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An experimental study for adapting electrocoagulation as a technique for fluoride removal from water

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Abstract. A small amount of fluoride in potable water can maximize teeth mineralization and minimizes their cavities. However, fluoride presence in excess quantities in drinking water could cause severe illnesses including Alzheimer's syndrome and women infertility. Thus, scholars are concerned over the last decades in developing techniques to minimize the concentration of fluoride from wastewater and lower its impact on human health. The current research aims to utilize the electrocoagulation technique to remove the fluoride from wastewater. Experiments were conducted to study fluoride removal using rectangular electrocoagulation cell and analyze the influence of four main parameters namely detention duration, electrodes spacing, pH and current density on the performance of the electrocoagulation techniques on fluoride removal. The results showed that 93% of the fluoride was removed from the wastewater after 20 minutes of treatment using 5mm electrodes spacing with a current density of 2 mA/cm² at a pH level of 7. The main parameters have a significant influence on the removal efficiency of the fluoride. A higher fluoride removal efficiency can be achieved in the acidic setting. The removal efficiency is positively related to the electrical current and the detention time while it is negatively related to the electrode spacing.

1. Introduction

Water is an essential component of the existence of life on the planet earth. However, only 1% or less is fresh which is suitable for drinking purposes [1-3]. The provision of adequate quantities of potable water is considered one of the main challenges currently facing human civilization due to population growth [4-6]. Researchers, [7-9], reported that 50% of the earth's population will have no access to freshwater sources by the middle of this century. Other scholars highlighted that millions of cubic meters of polluted water are yearly discharged into the water bodies (surface or groundwater bodies) due to industrial growth making the problem is even more challenging, for example, more than 33.6 million barrels of heavily polluted water is disposed to water bodies to produce only 84 million barrels of oil [10-12].

Fluoride is one of the most abundant elements where about 5% of the earth's crust contains fluoride in its composition [13, 14]. Therefore, most water bodies naturally contain a low concentration of fluoride in the water. Industrial development has led to increasing the fluoride concentration in water bodies; for example, the production of the semiconductor such as steel, and aluminum using high-temperature chemicals deposit large quantities of fluoride contaminated wastewater. The presence of fluoride in low concentration in freshwater helps to avoid tooth cavitation; however, high concentration could lead



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to serious diseases such as Alzheimer's syndrome and paralysis of the skeleton. Thus, the World Health Organization has limited the fluoride concentration in drinking water to 1.2 mg/l [15]. Therefore, researchers currently employing different treatment techniques to remove contaminants from water or wastewater [16, 17]. The electrocoagulation techniques are promising treatment techniques for water and wastewater contamination as it supports three conventional treatment processes that are flotation, coagulation, and electrochemistry [18, 19], it costs less operation cost [20], it gives a good flexibility to combine it with other methods [21-23], it generates few sludge [24, 25], its sludge is rich with metal ions that enhances the possibility of recycling it as cement alternative [26-32] rather than dispose it in costly landfills [33-35], and finally it could be operated using sensors, such as microwave sensors [36-40].

Scholars, [41-44], used electrocoagulation process to remove a variety of pollutants including heavy metals and biological pollutants from the water and the wastewater. For instance, electrocoagulation technique has been used to remove arsenic from wastewater using Fe-Al electrodes where about 100% of the Arsenic were removed after 30 minutes of treatment [45-47]. Other metals including nickel and copper were removed from wastewater using electrocoagulation after 20 minutes of treatment using Aluminum electrodes at the acidic environment (pH =3). Electrocoagulation was also implemented to remove organic pollutants from industrial wastewater, and it was able to remove organic pollutants from tannery wastewater with removal efficiency of about 85% after 45 minutes [44, 47].

Electrocoagulation is also applied to remove fluoride from wastewater. For instance, the concentration of the fluoride was minimized in synthetic wastewater samples using aluminum electrodes. The impact of several parameters including current density, and the pH on the fluoride removal using electrocoagulation process were explored by many scholars and it was found that higher current density to higher fluoride removal efficiency. Electrocoagulation was also proved effective in removing fluoride from potable water. Hashim *et al.*, [15] used this technique to remove the fluoride of the initial concentration of 2 to 10 mg/l from synthetic potable water using monopolar and bipolar electrodes. It was confirmed that bipolar connection has a higher removal efficiency than monopolar connection.

