source, 5. Cathode, 6. Steel rod, 7. Cylindrical shaped Plexiglas, 8. All ball electrodes, 9. Air diffuser [25].

3 Results and discussion

The EC process can be affected by number of operating variables, as follows, applied current, pH, EC time, air flow rate, size of Al ball electrodes, and electrode height in the EC reactor. As shown in Table 1, the low, middle, and high values of operating parameters were specified based on our previous work [25].

Table 1: Operating parameters and levels.

Operating Parameters	Unit	Levels		
		Low	Middle	High
Initial pH	-	5.5	6.5-7.5	8.5
Current density	A	0.075	0.15	0.30
Air flow	L/min	0	2	6
Size of electrode	mm	5	7.5	10
Electrode height	cm	2	5	8

3.1 Effect of applied current

In EC process, EC time and current are the most important operating variables which are controlling the system efficiency. Applied current affects the amount of coagulant and mixing rate of the solution in EC reactor [26]-[28]. Figure 2. demonstrates the effect of current density on residual As (V) concentration during the EC time of 18 min. During the experiments, the initial pH, electrode height in EC reactor, air flow rate, and size of Al ball anodes, selected as 7.5, 5.0 cm, 6 L/min, and 7.5 mm, respectively. In Figure 2, the EC time necessary to meet limit of <10 μg/L of WHO limit arsenic concentration decreased with increasing applied current due to the efficiency of OH⁻ ions production at cathode and dissolution of Al³⁺ ions at anode increased. The observed effluent As (V) concentrations for applied currents of 0.075, 0.15, and 0.3 A were 11.02 μg/L, 7.42 μg/L, and 2.6 μg/L at operating time of 18 min. It can be also concluded that the residual As(V) concentration at 0.075 A was not lower than 10 μg/L for EC time of 18 min, while it took almost 14 min at 0.150 A and 10.5 min at 0.3 A to reached limit arsenic concentration of 10 μg/L. The EC time for As (V) removal was reduced from 18 min to 10.5 min with the increment of applied current from 0.075 A to 0.3 A.

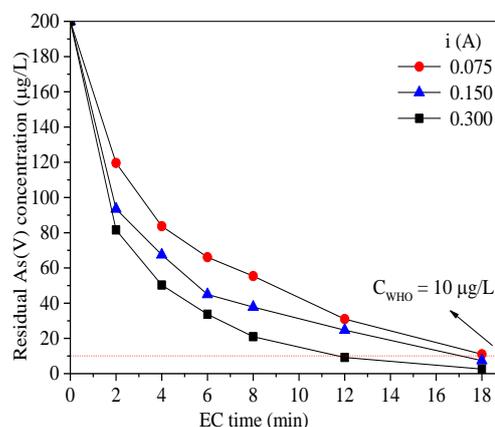


Figure 2: Effect of applied current on residual As (V).

Based on the Faraday's law, consumption of the anode material is directly correlated with the charge loading. The production of coagulant to remove arsenic increases when the charge loading was increased. Therefore, charge loading is another important parameter which controls the reactions in EC reactor. Charge loading was calculated with equation represented as below:

$$q \text{ (F/m}^3\text{)} = \frac{i \times t_{EC}}{F \times v} \quad (1)$$

where q is the charge loading (F/m³ water), t_{EC} is the EC time (min), F is the Faraday's constant (96487 Coulomb), and v is the solution volume (m³) in EC reactor. The high amount of aluminum hydroxide flocs forms with the high charge loadings as it increases Al²⁺ ions in solution. Also, the operating cost of EC process increased with the increment of charge loading. Hence, the charge loading needed to be optimized to minimize the operating cost. The charge loading required to reach <10 µg/L limit value were 1.1993 F/m³ at 0.075 A, 2.3986 F/m³ at 0.15 A, and 4.7971 F/m³ at 0.3 A in EC time of 18 min. The operating cost of the treated water were also determined by considering the amounts of electrode consumption (ELC) and energy consumption (ENC) in the EC process. Based on the Turkish suppliers in March 2018, charges for Al ball electrodes (β) and electrical energy (α) were 15 \$/kg and 0.19 \$/kWh, respectively. The operating cost (OC) of the EC reactor was determined as shown in equation:

$$OC(\$/m^3) = (\alpha \times ENC) + (\beta + ELC) \quad (2)$$

The ENC and ELC was also calculated with the following equations:

$$ENC(kWh/m^3) = \frac{i \times t_{EC} \times U}{v} \quad (3)$$

$$ELC(kg/m^3) = \frac{i \times t_{EC} \times M_{Al}}{z_{Al} \times F \times v} \quad (4)$$

