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ELECTROCOAGULATION PROCESS FOR SHORT TREATMENT PERIOD OF ARSENIC CONTAMINATED WATER

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Abstract

The effective arsenic removal rate was achieved by the electrocoagulation (EC) method. The aim of this paper is to study the arsenic removal by EC method applied for short period and to investigate the effects of current density, pH, salt addition, and conductivity on the removal rate and energy consumption. The EC experiments started in batch mode in 10 L plexiglass reactor with five aluminum electrodes. The electrocoagulation was started with the initial arsenic (As) concentration of 1.00 mg/L. Current intensity and voltage values ranged between 1-2 A and 2-15 V, respectively for the process time of 10 min. The highest As removal rate (99%) was found for 5 V and 1 A current application. 1.29 mA/cm^2 current density provided the optimum energy consumption (0.60 Wh) for 99% As removal. Increased removal was noticed above pH 8. Current density, pH, and conductivity were found effective factors on the As removal.

Key words: arsenic, contamination, electrocoagulation, current density

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1. Introduction

Arsenic (As) contaminates groundwater all over the world through the hydrogeological processes (Das and Nandi, 2021). It is found in natural environments due to geological formations such as lake sediments and volcanic rocks. In many parts of the world, groundwater is the most important source to supply drinking water; one-third of the world's population depends on groundwater resource. Smaller societies, such as in rural areas, notify a greater dependence on groundwater than surface water (Mohammadi et al., 2018). By its nature, groundwater is responsive to contamination by sources in surface water and from surrounding geological processes (Oh et al., 2019). Arsenic concentration in drinking water above a certain limit is carcinogenic to humans (Das and Nandi, 2021). The World Health Organization (WHO) determined the permissible limit of arsenic in drinking water as 10 µg/L and declared toxic water containing arsenic above this amount (WHO, 1996). In Turkey, arsenic amount in drinking water is determined as <10 µg/L in Turkish Drinking Water Standards (TSE 266) by the Ministry of Health (Anonymous, 2005). So, the control of arsenic in water is the urgent issue for local governments. By now a number of researchers studied on As removal by electrocoagulation (Dhadge et al., 2018; Kobya et al., 2016; Kobya et al., 2018; Mroczek et al., 2019; Silva et al., 2018). Electrocoagulation has been successfully used to treat the arsenic contaminated waters.

Electrocoagulation (EC) process has some advantages over the other water treatment methods such as high removal efficiency, lower operating costs, no chemical addition, reduced sludge

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generation, and energy efficiency (Ozyonar and Aksoy, 2016). The most effective factors that determine and govern the reactions in the system and the generation of the coagulant are applied charge density, type and condition of electrodes, and the nature and concentration of the pollutants (Mena et al., 2019).

Electrocoagulation technology requires applying an electric current to an aqueous solution through metal electrodes such as iron (Fe) and/or aluminum (Al) (Emamjomeh et al., 2020). The most used electrode materials are Al and Fe and they lead to high arsenic removal efficiencies of up to 99% (Mohora et al., 2019). For wastewater treatment, EC initiates the metal oxidation and dissolution, thus leading to the production of ions and gases (oxygen and hydrogen), which generate the coagulation/flocculation at suitable pH (Jung et al., 2015; Mores et al., 2018). In EC process, when a direct current is applied, aluminium or iron plates dissolve by generating metallic cations such as $Al^{3+,}$ Fe^{2+} or Fe^{3+}. Cations form a range of coagulant species or metal hydroxide which adsorb or coprecipitate the dissolved pollutants (Emamjomeh et al 2019; Song et al., 2015). The basic steps include in EC process are (i) electrolytic dissolution of anode plate due to oxidation (Eq. 1), (ii) formation of hydroxide ions and hydrogen gas at the cathode (Eq. 2), (iii) formation of metal hydroxide in solution (Eq. 3), (iv) adsorption of contaminants at metal hydroxide surface and charge neutralization (Islam et al., 2018) and (v) contaminants removal by settling.

