Simultaneous removal of fluoride and arsenic from well water by electrocoagulation

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HIGHLIGHTS

- Electrocoagulation is a process able to treat and remove fluoride and arsenic simultaneously from drinking water.
- Critical factors in the electrocoagulation process for simultaneous removal are pH, current density and treatment time.
- Acidic pH is more favorable for fluoride removal
- Arsenic removal is less complex than fluoride removal, high removal efficiencies were reached without initial pH adjustment.

GRAPHICAL ABSTRACT
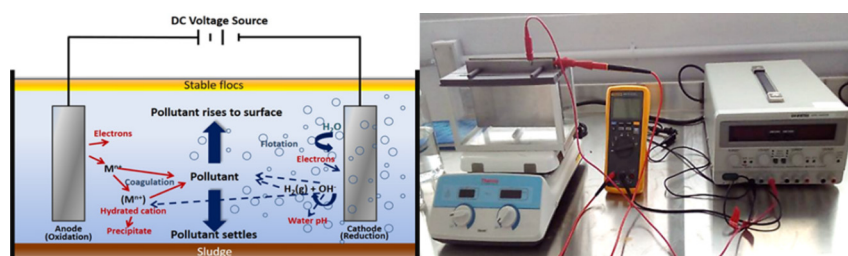
ARTICLE INFO

ABSTRACT

The co-occurrence of fluoride and arsenic in groundwater presents a problem in many, mostly arid, regions of Latin America and the world. These pollutants cause significant health problems and are difficult to remove simultaneously from drinking water. In this study, the electrocoagulation process for the simultaneous removal of fluoride and arsenic was evaluated in well from the state of Durango, Mexico, in order to both solve the local problem and determine how to apply the method generally. Tests were carried out with different times, concentrations, initial pH values, and electric current densities, with iron and aluminum as electrode materials. The removal efficiencies in simultaneous presence were 85.68% for fluoride and approximately 100% for arsenic. The final concentrations for both pollutants were below the drinking water limits established by the World Health Organization (WHO) and Mexican regulations. The optimum conditions of the electrocoagulation process found were a current density of 4.5 mA/cm^2, an initial pH of 5, and a treatment time of 15 min, considering initial fluoride and arsenic concentrations of 5 mg/L and 80 µg/L, respectively.

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1. Introduction

The increasing demand for drinking water has resulted in increased use of groundwater, which has become the primary source of supply for many regions of the world, including in Latin America. It is estimated that >50% of the global population satisfies their water needs from groundwater (Jadhav et al., 2015). Generally, groundwater is considered safer than surface water, as a source of drinking water. However, its quality could be compromised by geogenic contaminants such as arsenic and fluoride (Bretzler and Johnson, 2015). Furthermore, indiscriminate groundwater use has caused problems of overexploitation of aquifers, for example, lacking recharge, which has resulted in an increase in the levels of natural pollutants, due to the water-rock

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interaction and hindering the dilution of waters rich in arsenic and fluorine (Alarcon-Herrera et al., 2013).

Fluorine is one of the most abundant trace elements in the Earth's crust and water, and it is usually present as fluoride ion. The main sources of fluorine in the environment are various minerals that contain fluorides in their structure. Similarly, arsenic is found as a significant constituent in >200 minerals. Both pollutants are released into the groundwater due to water-rock interactions under favorable geochemical conditions (Basu et al., 2014; Vithanage and Bhattacharya, 2015).

The presence of these pollutants in groundwater has affected at least 300 million people in the world (Bibi et al., 2017). It is estimated that 5 million people in Mexico are exposed to fluoride; moreover, around 4% of the population is exposed to arsenic, >5 million people and increasing every day (Jadhav et al., 2015; McClintock et al., 2012). Arsenic has been reported in 14 of the 20 countries in Latin America, with an estimated total exposed population of 14 million people. The level of exposure depends on the maximum permissible limits established. In Argentina, the exposed population of 1.2 to 2 million people was estimated using a maximum limit of 50 μg/L. If this limit were reduced to 10 μg/L, the number of people exposed would increase from 3 to 8 million (Bundschuh et al., 2012). In Mexico, Chile, and Argentina, the co-occurrence in groundwater of fluoride and arsenic has been reported. The origin of this dual contamination is mainly geological (Alarcon-Herrera et al., 2013).

The ingestion of high concentrations of fluoride and arsenic for long periods has adverse effects on health. Because of this, the World Health Organization (WHO) has cataloged them as the most critical inorganic contaminants in water. This institution has established a maximum recommended a limit of 0.5 to 1.0 mg/L for fluoride and 10 μg/L for arsenic in drinking water (WHO, 2011). For its part, the current Mexican regulations establish 1.5 mg/L for fluoride and 25 μg/L for arsenic (SSA, 2000).