where U is the cell voltage (V), M_{Al} is the molecular weight of the Al ball anode (M_{Al} : 26.98 g/mol), and z_{Al} is the electron number in oxidation/reduction reactions (z_{Al} : 3). The operating costs, electrode and energy consumptions calculated from equations (2), (3), and (4) were 0.350, 0.389, and 0.458 \$/m³-0.0055, 0.0114, and 0.0347 kg/m³- 0.3153, 0.9360, and 2.7060 kWh/m³ at 0.075, 0.15, and 0.3 A, respectively. The sludge production were also determined as 0.096 kg/m³, 0.079 kg/m³, and 0.147 kg/m³ for the currents of 0.075, 0.15, and 0.3 A. In conclusion, the results indicated removal efficiency of arsenate increased with the increment of applied current. Thus, the optimum applied current for effective arsenate removal were selected as 0.3 A for the rest of experiments.

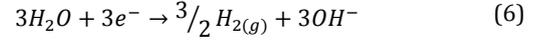
3.2 Effect of solution pH

The solution pH has great impact on removal of arsenate in aqueous solutions and performance of EC process [23]. The negatively charged dominant As(V) species in the aqueous solution are AsO₄³⁻ at pH >12.4, H₂AsO₄⁻ at 3.6 < pH < 7.2, and HAsO₄²⁻ at 7.2 < pH < 12.4. According to the literature, the adsorption of As (V) on amorphous Al(OH)_{3(s)} at As(V) concentration of 1600 mg/L and 7492 µg/g were observed as >100 mg/g, and 118.9 µg/g at pH 6, respectively [29]. In the case of EC reactor using Al electrodes, Al³⁺ ions dissolved from the anode and react with the hydroxyl ions dissolved from the cathode to form aluminum hydroxide (Al(OH)_{3(s)}). The main anode, cathode, and As (V) removal mechanisms were represented with the following equations:

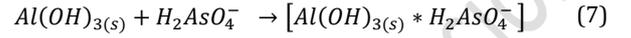
Anode reaction:



Cathode reaction:



Adsorption reactions:



Co-precipitation reaction:

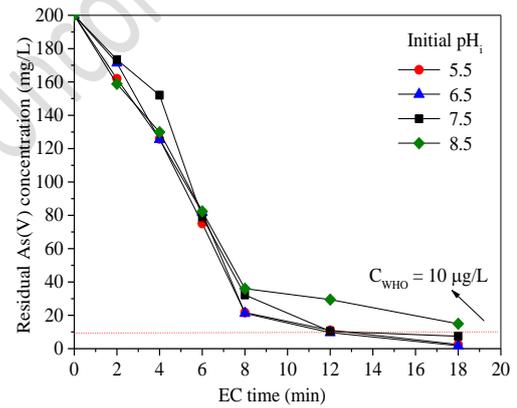
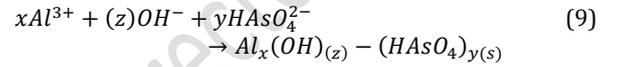


Figure 3: Effect of initial pH on residual As (V).

In this study to examine the pH effect on arsenate removal by the EC, experiments was carried out at pH of 5.5-8.5 since most of groundwater have the pH range of 6-9 [23]. As shown in Figure 3. As (V) removal percentages and residual concentrations (<10 µg/L) were found as 99.20% and 8.02 µg/L at 12 min for pH 5.5, 96.22% and 9.65 µg/L at 12 min for pH 6.5, 95.65% and 9.20 µg/L at 12 min for pH 7.5, and 92.01% and 7.9 µg/L at 18 min for pH 8.5, respectively. According to results, the air-injected EC reactor was remove As (V) efficiently at pH 5.5-8.5. However, the increase of removal efficiency was slight at pH 7.5-8.5, while the removal was significant at pH 5.5-7.5. Since the Al³⁺ and OH⁻ react to form monomeric and polymeric Al(OH)_x species at neutral conditions, which have large hydroxyl groups to adsorb arsenic. At neutral conditions, the arsenic removal was dominated by adsorption and precipitation while at basic pH conditions the removal was only dominated by precipitation. Hence, the arsenate removal efficiencies using Al ball electrodes were better at neutral pH (7.5).