Anodic reaction:

$$Al \rightarrow Al^{3+} + 3e^- \qquad (1)$$

Cathodic reaction:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \qquad (2)$$

Reaction in aqueous solution:

$$Al^{3+} + 3OH^- \rightarrow Al(OH)_3\ (s) \qquad (3)$$

In electrocoagulation, aluminium and hydroxyl ion generated by reactions (1) and (2), undergo hydrolysis and form various monomeric and polymeric species, that are further transformed into amorphous aluminium hydroxide. This precipitation-prone aluminium hydroxide complex is imagined adsorbing arsenic (Thakur et al., 2019). Typically, aluminium, iron, graphite, titanium, carbon, mild and steel plates are used as electrodes in the electrocoagulation process. Iron and aluminium have been reported to be very effective and successful in contaminant removal at appropriate operating conditions (Can et al., 2016). Although the both are the most used electrodes in EC processes, other materials have produced good results in different kinds of water treatment. For instance, the use cooper to remove phosphorus from water (Hong et al., 2013), zinc to reduce the organic load from wastewater (Fajardo et al., 2015), combinations of titanium and platinum electrodes for treating dye solutions (Fajardo et al., 2017). The aim of this study is to investigate the removal efficiency of arsenic by electrocoagulation method for short period (10 min) and to evaluate the effects of current change for different amperage and voltage conditions, impacts of different pH, salt addition and conductivity.

2. Material and methods

2.1. Experimental setup, electrocoagulation process and analytical methods

The EC experiments were performed in batch mode in a plexiglass reactor with a useful volume of 10 L (Fig. 1). Characteristics of the electrocoagulation and process conditions are presented in Table 1. Five aluminum electrodes with 175 mm height and 105 mm of width that connected monopolar parallel were placed at 5 cm intervals. The mixing process in the plexiglass reactor was provided by two IKA RH Basic 2 model magnetic stirrers. Electrodes were connected with GW INSTEK GPS-3303 DC regulated power source operating at a maximum of 30 V and 3 A. During the electrocoagulation experiment, 10 g of NaCl was added to the electrocoagulation as supporting electrolyte to ensure electrical conductivity.

Aluminum and arsenic analyze were performed in the laboratory at Optima 2100 DV brand ICP (Inductively Coupled Plasma-Optical Emission Spectrometer) (ICP hydride system was used for arsenic). Electrical conductivity and pH were measured with HQ 40d EC meter and WTW 340I pH meter, respectively. Samples were passed through single-use 0.45 μm syringe filters before the measurements. All measurements were carried out in accordance with Standard Methods 1999 (Apha-Awwa-Wef, 1999), as suggested by the American Public Health Association. Samples were stored in polypropylene containers and all chemicals used in the experiments were of analytical purity.

Synthetic arsenic water was prepared by stock solution of 1000 mg/L As standard. The electrocoagulation was started with the initial As concentration of 1.00 mg/L. Digital magnetic stirrer was used for providing necessary agitation (250 rpm) in water. During EC, treated water samples were taken under the suspended flocks with 15 cc syringes and were filtered into the 15 mL sample tubes with a 0.2 mL (0.1 M) of nitric acid and then analyzed. Washing acid and plenty of distilled water were used for cleaning the electrocoagulation after each operation. Aluminum electrodes were also washed with pure water and dried immediately after the experiments. Various operating parameters used during EC experiments are outlined in Table 1.

During the EC process, aluminum ions dissolved from Al electrode surface formed a flock with the destabilized components in water and aluminum flocs were collected on the surface of water in electrocoagulation with the rising upwards hydrogen gas that formed in the system. The flock layer has been observed as in the form of foam. While a foam layer is formed on the upper surface of the reactor, the presence of purified water in the middle of the reactor and the precipitated layer formed by the effect of gravity on the bottom of the reactor were noticed as the reactor was stopped for a short time.

