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Investigation on Efficiency of electrocoagulation using monopolar iron and aluminum electrodes for fluoride removal from water

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Abstract: The present work was carried out to assess the efficiency of iron (Fe) and aluminum (Al) electrodes in electrocoagulation (EC) process for removal of fluoride using aqueous solutions. Several operating parameters such as type of electrode, initial concentration of fluoride, current density, electrolysis time and NaCl amount were studied to achieve optimum conditions of high fluoride removal and energy consumption. Using Al electrodes, fluoride removal efficiencies of 80%, 83% and 87% were achieved while the Fe electrodes removed 51%, 54% and 56% using initial fluoride C_0 =15, 10 and 5 mg/L, respectively. Besides, the increase of initial fluoride has caused a decrease of fluoride removal efficiency. By adding 0.5 g/L of NaCl to fluoride solutions, fluoride removal increased from 45 to 61.76% using Fe electrodes, while this efficiency passed from 56 to 66.47% using Al electrodes. Energy consumption was voltage, intensity and time dependency. In optimum conditions (t = 60 min and DC= 27.8 mA/cm^2), Al electrodes could remove up to 100% while Fe electrodes removed 77.4% during the whole of electrolysis. . Therefore, Aluminum electrodes were more efficient in fluoride removal using EC but they require more energy during the EC process.

1. Introduction

Depending on its concentration, fluoride in water can have both beneficial and harmful effects on the environment and human health. The concentration of fluoride around 1 mg/L in drinking water can help in teeth decay prevention (Graça *et al*., 2019). However, the long-term consumption of water containing an excess of fluoride can lead to fluorosis of teeth and bones (WHO and IPCS, 1996). In addition, the role of fluoride in reducing the risk of dental caries, especially among children, is well recognized (Pehrsson *et al.*, 2006). At this regard, a limit value of fluoride in drinking water has been fixed to 1.5 mg/L by the World Health Organization (Nemerow *et al*., 2009). Fluoride removal from contaminated water can be carried out using various processes such as adsorption, chemical precipitation, reverse osmosis, electrocoagulation, and ion exchange (Loganathan *et al*., 2013). Electrocoagulation (EC) process is one of the electrochemical methods developed for water treatment because it is based on in situ production of metal ions by progressive dissolution of the sacrificial anode of an electrochemical cell receptor under the effect of an electric current. The electrocoagulation process has been suggested as a good alternative to conventional chemical coagulation in water treatment (Mills *et al*., 2000), (Miwa *et al*., 2006). Several studies showed the effectiveness of electrocoagulation on defluoridation of water supplies and industrial wastewaters (Drondina and Drako, 1994) ,(Mameri *et al*., 1998), (Yang and Dluhy, 2002), (Shen *et al*., 2003), (Un *et al*., 2013), (Sandoval *et al*., 2014).The application of electrochemical processes to remove pollutants from water has been gaining an increasing interest due to its low associated costs, high efficiency, and relatively simple operation and control (Rajeshwar *et al*., 1994). Most EC applications use iron or aluminum electrodes because of their low cost and high valences of the cations that they generate.

In this study, we investigated the electrochemical removal of fluoride in water using monopolar Fe and Al electrodes as anode and cathode. In addition, the influence of operating variables such as current density, initial fluoride concentration, electrodes nature and NaCl amount on EC efficiency and energy consumption has been studied.

2. Materials and methods

2.1 Experimental device

An acrylic cubic shaped flat bottom vessel (7 cm long, 5.9 cm larger, 0.5 cm thickness) was used as a reactor in electrocoagulation (EC) process as shown in **Figure 1a**. In the reactor (0.4 L of capacity), were putting two homogenous aluminum or iron plate electrodes as cathode and anode, with an inter-electrode distance fixed to 1 cm on supporting rod. The electrochemical reactor (14.7 cm of height) was made of Plexiglas. The electrodes were dimensioned as follows: 15 cm × 4.7 cm × 1 mm. The thickness of each electrode and the effective surface area was 1 mm and 55 cm^2, respectively. A constant current was fixed using DC power supply (LODESTAR, LP3005D) and stirring was carried out using a magnetic stirrer. The schematic representation of experimental setup is shown in **Figure 1b**. This experimental device has been setup and used in previous studies conducted in our laboratory on fluoride removal by EC process (Sanou *et al*., 2022; Sedou *et al*., 2022).