The most significant adverse health effects of fluoride ingestion are dental and skeletal fluorosis. The World Health Organization estimates that tens of millions of people suffer from this disorder (WHO, 2014). In Mexico, some populations have been diagnosed with hydrofluorosis in semi-arid central and northeastern regions (Alarcon-Herrera et al., 2013), a condition characterized by stains on teeth, bone deformities and neurological damage in the most severe cases (Bibi et al., 2017).

In the case of arsenic, carcinogenic effects are reported (Banerji and Chaudhari, 2016), being the poor and the infant population the most vulnerable. The most toxic forms in the water of arsenic are the inorganic As(III) species, which accumulates in the organism causing health alterations in some organs (lungs, liver, kidneys, skin) and bladder cancer. In high concentrations arsenic causes gastrointestinal problems, arsenicosis, thinning of the skin and discoloration, numbness of the hands and feet, partial paralysis, blindness, type II diabetes mellitus, cardiovascular diseases, and neurological defects. Furthermore, it is considered teratogenic, that is, it crosses the placenta and enters the metabolic system of the gestational fetus developing (Basu et al., 2014; Ali et al., 2012; Jadhav et al., 2015; McClintock et al., 2012).

The health risks of exposure to each pollutant are well documented. However, there are only a few references about the risk of simultaneous exposure to both pollutants. Some nations as Argentina, China, Mexico, Vietnam, and Cambodia have been affected by the presence of As and F⁻ groundwater contamination (Kim et al., 2012). Arreola-Mendoza et al. (2012) suggest that the toxic potential of these elements increase when chemicals mix, presenting kinetic interactions and altering their average life and toxicity. Moreover, Rasool et al. (2015) discussed the health risk for habitants of Punjab, Pakistan, which drink groundwater with As and F⁻. However, they only describe the effects of excessive and long-term human intake of As and F⁻ for human health, separately.

In Mexico, it has been reported that the infant population in the state of Aguascalientes, as well as in central and northern areas of the country, has an elevated incidence of chronic kidney disease (CKD) due to simultaneous exposure of heavy metals, fluoride, and arsenic, in drinking
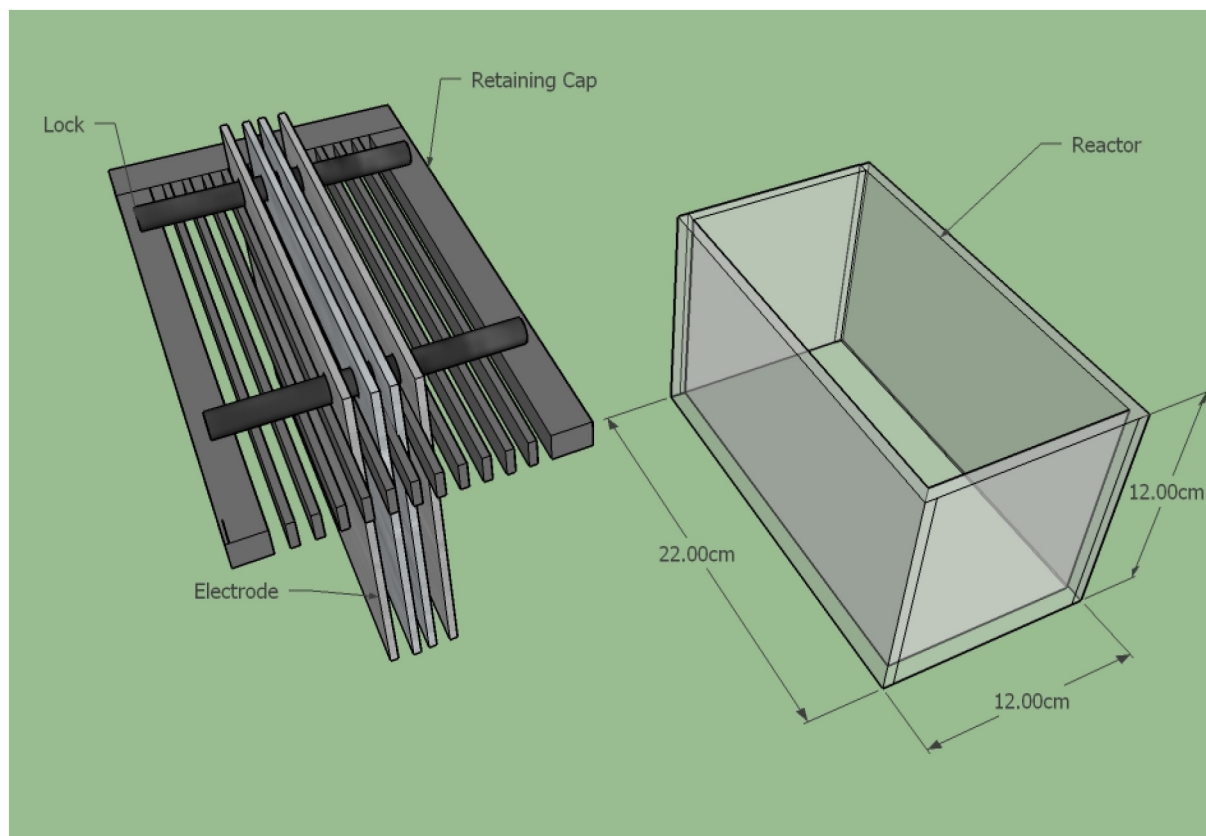