Fig. 1. The EC setup with electrocoagulation and DC power source

The scope of this study includes construction and operation stages of 3-pass fire tube boilers. The diameter and the length of both types of boilers are 2.5 and 5.0 meter, respectively. The boilers produce the steam of 3 ton/hr, having the temperature of 165 ^{o}C, using rubber wood chips (10.5% moisture content) of 667 kg/hr. The pressure of the steam is 7 bars. After the steam is used, the condensate goes back to the boiler along with the feeding water. Simple water spraying is used to capture ashes from flue gas. After the end of life, the boilers are brought to a recycling plant and thus the data at the disposal stage are excluded.

The natural resources used for the production of raw materials, energy and water as well as emissions and waste are also included. Description of the system and data under study is shown in Fig. 1.

New boiler: The new boiler is made of virgin materials. After all parts are assembled, a hydro pressure test is performed. The steam production capacity is 3 ton/hr, having an efficiency of 70% (CIBO). The operating lifetime is 42,000 hr (Ecoinvent, 2007).

Recycled boiler: A coal boiler having a steam production capacity of 5-6 ton/hr is converted to the recycled boiler. The construction processes consist of the transfer of the old boiler to the construction factory, cleaning of the boiler as well as fire and water tubes, changing and repairing the insulator, gasket and the out of specification parts. The next step is constructing an additional furnace having the width, length and height of 1.5, 2.5 and 3.0 meter, respectively. The additional furnace is modified in order to fit biomass burning. Then the additional furnace is assembled to the boiler and a hydro pressure test is performed. Fig. 2 shows the picture of the recycled boiler and additional furnace. The steam production capacity is 3 ton/hr, having an efficiency of 60% according to boiler specification. The operating lifetime is 27,270 hour. The recycled boiler can have efficiency as high as the new boiler (70%) if the heat loss is reduced or it is produced from a relatively large boiler.

2.2. Calculation methods

The As removal efficiency (RAs, %) was calculated as Eq. (4) (Mousazadeh et al., 2021):

$$\mathrm{RAs}\ (\%) = \frac{(\mathrm{C0}-\mathrm{Ct})}{\mathrm{C0}}\ x\ 100 \qquad (4)$$

where: R_{As} is arsenic removal efficiency and C_0 and C_t are the initial arsenic concentration and concentration of arsenic at time t in solution (mg/L), respectively.

Energy consumption was calculated in watt-hour (Wh) as Eq. (5) (Mousazadeh et al., 2021):

$$Ec = V \times I \times t \qquad (5)$$

where: Ec is energy consumption (Wh), V is voltage (volt), I is applied current (A) and t is time (h).

Table 1. Main properties of experimental setup in EC study

Parameters	***Value***
EC reactor volume, V (dm^3)	10
Dimensions of reactor. (cm)	30x15x25
Dimensions of electrode (cm)	10.5x17.5x0.2
Effective electrode surface area, S (cm^2)	735
Number of electrodes	5
Number of anode	3
Number of cathode	2
Distance between the electrodes (cm)	5
Electrode material	Al
Applied current (A)	0-3
Applied cell voltage (V)	0-30

3. Results and discussions

3.1. Effects of applied current and cell voltage on As removal

Time dependent As removal percentages at different voltage values for 1 A and 2 A applied current within 10 minutes are given in Fig. 2 and Fig. 3, respectively. Especially for 5 V, 10 V and 15 V voltage applications, As removal rates increased significantly with increasing time. However, no significant change in As removal over time has been observed in the low voltage values (2 V, 2.5 V and 3 V). As seen from the Figures, As removal increased significantly with the increasing voltage values. However, in the process period of 10 minutes, very low removal rates were recorded for 2 V, 2.5 V and 3 V (max 24%). Likewise, increasing the applied current from 1 A to 2 A for the same voltage values did not have a significant effect on the removal rates. But higher as removal rate was observed with 5 V of applied voltage compared to lower values (Karabulut et al., 2021). Energy consumption increased above this value (Uzun and Debik, 2019). At the end of 10 minutes process for 5 V, 99% As removal efficiency was achieved for both 1 A and 2 A current values, but a faster removal occurred at 1 A. The processing time was completed in short period, considering the high energy cost of the electrocoagulation process. It was preferred that the reactor process time should not exceed 10 minutes since it was set out to reach the current and voltage values that would provide maximum removal within the minimum process time.