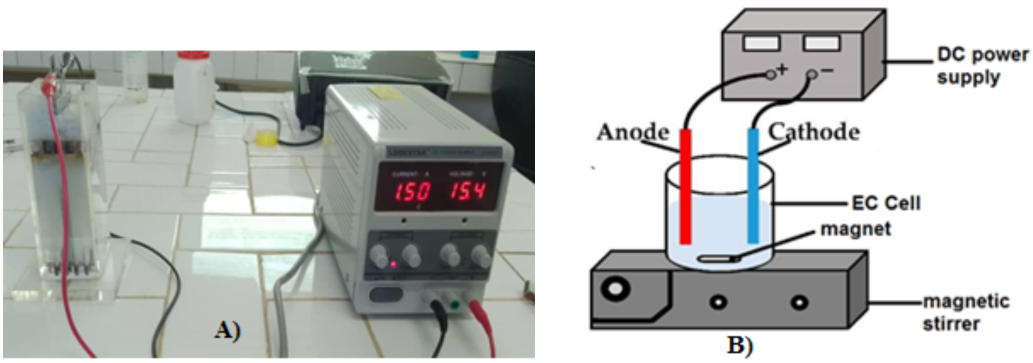

Figure 1. Experimental device of EC reactor (A) EC reactor (B) Assembly diagram of the EC

2.2 Fluoride removal experiments

The feed solution was prepared by dissolving the appropriated mass of sodium fluoride (Sigma-Andrich; assay ≥ 99%) in distilled water and adjusting the conductivity with 0.5 g/L NaCl solution. At the beginning of each run, 350 mL of water (11.7 cm height) was fed in the EC reactor and 0.5 g/L of NaCl was added to increase the conductivity of the solution. The current density was adjusted to 14, 18.5 and 27.8 mA/cm^2 using a digital DC power supply.

Fluoride removal experiments were carried out with Fe and Al electrodes as anode and cathode during electrolysis time of 60 min. Treated fluoride solutions samples were collected each 10 minutes and Fluoride concentration was determined. An Ultraviolet-Visible spectrophotometer (DIV-UV5600) analyzed fluoride concentration using molecular absorption spectrometry with zirconium and Erichrome-cyanine R. All analyses were carried out twice according to APHA standard methods (APHA, 2012). Experiments were repeated three times and average values were used for calculations and the representation of graphs or figures.

Fluoride removal efficiency (%) was evaluated using the following relation:

$$Removal\ efficiency\ (\%) = \frac{C_0 - C_f}{C_0} \times 100 \qquad \textbf{Eqn. 1}$$

Where C_0 and C_f represent respectively the initial and final fluoride concentrations (mg/L).

Energy consumption (E) expressed in kWh/m^3 was calculated as follow:

$$E = \frac{\mathrm{U \times I \times t}}{\mathrm{V}} \qquad \textbf{Eqn. 2}$$

Where U: Voltage (V); I: Current intensity (A) and V: Volume of water (m^3) and t: time (h)

3. Results and Discussion

3.1 Effects of operating conditions on fluoride removal efficiency

3.1.1. Influence of electrode type

In electrocoagulation process, the nature of electrodes is an influent parameter which affect significantly the efficiency of fluoride removal (Sanou *et al*., 2022). In this study, both iron and aluminum electrodes were used as anode and cathode during the electrolysis time and their performance in fluoride removal were assessed during the whole of experiment. Experimental results showed that all removal efficiencies obtained with Al electrodes were up to those obtained with Fe electrodes indicating the high efficiency of Al electrodes (**Fig. 2**). Indeed, Fe and Al electrodes removed 56% and 87.5% of fluoride for 60 min when DC=14 mA/cm^2 was applied (**Figure 2a**).