Fig. 1. Design of the electrocoagulation unit.

water. CKD at national level reaches 3.5 new cases per million children under 15 years; while, in Aguascalientes, has been reported 7000 cases per million children under 15 years (Arreola-Mendoza et al., 2012).

Rocha-Amador et al. (2011) evaluated the effect on the induction of apoptosis in immune cells in children who are exposed to both pollutants. Children were sampled from the town 5 de Febrero in the state of Durango and compared with children from another town where the maximum permissible limits established by WHO are met. The average values of arsenic and fluoride in 5 de Febrero were 157.9 μg/L and 8.19 mg/L, respectively, according to a sampling in drinking water. Children exposed to high concentrations of pollutants showed higher levels of apoptosis, which means significant risks to develop problems such as immunodeficiency and cancer.

In the state of Zacatecas, a neighbor of the state of Durango, it is estimated that 80% of the population is exposed to high levels of arsenic in drinking water, and 22% is exposed to fluoride. In this state, a community fluorosis index (CFI) above 1 has been reported; this is similar to that reported in the states of Durango, San Luis Potosí and Aguascalientes (Martínez-Acuña et al., 2016).

Nevertheless, the health effect from simultaneous exposure remains as a controversial issue. Differences in the effects, such as gene expression or cellular processes alterations, may be due to the diversity in biological systems, the doses and the proportion of both pollutants (Barbier et al., 2010).

Exposure to fluoride in Durango state, located in northern Mexico, is evident as there are cases of severe dental fluorosis in children. Fluoride and arsenic concentrations of 1.5 to 5.67 mg/L and 0.2 to 0.4 mg/L and up have been reported, respectively; in the groundwater of the Guadiana Valley (Armienta and Segovia, 2008; Alarcon-Herrera et al., 2013).

The selective removal of fluoride and arsenic has been evaluated by several methods (coagulation/chemical precipitation, electrocoagulation, adsorption, ion exchange, membrane technologies), which are efficient. However, simultaneous removal represents a considerable challenge since, in general, there is a decrease in the removal efficiency of one of the two pollutants.

The removal technologies that have been used to remove these two pollutants simultaneously are coagulation, adsorption, membrane filtration and electrocoagulation (Jadhav et al., 2015). Among these technologies, electrocoagulation (EC) has gained much interest in recent years as an efficient removal method for fluoride and arsenic. This technology is easy to operate at both scales (batch and continuous flow), it is inexpensive by not requiring the addition of chemicals, and it does not produce a large quantity of sludge that would require a subsequent treatment. Besides, its automation is possible (Thakur and Mondal, 2017; Vasudevan et al., 2010).

The EC method consists of the in situ generation of coagulant species by applying an electric current on the electrodes. The generated species form a complex with the contaminants and then precipitate (Hakizimana et al., 2017). The efficiency of the EC processes depends on the operational parameters such as initial concentration, current density, pH, electrode materials; number, arrangement of the electrodes and the distance between them; conductivity of the water, as well as the electrolysis time (Ali et al., 2012; Banerji and Chaudhari, 2016; Islam, 2017; Kobya et al., 2011).

Thakur and Mondal (2017) evaluate the performance of the EC processes for the individual and simultaneous removal of fluoride and arsenic. Their results revealed that EC is suitable for both cases (individual and simultaneous removal), and notice that EC achieved better removal efficiencies for arsenic than for fluoride (98.51 and 88.3, respectively). In the same way, Zhao et al. (2011) used a combination of electrooxidation and electrocoagulation. Their EC unit was built with Al and Fe electrodes and reported 77.7% removal for fluoride and 99% for arsenic.