The effect of applied voltage on As removal rates for 1 A and 2 A current values is shown in Fig. 4. Up to 5 V, 2 A current caused a slightly higher As removal, however, to increase the current from 1 A to 2 A for higher voltages had no effect on the treatment. While the As removal for 5 V reached approximately the same maximum percentages, the energy consumption was 0.618 Wh and 1.275 Wh for 1 A and 2 A, respectively (Fig. 5). For 2 A current, energy consumption increased significantly for 5 V and higher voltage values. In this case, the application of 5 V and 1 A current provides the optimum energy consumption for the highest removal rate.

Energy consumption and As removal rate increased with the increasing current density (Fig. 6). This phenomenon can be related by the fact that the rate of anodic dissolution increases with the increasing current density and as a consequences larger size of floc is generated.

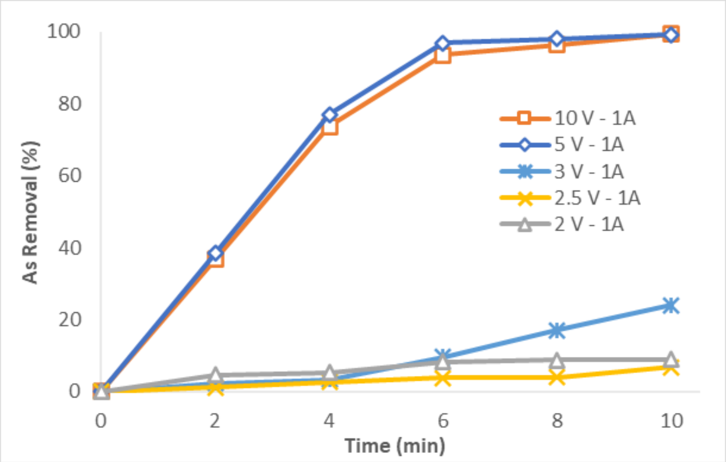

Fig. 2. Time dependent As removal percentages for 1 A (Initial pH:7.03-9.81, T: 19.80 °C-20.60 °C)

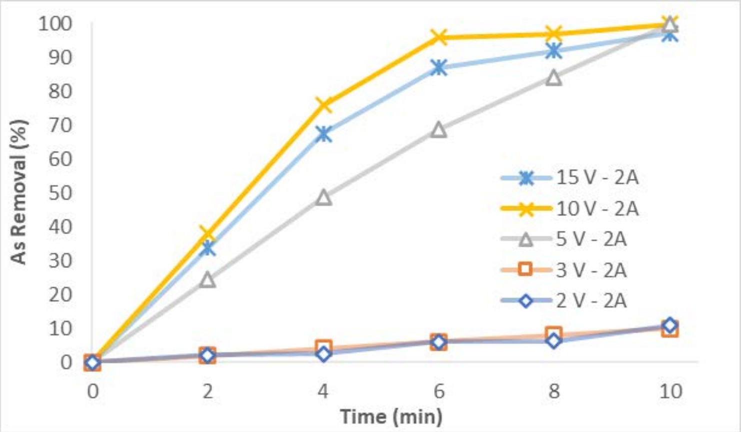

Fig. 3. Time dependent As removal percentages for 2 A (Initial pH:6.34-10.01, T: 18.60 °C-20.60 °C)