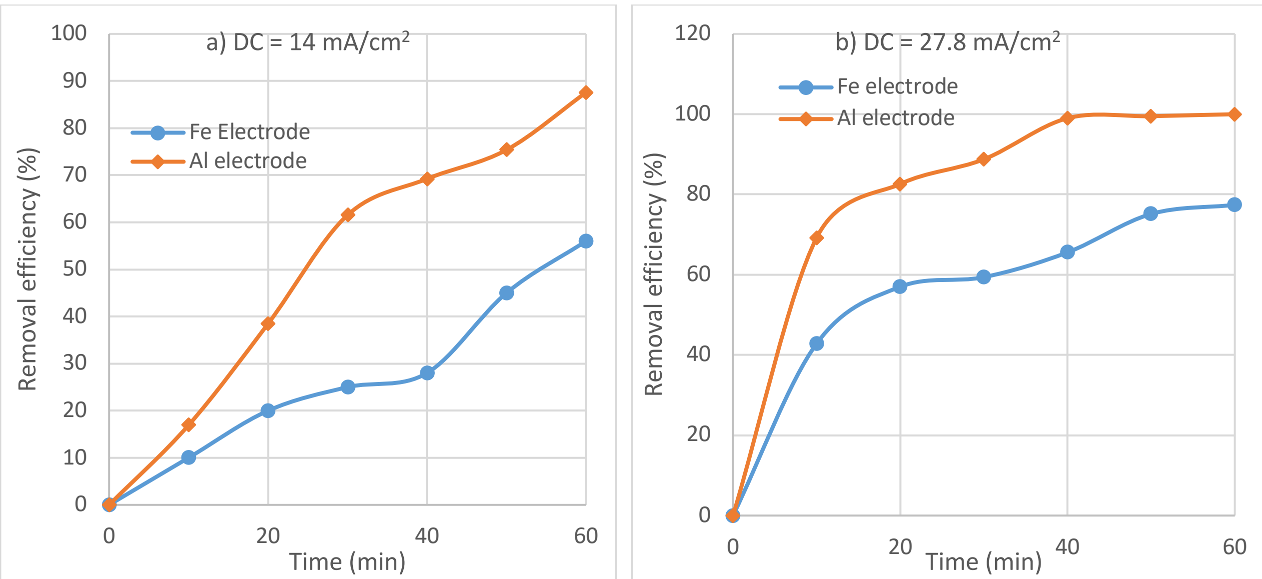

Figure 2. Effect of electrode type on fluoride removal efficiency using Al and Fe electrodes with C_0 = 5 mg/L; 0.5 g/L NaCl, and V=350 mL, (**a**) DC = 14 mA/cm^2 (**b**)) DC = 27.8 mA/cm^2

In addition, when the DC was increased up to 27.8 mA/cm^2, Al electrodes could remove up to 100% while Fe electrodes removed 77.4% during 60 min (**Fig. 2b**). All results revealed the efficiency of Al electrodes due to their low density. High efficiency of Al electrodes in EC compared to Fe electrodes in this study is in agreement with data obtained in literature (Essadki *et al*., 2009; Bazrafshan *et al*., 2012; Takdastan *et al*., 2015).

3.1.2 Effect of current density

The current density is electric current supplied per unit surface area of an electrode. It is a critical parameter of electrochemical process because it is directly linked to the imposed current and the active surface of the electrodes used. The current density (DC) determines the dosage coagulant level, the rate of bubble production, size and growth of the floc which may affect the efficiency of electrocoagulation process (Khatibikamala *et al*., 2010). From **Figure 3**, an increase in fluoride removal was observed with the increase in current density from 14 to 27.8 mA/cm^2 using each electrode. This result can be explained by the fact that more current was supplied to electrodes, more coagulant dosage is high into water (Un *et al*., 2009; Sedou *et al*., 2022).