Values of electrode and energy consumptions, operating cost, and amount of sludge formation were also calculated as 0.0318

kg/m³ and 2.8941 kWh/m³- 0.718 \$/m³-0.2825 kg/m³ for pH 5.5, 0.0221 kg/m³ and 3.2027 kWh/m³-0.726 \$/m³ - 0.2330 kg/m³ for pH 6.5, 0.0334 kg/m³ and 3.3069 kWh/m³ - 0.805 \$/m³ - 0.2309 kg/m³ for pH 7.5, and 0.0225 kg/m³ and 2.7694 kWh/m³ - 0.645 \$/m³ - 0.1700 kg/m³ for pH 8.5, respectively. The pH values after the EC process changed from 5.5, 6.5, 7.5 and 8.5 to 6.01, 7.0, 7.84 and 8.96, respectively. This slight increase of pH values related to the formation of H_{2(g)} and OH⁻ ions on cathode which compensated the acid buffer thus form an alkaline solution [30].

3.3 Effect of electrode level in EC reactor and size of Al ball electrodes

The electrode height in EC reactor is another important parameter which effects the arsenate removal efficiency and electrode life time. The effect of electrode height in EC reactor was investigated at air flow rate of 6 L/min, size of Al ball electrodes of 7.5 mm, pH of 7.5 and applied current of 0.3 A. Figure 4. shows the effect of electrode height on residual As (V) concentration. The residual As (V) concentration decreased with the increasing electrode height in EC reactor since increase in total Al anode surface area. The increasing arsenate removal efficiency with the increment of electrode height could be explained by Faraday's Law.

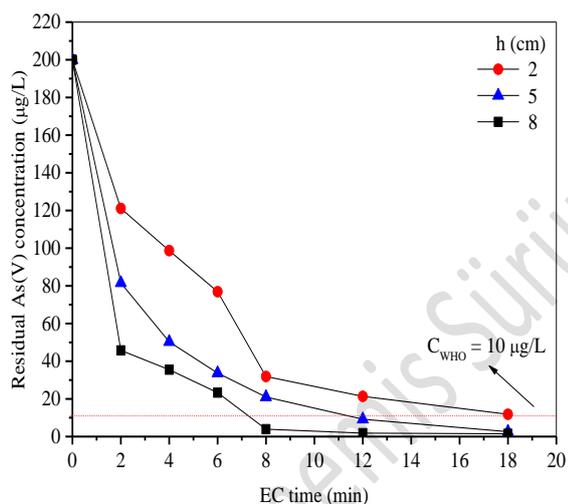


Figure 4: Effect electrode height on residual As (V).

Total Al anode surface area were 0.032 m², 0.076 m², and 0.117 m² for electrode height of 2 cm, 5 cm, and 8 cm, respectively. As seen in Figure 4, the residual As (V) concentration and minimum operating time were 1.14 µg/L and 8min for 8 cm, 9.07 µg/L and 12 min for 5 cm, and 10.12 µg/L and 18 min for electrode height of 2 cm, respectively.

Another important operating condition is the size of Al ball electrodes which influences the residual As (V) concentration. The effect of size of Al balls on As (V) removal shown in Figure 5. The size of Al ball electrodes in EC reactor decreases at a specified electrode height in EC reactor (8 cm), the surface area of Al ball electrodes used in the EC increases. The residual As (V) concentrations for 5.0 mm, 7.5 mm, and 10 mm Al electrode sizes were calculated as 3.26 µg/L, 9.52 µg/L, and 11.97 µg/L at EC time of 12 min, respectively. When the surface area of Al ball electrodes increased, the As (V) removal efficiency increased due to the formation of significantly high amount of Al³⁺ ions in solution. The optimum size of Al ball electrodes were selected

as 7.5 mm since there was no difference between 5 mm and 7.5 mm of the size of Al ball electrode size in terms of arsenic removal efficiency. However, the Al ball size of 7.5 mm was chosen as it has higher surface area.

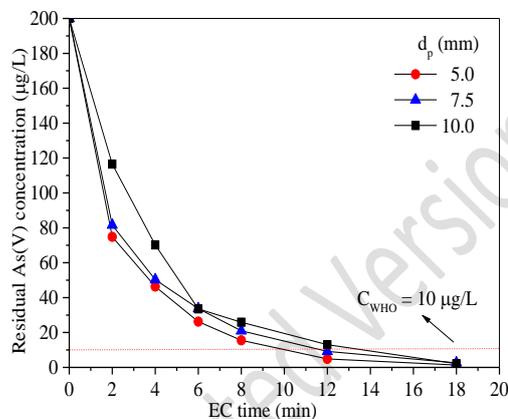


Figure 5: Effect of size of Al ball electrodes on residual As (V).