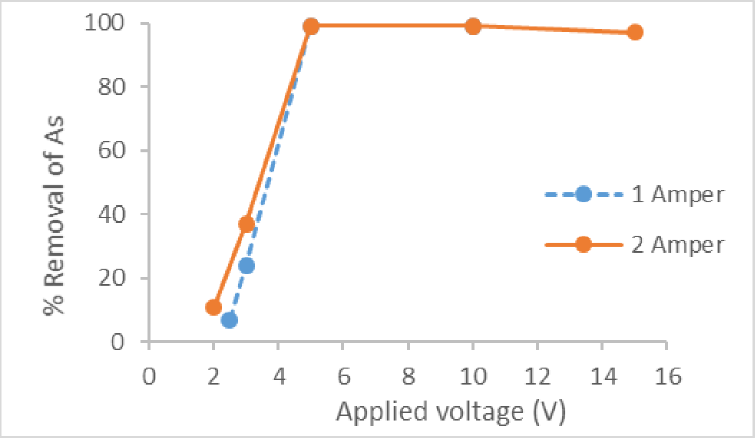

Fig. 4. The effect of applied voltage on As removal rates

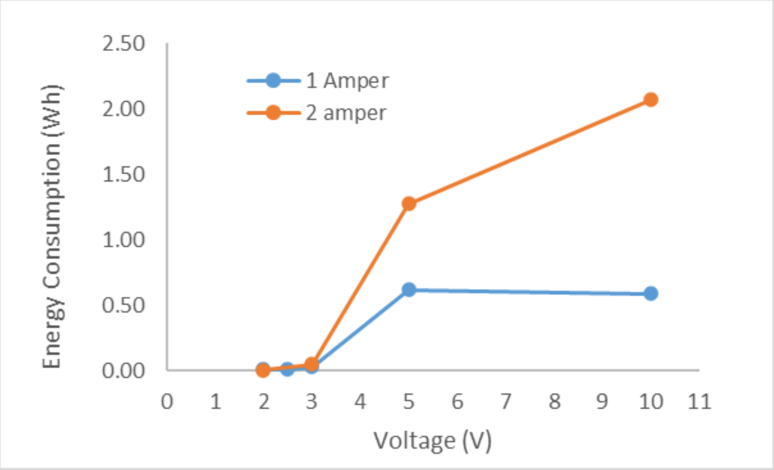

Fig. 5. Energy consumption for increasing voltage values

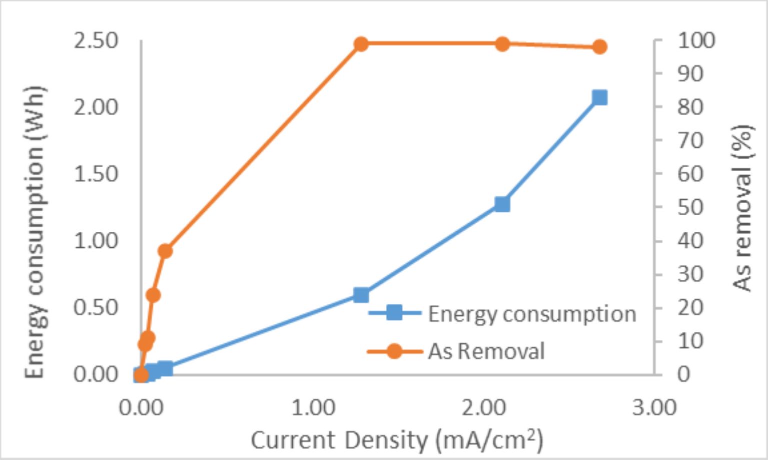

Fig. 6. Energy consumption and As removal rate for increasing current density

These larger flocs with larger surface area increase the adsorption/coagulation rate and thus ensure faster removal of As ions (Das and Nandi, 2021). Current density of 1.29 mA/cm2 provided the optimum energy consumption (0.60 Wh) for 99% removal rate. Lopez-Guzman et al. (2019) obtained approximately 100% arsenic removal for current density of 4.5 mA/cm^2, initial pH of 5, and 15 min of treatment time.

3.2. Effect of pH on As removal

The proportions and distributions of the species at certain concentrations can be found by calculating the electrochemical stresses or with electrochemical equations at the specific oxidation level and temperature. Pourbaix diagram for arsenic types is given in Fig. 7. In this graph, the pH value of the solution is given on the horizontal axis and the Redox potential (Eh) is given in Volt unit on the vertical axis. The +3 valence arsenite species (negative voltage values) are dominant in reducing conditions. Arsenate species with +5 valence are dominant in positive voltage values, ie in oxidizing conditions.