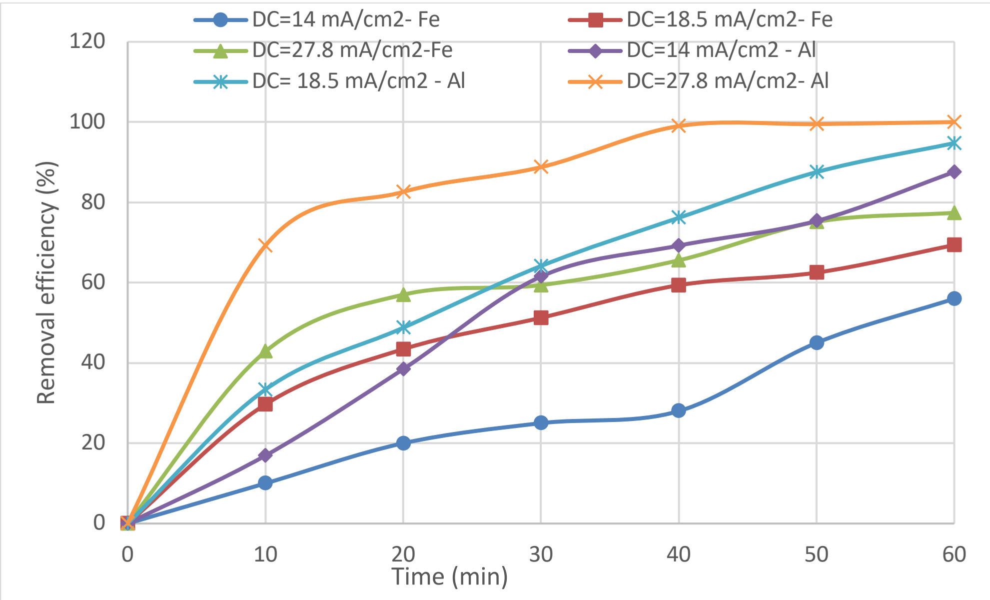

Figure 3. Effect of current density on Fluoride removal by Al and Fe electrodes using C_0 = 5 mg/L, pH= 7.68; 0.5 g/L NaCl, and V=350 mL.

By comparing the curves in **figure 3**, we noticed that a maximum treatment yield (100%) was achieved using Al electrodes under 27.8 mA/cm^2 during 60 min of electrolysis. In addition, Al electrodes showed higher removal efficiency at any time comparatively to Fe electrodes. This can be described to the fact that the amount of aluminum oxidized increased at higher voltage, resulting in a greater amount of precipitate for the removal of pollutants. Besides, it was demonstrated that bubbles density increases and their size decreases with increasing current density, resulting in a greater upwards flux and a faster removal of pollutants and sludge flotation (Malakootian and Yousefi, 2009; Bazrafshan *et al*., 2012). So, fluoride removal was more efficient at high values of current density. When the current density increases, the production of cations (Al^{3+}, Fe^{2+} or Fe^{3+}) on electrodes also increases. This leads to production of $Al(OH)_3$ and $Fe(OH)_3$ flocs in the solution and hence efficiency of the EC process is improved. Consequently, we concluded that Al electrodes and high current density constitute optimum variables for efficient fluoride removal using EC process. But, the use of high DC caused high energy consumption and calculations revealed an anode decrease of 4.86% when 27.8 mA/cm^2 was applied.

3.1.3. Effect of initial fluoride concentration

The initial fluoride concentration in water has been reported to greatly influence the defluoridation efficiency (Battula *et al.*, 2014). To assess the influence of initial fluoride on EC efficiency, three initial concentrations (5, 10 and 15 mg/L) were tested, whereas other parameters such current density and NaCl concentration remain fixed. From **Figure 4**, which shows the fluoride removal efficiency over electrolysis time; it was observed that the solution containing the highest fluoride concentration required more Al^{3+} or Fe^{3+} to be dissolved as compared to others having lower fluoride concentrations.

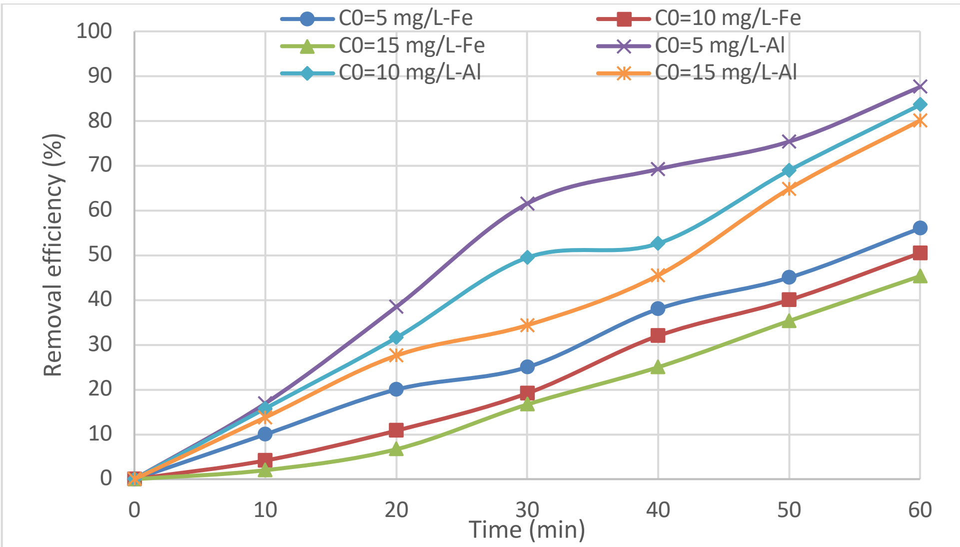

Figure 4. Effect of initial fluoride concentration on removal efficiency using Al and Fe electrodes with DC=14 mA/cm^2, 0.5 g/L NaCl, and V=350 mL.