3.4 Effect of air/oxygen flow in EC reactor

The air flow is a notable operating parameter in EC system to provide homogenous mixture in solution. In addition, the air flow also supports the coagulation/flocculation mechanisms as it supply the mixing in the solution in EC reactor. The formation of passivation layer on anode surfaces also prevents with the air flow rate [31].

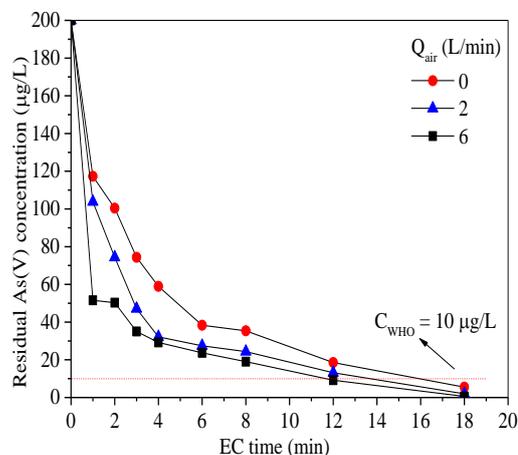


Figure 6: Effect of air/oxygen flow on residual As (V).

The effect of air flow on treatment efficiency were investigated at 0, 2, and 6 L/min of air flows. The residual As (V) amount diminished with the increasing air flow as seen in Figure 6. The residual As (V) concentration at EC time of 12 min were found as 7.06 µg/L for 6 L/min while the residual arsenic concentration was not achieved <10 µg/L limit value for 0 and 2 L/min air flows. Also, the residual As (V) concentrations reached <10 µg/L limit arsenic value at the end of the EC time of 14 min. At EC time of 18 min, the residual As (V) concentrations were 5.51 µg/L, 2.64 µg/L, and 2.58 µg/L for air flow rate of 0, 2, and 6 L/min, respectively. The operating cost,

energy consumption, and charge loading at the end of the EC time of 18 min were also calculated as 0.913 \$/m³, 3.8867 kWh/m³, and 0.0354 F/m³ for 0 L/min, and 0.762 \$/m³, 3.4689 kWh/m³, and 0.0329 F/m³ for 2 L/min, and 0.609 \$/m³, 3.7694 kWh/m³, and 0.0195 F/m³ for 6 L/min.

Consequently, it can be concluded that the novel air-injected EC reactor significantly increased the removal efficiency of As (V). Since, the homogenous mixing in solution increased with the increasing air flow rate. Also, the passivation layer on the surfaces of Al ball anodes prevented with the increment of air flow rate.

3.5 Comparison of arsenic removal by EC process

The removal of arsenic by EC process was studied by many authors. This study compared with the other studies in the literature based on arsenic concentrations, electrode types (Al, Fe, Cu and hybrid electrode combinations), and electrode shapes (rod, plate and ball).

The arsenic removal efficiency of EC process for groundwater containing 285 µg/L arsenate achieved a maximum removal efficiency of 96.9% and 99.3% using Fe plate and Fe ball electrodes, respectively [26]. In a separate study, the removal efficiency and operating cost in a batch mode EC were 99.2% and 0.031 \$/m³ using Fe ball electrodes at 150 µg As (V)/L [25]. The authors also observed that the arsenic removal efficiency and energy consumption were found as 92.6% and 1.19 kWh/m³ using Al plate type electrodes [32]. Arsenic removal by EC process carried out using Al plate at operating time of 62.8 min and 85% of arsenic removal efficiency were observed [33]. In our previous work, the comparison of arsenic removal by EC process were reported in more detail [34]. The removal efficiency in the EC process using Al ball electrodes was calculated as 98.68% which was higher compared to the literature values. Also, conventional treatment processes used for the removal of arsenic were presented in the introduction. The EC process compared with the other studies, EC turns out to be an effective and environmentally friendly method for the removal of arsenic from groundwater.

4 Conclusion

In this paper, EC reactor was investigated for removal of As (V) and observed to influence of operational variables. The results showed that the increasing applied current, electrode level in EC reactor, EC time, and air flow rate have positive effect on As(V) treatment efficiency while the increasing size of Al ball electrodes have negative effect on removal efficiency. On the other hand, the effect of initial pH on As (V) removal was not notable in specified pH values. According to results, the minimum residual As(V) and minimum operating cost were found as 2.64 µg/L and 0.609 \$/m³ at initial pH of 7.5, applied current of 0.3 A, size of Al balls of 7.5mm, height of electrode in EC of 5 cm, and air flow rate of 6 L/min. It was concluded from this work that As (V) could be removed effectively by EC reactor using Al ball electrodes.

5 Acknowledgement

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