Theoretically, As (V) anions are captured by the $Al(OH)_3$ which are removed by sedimentation or by H_2 flotation. Arsenic was removed from the solution due to the negatively charged arsenate species that electrostatically pulling the positively charged metal hydroxides (Can et al., 2016). As (V) anions are adsorbed onto the aluminum hydroxide precipitates that are finally filtered out of solution. Aluminum and arsenic change their forms depending on pH in aqueous solution. Along the pH intervals (pH 4.7 and 10.5), existence of polymeric aluminum hydroxides would allow significantly larger surface areas for arsenic adsorption due to their amorphous nature (Gomes et al., 2007). Our experiments were carried with 1 mg/L initial As concentration for 3V and 2 A current in 10 min between pHs ranging from 2.40 to 11. The capacity of metal flocs to adsorb arsenic increases at low pH but the acidity is neutralized by electrolysis (Mroczek et al., 2019). As shown in Fig.8, significant As removal did not observed between pH 2 and pH 8 probably because of the neutralization by electrolysis. Increasing As removal was noticed above pH 8 due to the increasing Al flocs adsorbing As. The highest As removal and energy consumption for pH 11 were achieved as 64% and 0.11 Wh, respectively. Considering the previous high As removal percentages (99%) obtained for higher cell voltage and current (5 V-2 A); the high voltage/current application was found to be more effective on As removal than the high pH of electrocoagulation process. Similarly, Can et al., 2016 obtained the lowest arsenic removal efficiency at initial pH of 2. They carried out the experiments with different pHs ranging from 2 to 8 and found that initial pH was highly effective on the As removal efficiency.

However, Lopez-Guzman et al., (2019) defended that the initial pH adjustment did not have a significant effect on the removal of arsenic. In this case, it is thought that other factors such as electrodes, current density, temperature or other anions and cations may affect the process conditions in obtaining different removal results at similar pH ranges.

3.3. Effects of salt addition and conductivity

NaCl was used to increase the conductivity. While the initial conductivity of tap water was 350-410 μS/cm, it was increased to 2430 μS/cm with the addition of 1 g/L NaCl (for 5 V and 1 A). The electrical conductivity and salinity of solutions increases significantly with the added NaCl in water and slight increase occurs in TDS (Total dissolved soils) and turbidity. Time varying initial and final conductivity was measured on the synthetic arsenic water prepared in laboratory. The graph of conductivity and As concentration change depending on time is given in Fig. 9. As can be seen in the figure, the initial conductivity decreased from 2430 to 2410 μS/cm in 30 min time period. This shows that the dissolved matters initiated the increase in conductivity were removed from water by forming compounds either by settling on the bottom or by accumulating on the surface with the flocks. This case allowed the decrease of arsenic concentration with the precipitating or floating flocs.

Das and Nandi (2021) achieved a similar decrease in As amount arising from better transport of As ions to the flocs. From the experimental results they observed that, addition of NaCl with higher concentration increased the removal efficiency of the process. However, lower concentration of NaCl may be preferred to prevent undesired salty taste of water after the treatment.

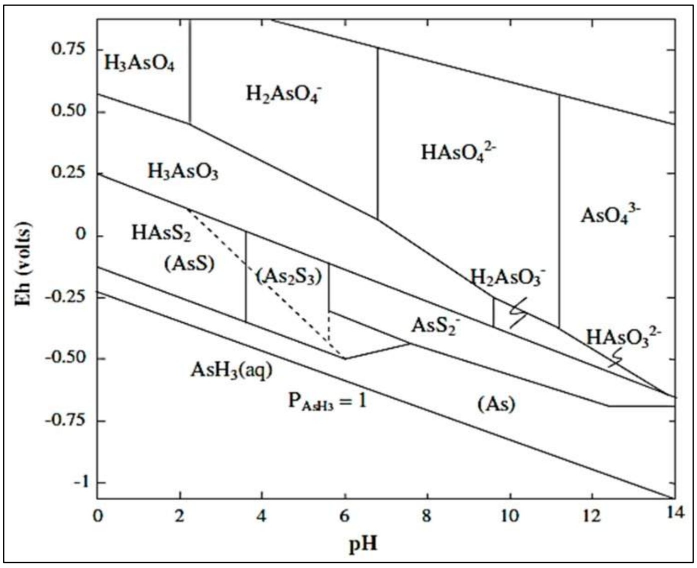

Fig. 7. Pourbaix diagram for Arsenic (Can et al., 2016)