The Figure 4 demonstrates that the rate of defluoridation was significantly influenced by the initial concentration of fluoride. Experimental results indicate the decrease of EC efficiency when initial fluoride increased at any time of electrolysis (**Fig. 4**). Using Al electrodes, fluoride removal efficiencies of 80%, 83% and 87% were achieved with C_0 =15, 10 and 5 mg/L, respectively during 60 min (**Fig.4**). While, Fe electrodes removed 51%, 54% and 56% using the same respectively initial fluoride (**Fig.4**).

The decrease of fluoride removal with initial fluoride increase was demonstrated in previous study under similar conditions (Vasudevan *et al.*, 2009). High EC efficiency with lower fluoride concentrations can be explained by the theory of dilute solution. Indeed, the formation of the diffusion layer at the vicinity of the electrode causes a slower reaction rate in dilute solution, while this diffusion layer has no effect on the rate of diffusion or migration of metal ions to the electrode surface in concentrated solution (Chaudhary *et al*., 2003; Nouri *et al*., 2010; Bazrafshan *et al*., 2011). Higher of EC efficiency was obtained using Al electrodes with any fluoride amount in the feed solution comparatively to Fe electrodes which were less efficiency in fluoride removal. By varying the value of current density, experimental results were given by **figures 5a** and **5b**. It was clearly observed an increase of fluoride removal efficiency when the initial fluoride decreased from 15 to 5 mg/L. In addition, the increase of current intensity caused an increase in EC efficiency due to high energy consumption.

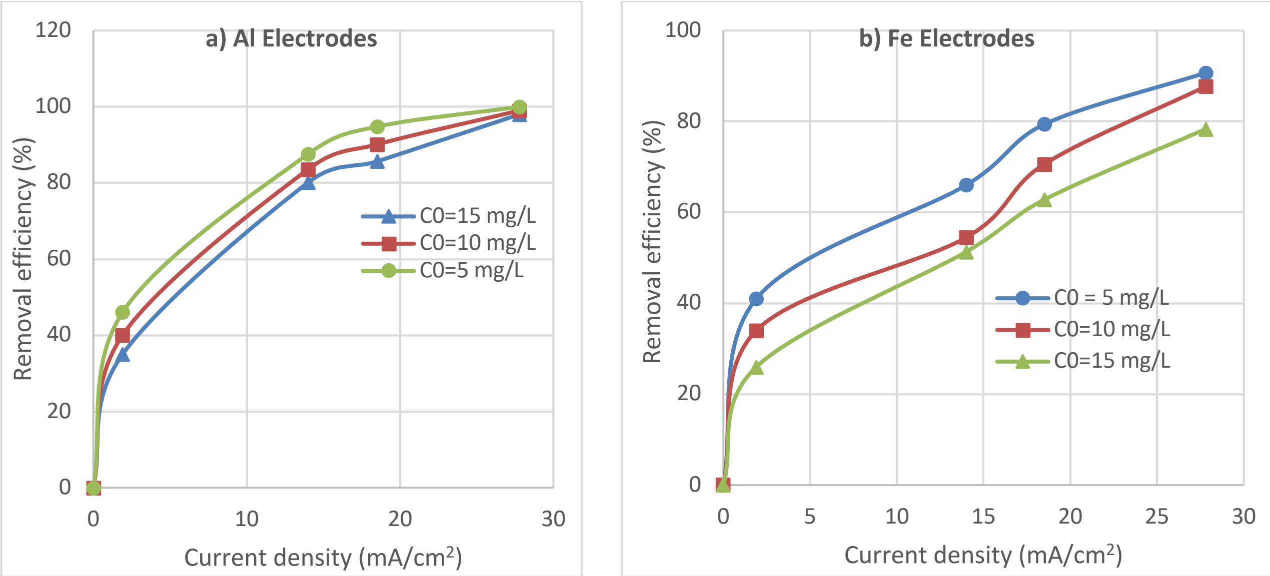

Figure 5. Influence of initial fluoride amount on removal efficiency using Al and Fe electrodes with current density varying from 14 to 27.8 mA/cm^2, and V=350 mL, **(a)** Al Electrodes and **(b)** Fe Electrodes