The current problem of contamination by fluoride and arsenic, as well as the lack of efficient and economically viable technologies for vulnerable populations, makes it necessary to generate proposals to solve this problem through the evaluation of conventional technologies. In the present study, an electrocoagulation device was designed and constructed for the simultaneous removal of fluoride and arsenic from well water, to achieve potable quality according to Mexican regulations.

Table 1
Factors and levels of the factorial design.

Factor	Symbol	Low level (−1)	High level (+1)	Unit
As concentration	CAs	40	80	μg/L
F⁻ concentration	CF	5	10	mg/L
Current density	C.D.	0.5	4.5	mA/cm²
Time	T	5	15	min

2. Materials and methods

2.1. Sampling and water characterization

Water samples were obtained from wells in the Guadiana Valley, Durango state, Mexico. Conductivity and pH measurements were carried out using an ORION VERSA STAR meter (Thermo Scientific). Calibration by 3 points was performed daily at the beginning of the measurements. Calibration standards (pH and conductivity) were purchased from Thermo Scientific.

Major cations (Na^+, K^+, Ca^{2+}, Mg^{2+}) quantification was performed by atomic absorption spectrophotometry (AAS - GBC model XplorAA). Calibration standards of each element were purchased from Merck. Quantification of CO_3^{2-} and HCO_3^- was carried out by volumetric titration with sulfuric acid (1 N). Alkalinity and major cations were carried out using the methodology NMX-AA-036-SCFI-2001 and the standard method for water analysis (APHA/AWWA/WEF, 2012).

To measure the concentration of Cl^-, SO_4^{2-} and NO_3^- anions, a Thermo Scientific ion chromatograph (Dionex Integrion HPIC) was used, with conductivity detector, isocratic elution, IonPack AsII-HC column 2 × 250 mm and ion suppressor AERS 2 mm, according to method 300.0 of the Environmental Protection Agency (EPA, 1993). Dionex Seven Anion standard was purchased from Thermo Scientific.

The fluoride determination was carried out by potentiometric method using an ISE Fluoride Ion Plus electrode (Orion VersaStar-Thermo Scientific multiparameter equipment). Arsenic quantification was performed by AAS (Avanta GBC model XplorAA with hydride generator model HG300). Calibration standards were purchased from Fluka Analytical. All standards used were traceable to NIST reference materials.

2.2. Preparation of solutions

All the reagents used were analytical grade. With stock solutions were prepared, working solutions at the concentrations required for the process by proper dilution in well water. NaCl was used as a supporting electrolyte for the conductivity adjustment until reaching an approximate value of 1200 μS/cm. The pH adjustment was carried out with 1 M HCl.

2.3. Design and construction of the electrocoagulation cell

Fig. 1 shows the schematic representation of the electrocoagulation cell. The system comprises *a*) a rectangular acrylic reactor of 22 × 12

Table 2
Factors and values of experimental design levels of simultaneous removal tests with pH adjustment.

Factor	Symbol	Low level (−1)	High level (+1)	Unit
F⁻ concentration	CF	5	10	mg/L
Time	T	5	15	min
pH	pH	3	5	–

Table 3
Characterization of well water.

Coordinates		pH	Conductivity (µS/cm)	Fluoride (mg/L)	Total arsenic (µg/L)	Nitrate (mg/L)	Chloride (mg/L)
Lenght	Latitude						
104.602	24.062	7.7	409.9	4.18	24.48	14.07	18.43

				Ionic balance			
HCO_3^- (mg/L)	SO_4^{2-} (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	Na^+ (mg/L)	K^+ (mg/L)	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)
118.80	34.15	18.43	14.07	45.78	6.44	35.98	0.82

× 12 cm (1.5 L, as effective volume); *b*) a 3D printed PLA (polylactic acid) cap with 0.5 cm spacing, to insert the electrodes; *c*) iron and aluminum electrodes, the first ones were 17.5 × 12.5 cm carbon steel A36 plates with 0.5 cm thickness, and the second ones were aluminum plates (same dimensions), with a thickness of 0.3 cm. The plates were perforated to be secured in the cover by a lock bar.

Iron electrodes were treated with 1 M HCl. Aluminum electrodes were immersed in a 20% NaOH solution. Subsequently, the electrodes were washed with distilled water to clean the surface. In each experimental run, the electrodes were abraded with sandpaper and washed with distilled water, and then the plates were cleaned with ethyl alcohol to remove any trace of oily residue that could be adhered to them during handling.