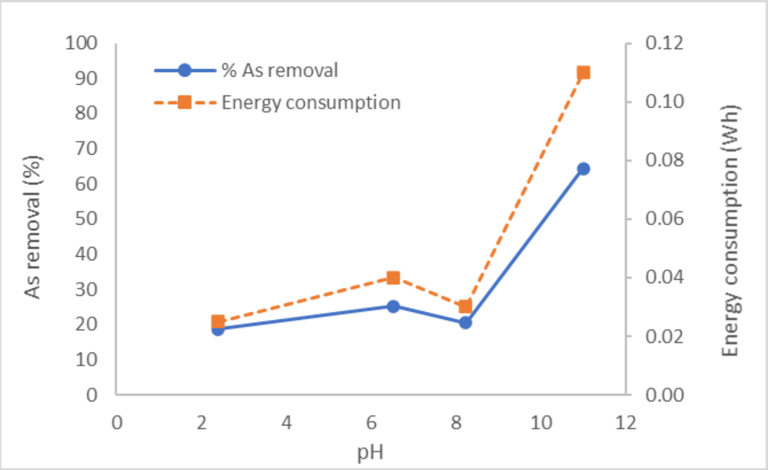

Fig. 8. Effect of pH on As removal and Energy consumption** (1 mg/L initial As concentration and 3V and 2 A for 10 min)

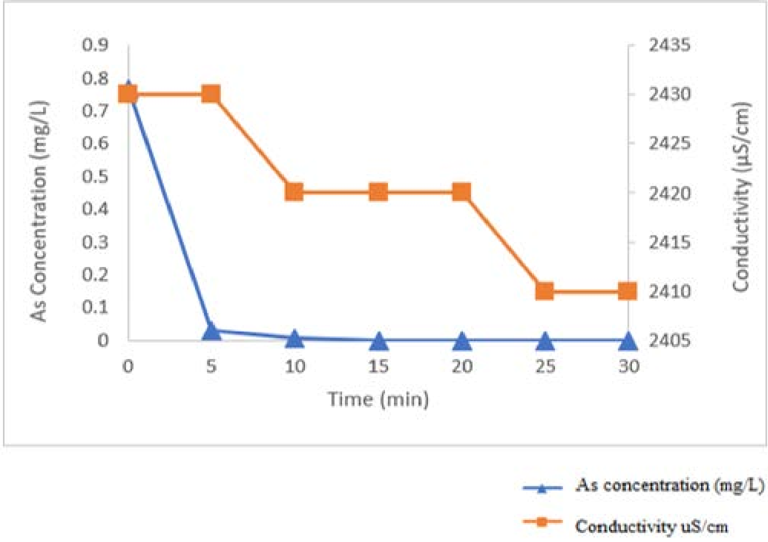

Fig. 9. Change of conductivity and As concentration depending on time (5 V-1 A Current Conditions)

4. Conclusions

The electrocoagulation with Al electrode for process period of 10 min was effective on arsenic removal. Removal rate and energy consumption increased with the increasing current density. 5 V and 1 A was found the optimum voltage-current for the highest removal rate. Current density of 1.29 mA/cm^2 provided the optimum energy consumption (0.60 Wh) for 99% As removal. Low removal rates were observed between pH 2 and pH 8. Increased removal was noticed above pH 8 and the highest removal rate and energy consumption for pH 11 were achieved as 64% and 0.11 Wh, respectively.

The high voltage/current application was found to be more effective on As removal than the high pH of electrocoagulation process. Increasing the initial conductivity to 2430 µS/cm with the addition of 1 g/L NaCl allowed the decrease of arsenic concentration with the precipitating or floating flocs. As a conclusion, current density, pH and conductivity are important on the As removal by electrocoagulation. However, other factors such as electrodes, current density, temperature, process time or other anions and cations may affect the process conditions and therefore the removal efficiency.

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