3.1.4 Effect of NaCl

The addition of an electrolyte increases the electrical conductivity of the medium and reduces its resistance to ensure better dissolution of anodes and to avoid the formation of an insulating layer on the surface of electrodes and thus increase the resistance of the electrochemical cell. The highest fluoride removal percentages (66.47% and 61.76%) were obtained by adding 0.5 g/L of NaCl to fluoride solution using Al and Fe electrodes, respectively (**Figure 6**). Without NaCl (0 g/L), 56% and 45.5% of fluoride were removed by Al and Fe electrodes, respectively during the EC process (**Fig. 6**). This result can be explained by the phenomenon of corrosion and oxidation under the effect of chloride ions on electrodes which induce the flocs formation and inhibit the formation of a passive film on the surface (Mameri *et al*., 1998). The addition of NaCl increases the flow of current in the EC cell, which leads to an overconsumption of the electrodes under the effect of undesirable corrosion (Merzouk *et al*., 2009). By adding high NaCl concentration (1 and 1.5 g/L), removal of fluoride reduces rapidly. The positive role could be offset by the negative role, and therefore, chloride had only little effect on the defluoridation (Zuo *et al*., 2008). It is therefore beneficial to choose a small amount of NaCl to be added for efficient removal and low consumption of electrodes, mainly anode.

According to a previous study (Takdastan *et al*., 2015), an examination of the chemical reactions occurring in the electrocoagulation process shows that the main reactions occurring at electrodes (aluminum and iron electrodes) are presented as follows:

At Anode: $M \longrightarrow M^{3+} + 3e^-$ (3) with M = Fe or Al

At Cathode: $3H_2O + 3e^- \longrightarrow \frac{3}{2}H_2 + 3OH^-$ (4)

If M = Al, Al^{3+} and OH^- ions generated at electrode surfaces react in the bulk water to form aluminum or iron hydroxide:

$M^{3+} + 3OH^- \longrightarrow M(OH)_3$ (5) with M = Fe or Al

In the study of removal mechanisms, the aluminum and iron hydroxide flocs normally act as adsorbents and therefore, they would remove fluoride from the solution through adsorption process (Bazrafshan *et al.*, 2012). In this work, the mechanism of fluoride removal could involve co-precipitation and adsorption occurred inside of EC reactor.

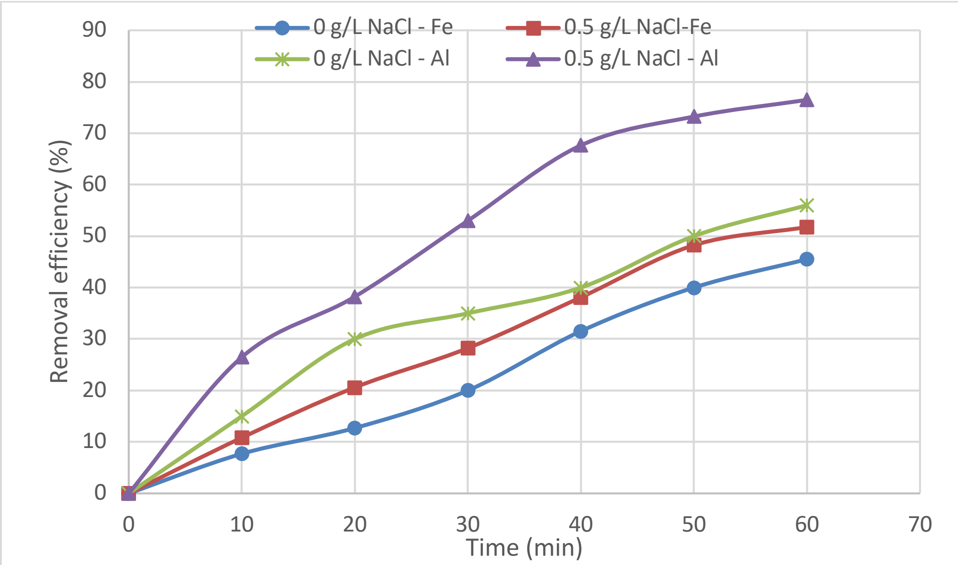

Figure 6. Effect of NaCl concentration on removal efficiency using Al and Fe electrodes with C_0 = 10 mg/L, I=10 mA; pH= 7.68; and V=350 mL.