2.3.1. Operating conditions

Tests were carried out in the electrocoagulation reactor using four electrodes: two iron electrodes (anode and cathode) and two aluminum electrodes (anode and cathode), in order to generate the largest amount of coagulant species for each of the contaminants; iron was used for arsenic species and aluminum for fluoride.

Electrodes were connected to a direct current source (GW INSTEK GPC 30300) through a monopolar connection in parallel. Two Fluke® multimeters were used, one to verify the imposed current and the other to record the voltage. The active area of the electrodes was 577.2 cm². The experiments were carried out with constant stirring (150 rpm) using a magnetic stirrer. After the treatment process, the water was filtered through a Whatman® 42 filter (2.5 µm). Final concentrations of fluoride and arsenic and pH were determined.

The removal yield was calculated according to Eq. (1).

$$\%\text{removal} = \frac{C_f - C_i}{C_i} * 100 \tag{1}$$

where C_f is the final concentration and C_i the initial concentration.

2.4. Experimental design

The experimental design for the electrocoagulation tests consisted of a 2^4 factorial design without replication, using a level of significance $\alpha = 0.05$; four factors with two levels, low (−1) and high (+1) (Table 1). According to Montgomery et al. (2013), it is feasible to obtain reliable estimates when performing an experimental design without replication, as long as the ranges of the factor levels are quite broad.

2.4.1. Effect of pH

Tests of initial pH variation were carried out at constant arsenic concentration (80 µg/L), the experimental design of which consisted of a factorial design 2^3 with one repetition (Table 2).

2.5. Statistical analysis

The statistical analysis of the results obtained in the electrocoagulation tests was carried out in the JMP-Pro® software. The fit of the model was expressed by R-square (r^2), and the statistical significance of the model obtained and factors involved in the model, was examined using F-value and *P*-value at 95% confidence level.
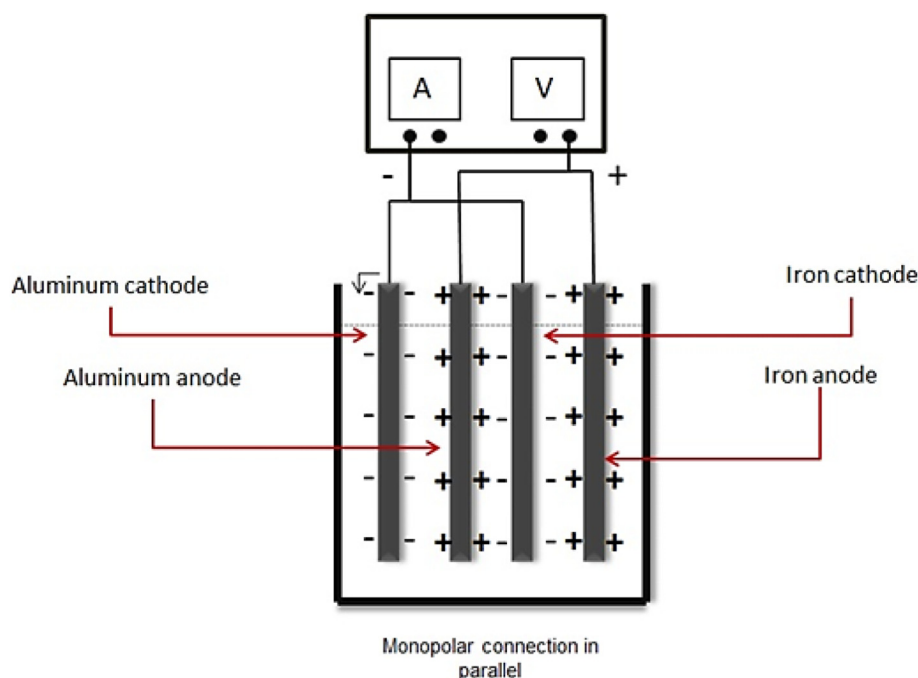

Fig. 2. Graphical representation of the type of connection used in simultaneous removal tests and the arrangement of the electrodes.

3. Results and discussion

3.1. Characterization of well water

The electrocoagulation process was evaluated with well water from the Guadiana Valley, in which fluoride and arsenic were present in co-occurrence (4.18 mg/L and 24.48 μg/L, respectively). The water sample selected for the study was classified by its ionic balance, according to Piper's diagram, as sodium-bicarbonated. The geographical coordinates of the sample site and the general characterization of the well water are shown in Table 3.

3.2. Simultaneous removal of arsenic and fluoride

Based on the results obtained in previous tests of independent fluoride and arsenic removal (data not shown); the aluminum electrodes were arranged in such a way that they had two active faces, to generate more aluminum species to promote the removal of fluoride, as shown in Fig. 2.