3.2. Influence of operating parameters on energy consumption

3.2.1. Nature of electrodes

Theoretically, the energy consumption depends on electrolysis time: the increase in electrolysis time leads to energy consumption increase. The electrical energy consumption during fluoride removal process was affected by the type of electrodes (iron and aluminum). In any electrical process, the cost is incurred due to electrical energy demand, which affects the operating cost. For EC process, the operating cost includes materials and electrical energy costs, as well as lab maintenance, sludge dewatering and disposal and fixed costs. Generally, energy consumption was taken into account as major cost item in the calculations of the operating cost. Looking at **figure 7**, it was observed an increase of energy consumption with electrolysis time increasing using each electrode.

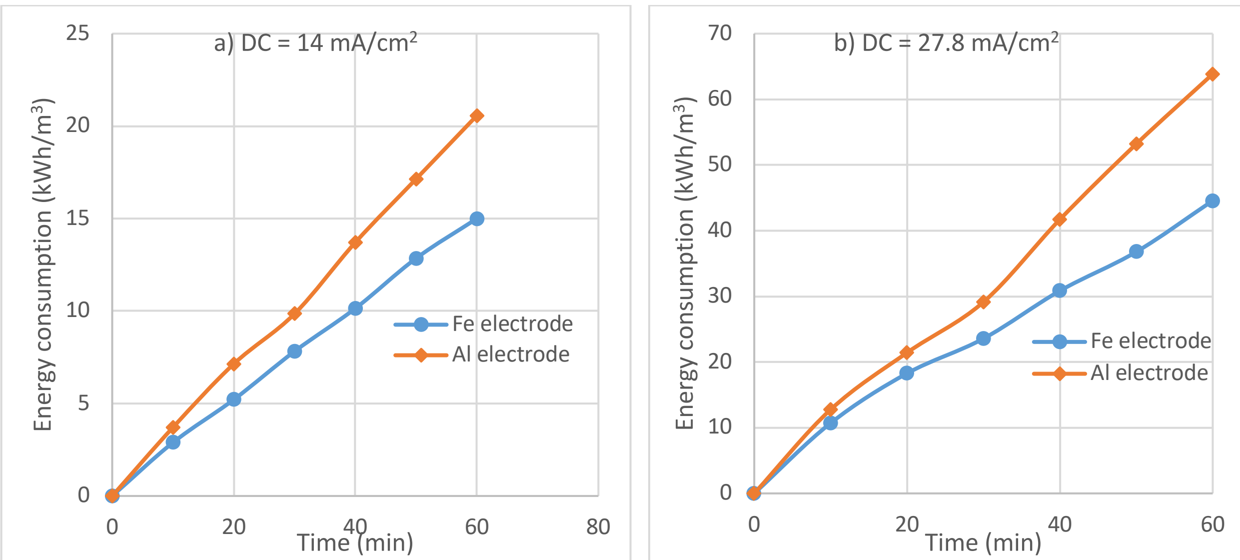

Figure 7. Effect of current intensity on energy consumption using Al and Fe electrodes with C_0 =5 mg/L; pH= 7.68; 0.5 g/L NaCl, and V=350 mL, (a) DC = 14 mA/cm^2 (b) DC = 27.8 mA/cm^2

When the current density increased, energetically efficiency was increased. However, we notice that Fe electrodes have less energy consumption and consequently, the type of electrode is more adapted for developing countries.

3.2.2. Current density

The current density is one of main parameters affecting the energy consumption in EC process because it is depending of intensity and voltage values. In this study, three values of DC (14, 18.5 and 27.8 mA/cm^2) were used in experiments with 350 mL of solution containing 5 mgF$^-$/L and 0.5 g/L of NaCl added. Results showed an increase of energy consumption when the DC increases (**Figure 8**). This increase of energy consumption confirms the voltage and intensity dependence. Iron electrodes have presented an energy consumption lower than Al electrodes due to their high density and this low energetically efficiency causes a low anode consumption during the EC process.