Table 4 shows the results from the simultaneous removal tests. Even when the surface area of the aluminum electrodes was increased, the percentage of fluoride removal was not satisfactory (from 13.0 to 64.8% removal). It was not possible to reach concentration levels below the maximum permissible limit established by the Mexican norm (NOM-127SSA1-1994), even with the most favorable conditions (low initial concentration, high current density and longer time). This is probably because fluoride removal efficiencies decrease with the presence of arsenate, which competes for adsorption sites in the aluminum hydroxides formed during the reaction; similar observations were reported by Vasudevan et al. (2011). Dhadge et al. (2018) report the decrease of the initial fluoride concentration from 10 to 0.92 mg/L in 35 min with a current density of 625 A/m^2; these operating conditions, however, increase energy consumption and reduce the electrode's useful life, thus increasing the overall cost of treatment.

According to the statistical analysis of the obtained model for fluoride removal ($r^2 = 0.99$; $F = 37.59$; $p = 0.0004$), current density ($F = 260.41$; $p < 0.0001$), treatment time ($F = 86.83$; $p = 0.0002$) and the interaction between both factors ($F = 9.80$; $p = 0.0259$) are the most relevant factors during EC process. For arsenic removal, the obtained model analysis ($r^2 = 0.97$; $F = 14.30$; $p = 0.0045$) shows that current density ($F = 90.07$; $p = 0.0002$) was the most relevant factor, while treatment time and the interaction between both factors had the same effect ($F = 13.74$; $p = 0.0139$).

Arsenic removal, under most operational conditions, initial concentration, current density and time, reached concentration levels below 25 μg/L required for drinking water in Mexico. The best results were obtained with the higher current density, reaching approximately 100% arsenic removal, without being affected by the initial concentration or the treatment time. According to Dhadge et al. (2018), the sacrificial anode generates the coagulant species, while the cathode facilitates the generation of hydrogen, which favors the process of removal. High efficiencies in arsenic removal may be due to the coagulant species generated in the iron electrodes, which possess a high affinity for arsenic, resulting in the adsorption of the element on the surface of the coagulant, favoring the removal process (Can et al., 2014).

3.3. pH effect

An important aspect to consider in the simultaneous removal tests was the behavior of the pH in the solution. For tests at low current density, there is no increase in the pH value, even in those that were carried out up to 15 min. According to Kim et al. (2016) under alkaline conditions, the electrocoagulation process has a buffer capacity that reduces changes in pH. This fact and the low current density applied (0.5 mA/cm^2), that does not promote a high generation of H$_2$ in the cathodes, can explain a constant pH in the low current density tests. However, in tests where the current density was 4.5 mA/cm^2 a change

Table 4
Results of tests of simultaneous removal of fluorides and arsenic.

Exp	CF	CAs	C.D.	t	Initial pH	Final pH	[F]$_f$ (mg/L)	[As]$_f$ (μg/L)	% Removal F	% Removal As
1	1	1	1	1	8.28	8.77	3.52	N.D.	64.8	≈100
2	1	1	1	−1	8.07	8.39	5.82	N.D	41.8	≈100
3	1	1	−1	1	8.23	8.09	7.61	26.16	23.9	83.65
4	1	1	−1	−1	8.08	8.15	7.90	38.20	21.0	76.12
5	1	−1	1	1	8.13	8.67	3.64	N.D.	63.6	≈100
6	1	−1	1	−1	7.97	8.23	5.69	N.D.	44.6	≈100
7	1	−1	−1	−1	8.24	8.12	7.74	26.7	22.6	83.31
8	1	−1	−1	1	8.10	8.10	6.84	5.84	31.6	96.35
9	−1	−1	1	1	8.28	9.01	2.23	N.D.	55.4	≈100
10	−1	−1	1	−1	8.26	8.15	3.40	N.D.	32.0	≈100
11	−1	−1	−1	−1	8.11	8.10	4.35	18.36	13.0	77.05
12	−1	−1	−1	1	8.19	8.04	3.48	8.58	30.4	89.27
13	−1	1	1	−1	7.97	8.23	2.77	N.D.	44.6	≈100
14	−1	1	−1	1	8.16	8.04	3.43	12.8	31.4	84.00
15	−1	1	1	−1	8.19	8.08	3.97	34.5	20.6	56.87
16	−1	1	1	1	8.20	8.97	2.08	N.D.	58.4	≈100

N.D. Not detected. ≈ Approximate percentage estimated, final concentration below the detection limit (0.6 μg/L).
Exp = experiment, i = initial, f = final.

in pH is observed, which is more noticeable in those experimental runs in which the treatment time was longer, due to higher generation of H$_2$.