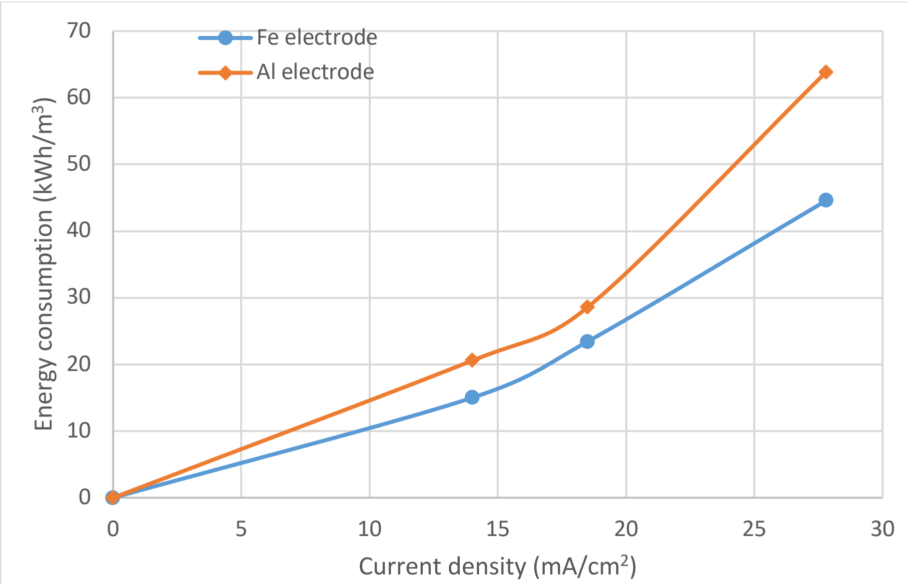

Figure 8. Influence of DC on energy consumption using Fe and Al electrodes.

3.2.3. Initial fluoride amount

To assess the effect of initial fluoride C_0, three values of C_0 were used under operating conditions such DC fixed to 27.8 mA/cm^2 and electrolysis time to 60 min. Experimental results summarized in **Table 1** indicated a decrease of energy consumption when the initial fluoride concentration increased. This result can be due to the decrease of fluoride removal efficiency with increase of initial fluoride. In addition, the increase of initial fluoride provides more fluoride ions which act on energetically efficiency.

Table 1. Value of energy consumption vs initial fluoride.

C_0 (mg/L)	*Energy consumption (kWh/m^3)*	
	Using Fe electrodes	**Using Al electrodes**
5	15.71	20.57
10	15.37	19
15	15	16.91

3.2.4. NaCl amount

When any NaCl was added to fluoride solution, it was observed that the current intensity did turn around 10 mA and only the voltage can change by increasing. This increase of voltage caused an increase of energy consumption whose the values were lower comparatively to fluoride solution including NaCl solution. Experimental results (Table 2) showed a linear increase of energy consumption with electrolysis time using Fe electrodes while this energy doesn't change considerably (≃1 kWh/m^3) using Al electrodes.

Table 2. Influence of 0.5 g/L NaCl adding on energy consumption during EC process.

Time (min)	*Energy consumption (kWh/m^3)*	
	Using Fe electrodes	**Using Al electrodes**
10	1.7	0.3
20	3.1	0.61
30	4.65	0.75
40	6.2	1.03
50	7.74	1.06
60	9.31	1.21

Conclusion

This study could investigate the application of electrocoagulation process using aluminum and iron electrodes in fluoride removal under various conditions such as electrode type, electrolysis time, current density, initial fluoride concentration and NaCl amount, requiring their optimization at lab scale. Results showed that iron and aluminum electrodes could successfully remove fluoride using EC process. Removal efficiency increased with increasing of DC and NaCl but the increase of initial fluoride has caused a decrease of EC efficiency. In addition, Al electrodes could allow to have a total removal of fluoride (100%) while Fe electrodes were less efficient (77.44%) using C_0 = 5 mg/L and DC = 27.8 mA/cm^2 during 60 min. Operating conditions have affected the energy evaluation and its consumption increased with reaction time and current density. Indeed, the increase of initial fluoride decreased the energy consumption using each electrode. Aluminum electrodes required more energy demand comparatively to iron electrodes. Consequently, a treatment plant can be only set up with aluminum electrodes using solar energy in developing countries such Togo and Burkina Faso where energy supply is one of most challenges that governmental authorities are facing.

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