The highest fluoride removal efficiencies were obtained when a current density of 4.5 mA/cm^2 was applied, which coincides with the most significant increases in the final pH value (Table 4), being these: 64.8% for initial concentration of 10 mg/L (Exp 1); 58.4 and 55.4% for initial concentration of 5 mg/L (Exp 9 and 16), the final pH values of which were 8.77, 8.97, and 9.01, respectively.

3.3.1. Effect of initial pH on the removal process

Due to the latter results, simultaneous removal tests were performed adjusting the initial pH of the solution to enhance the removal of fluoride. Tests were carried out at two initial pH values, 3 and 5. The results showed an increase in the fluoride removal efficiency for both pH values, with pH 3 being the one that enhances the fluoride removal to a greater extent (Fig. 3), which coincides with observations by Kim et al. (2016) and Thakur and Mondal (2017). They mention that at a pH >3, *boehmite*, AlO(OH), is present in the solution, which contains the structures of hydroxyl groups necessary for adsorption; furthermore, at this pH fluorine also starts to appear as F$^-$ and this is constant for all pH values >3 in solution. Using aluminum electrodes, a major drawback
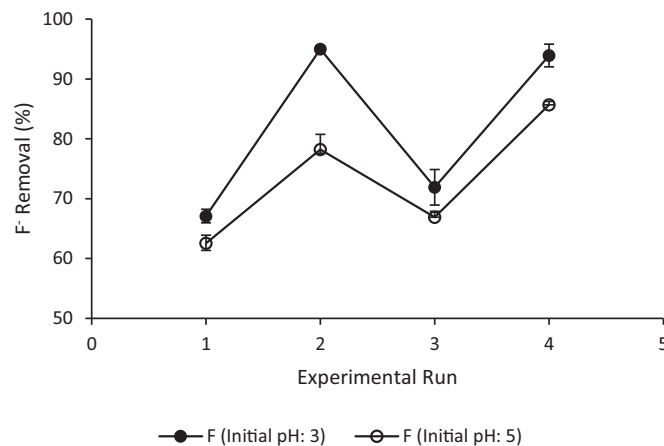

Fig. 3. Effect of initial pH on the fluoride removal efficiency. All the points in the chart represent the mean of the results ($n = 2$) and error bars represent ±1 standard deviation from the mean.
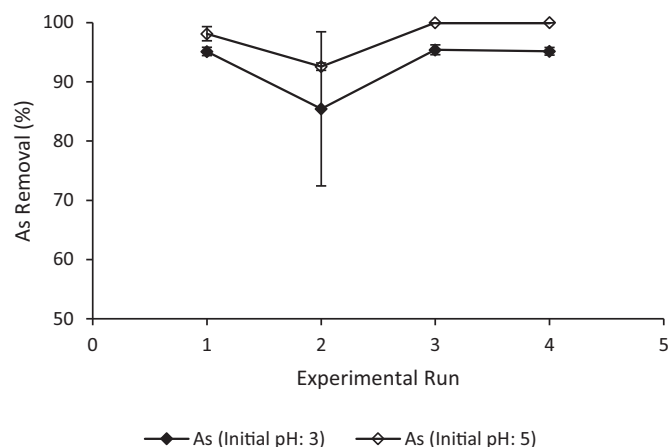
Fig. 4. Effect of initial pH on the arsenic removal efficiency. All the points in the chart represent the mean of the results (n = 2) and error bars represent ±1 standard deviation from the mean.

affecting removal yield is that, at values higher than pH 8, anionic species begin to appear, $Al(OH)_4^-$, which produces a repulsion of charges between that species and fluoride (Thakur and Mondal, 2017).

The initial pH 5 was more favorable for the removal of arsenic (Fig. 4) because the pH increase was greater from this value. The final pH reached from the initial pH 5 was in the range of 6.74 to 7.45 (Table 5). To carry out the removal of arsenic, it must be oxidized from As(III) to As(V) in almost all conventional removal processes, which involves the addition of reagents to achieve this. During the electrocoagulation process, the oxidation occurs during the process without the need to add reagents or intermediate stages. When that oxidation is carried out, the species $H_2AsO_4^{-2}$ is present in the solution in a pH range of 6 to 8, while the $Fe(OH)_3$ is constant in the solution from pH 3 (Can et al., 2014).

The removal of arsenic is promoted as a result of the increase in pH from 3 to 7, so, in a process with initial acidic conditions, it is necessary for the pH to increase throughout the process to increase the removal of arsenic, which is very likely to occur due to reactions at the cathode (Thakur and Mondal, 2017). Vasudevan et al. (2012) obtained a similar result, concluding that arsenate removal efficiency increases when pH

increases from a value of 2 to 7, value at which the highest removal of arsenate was obtained. According to Banerji and Chaudhari (2016), the oxidation of As(III) to As(V) is higher at a pH of 7 with the use of iron electrodes. The presence of the Fe(IV) oxidant species, which is an intermediary in the oxidation of Fe(II) to Fe(III), first oxidizes As(III) to As(IV), and if there is enough Fe(IV), the arsenic is wholly oxidized to As(V) and then the complexation and precipitation with Fe(III) is carried out.

The model obtained in the statistical analysis of the data ($r^2 = 0.98$; F = 74.58; p < 0.0001) shows that time (F = 359.2462; p < 0.0001) and pH (F = 60.2209; p < 0.0001) are the most relevant factors during EC process for fluoride. By increasing the treatment time and decreasing the initial pH value, the removal is maximized.

Although the tests at pH = 3 showed the highest removal efficiency (final concentrations below 1.5 mg/L for fluoride at 15 min of treatment), the selection of some of them as the optimal electrocoagulation process is not entirely feasible since final pH does not comply with Mexican regulation values of 6.5–8.5 (SSA, 2000). Based on the final pH of the water, the optimum conditions of the process were: pH 5, initial fluoride concentration of 5 mg/L, initial concentration of arsenic 80 μg/L, current density 4.5 mA/cm^2 and 15 min of treatment. With these operating conditions, satisfactory results were obtained: average final pH of 7.33, final fluoride concentration < 1.0 mg/L, almost total removal of arsenic, with a removal efficiency for fluorine of 85.68% and approximately 100% removal of arsenic.

Electrocoagulation has shown to simultaneously and efficiently remove fluoride and arsenic from well water. However, some drawbacks of its use, such as the consumption of electrical energy, may involve an additional cost and some technical problems associated with its installation and maintenance (Kabir and Chowdhury, 2017). Electrode passivation when applying direct current is another problem that must be considered since it reduces the efficiency of the process by increasing energy consumption. This drawback can be remedied by replacing the use of direct current with alternating current (Kamaraj et al., 2013).

4. Conclusions

The electrocoagulation cell prototype is effective in fluoride and arsenic simultaneous removal in well water. In the present EC process the most critical factors are the pH, the current density and the treatment time. The optimum operating conditions for well water samples with initial fluoride and arsenic concentrations of 5 mg/L and 80 μg/L, respectively, were: *a*) initial pH of 5, *b*) 15 min of treatment and *c*) current density of 4.5 mA/cm^2. The removal yield achieved under these conditions was 85.68% for fluorine and approximately 100% for arsenic, obtaining final concentrations lower than the values recommended by WHO and national regulations. The initial pH adjustment did not have a significant effect on the removal of arsenic; however, the acidic pH conditions favor the removal of fluoride reaching a maximum removal of 94.98% to achieve a final fluoride concentration < 1.0 mg/L in the water.

Table 5
Simultaneous removal test results with initial pH adjustment.

Exp	CF	t	pH$_f$	[F]$_f$ (mg/L)	% Removal F	[As]$_f$ (μg/L)	% Removal As
pH 3							
1	1	−1	6.14	3.21	67.90	4.28	94.65
2	1	−1	6.12	3.37	66.30	3.49	95.64
3	1	1	6.35	0.541	94.59	19.0	76.25
4	1	1	6.49	0.464	95.36	4.28	94.65
5	−1	−1	6.31	1.30	74.00	3.194	96.01
6	−1	−1	6.03	1.51	69.80	4.13	94.84
7	−1	1	6.15	0.371	92.58	3.5	95.63
8	−1	1	6.33	0.237	95.26	4.23	94.71
pH 5							
1	1	−1	6.89	3.65	63.50	2.165	97.29
2	1	−1	6.91	3.84	61.60	0.813	98.98
3	1	1	7.45	2.0	80.00	5.59	93.01
4	1	1	7.65	2.36	76.40	6.29	92.14
5	−1	−1	6.75	1.69	66.20	N.D.	≈100
6	−1	−1	6.74	1.62	67.60	N.D.	≈100
7	−1	1	7.35	0.696	86.08	N.D.	≈100
8	−1	1	7.32	0.736	85.28	N.D.	≈100

N.D. Not detected. ≈ Approximate percentage estimated, final concentration below the detection limit (0.6 μg/L).
CF = Initial fluoride concentration.
Exp = experiment, f = final.

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