The problem of water pollution threatens humanity for decades and safe water treatment methods are required to meet the future demand for potable water. Currently, the researcher's attention is the use of safe techniques to remove pollutants from water and wastewater. According to researchers, electrocoagulation provides robust treatment techniques comparing to other traditional water and wastewater treatment techniques due to its low cost, small unite size, low sludge quantities as there is no need to add coagulants. Thus, the current research aims to employ an electrocoagulation technique to remove fluoride from wastewater using a perforated plate of the Aluminum electrode which water mixing and aerating processes.

2. Methodology

2.1. Materials and chemicals

Sigma-Aldrich provided all chemicals used in this project. All materials were implemented in the experiments without adjustment or decontamination. According to Hashim *et al.*, [48], the Synthetic solution was prepared by dissolving a specific amount of sodium fluoride (NaF) to deionized water to reach a concentration of 500 mg/l of fluoride in this stock solution. From this solution, samples of 1 liter in size of lower fluoride concentrations were prepared by dilution. Additionally, HCl and NaOH were used to control the pH value in the solution to be in the range between 4 and 9 while NaCl was used to maintain the conductivity of polluted water at 0.3 mS/cm.

2.2. Electrocoagulation unit

The literature provides a comprehensive review of the electrocoagulation process showing that the rectangular unite still dominated [15]. Accordingly, a rectangular reactor of more than 1 liter in size was employed in this research with inner dimensions of 5, 20, and 10 cm for width, length, and height, respectively (Figure 1). The exterior of the reactor is Perspex which allows the examiner to check the mixing performance of the reactor. The interior of the reactor contains 4 Aluminum perforated plates of which were positioned vertically in the reactor as shown in Figure 1. The Aluminum plates used in the

electrocoagulation unit was provided by Liverpool John Moores University. The high affinity of the Aluminum to fluoride ions makes it an effective electrode to remove the fluoride from the water.

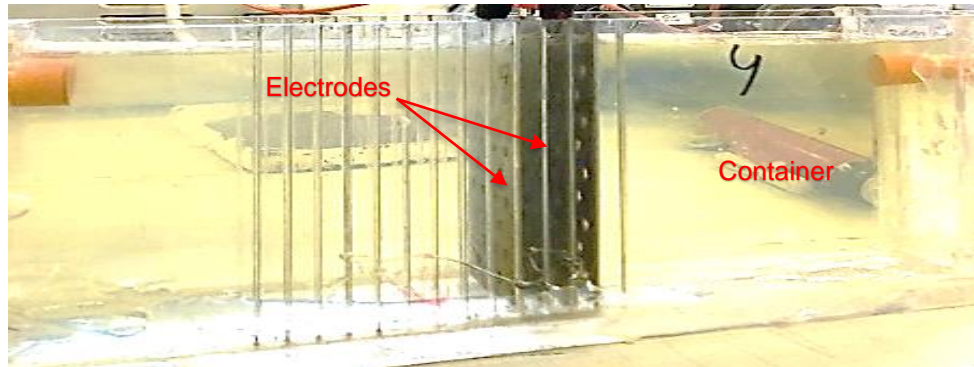


Figure 1. The electrocoagulation reactor used in this study.

2.3. Experimental procedures

500 ml of contaminated water was placed in the reactor. To complete the electrolyzing process, the inlet and the outlet of the reactor were connected and the water was circulated using a peristaltic pump. The power supply was connected to the four perforated Aluminum plated to maintain a stable electrical current. Besides, a pH meter, conductivity meter, and thermometer were used to measure the pH, the solution conductivity, and the solution temperature. All trials in this research were conducted at room temperature of $20 \pm 1^\circ\text{C}$. The efficiency of the reactor to remove the fluoride from the water was calculated following equation [15]:

$$\text{fluoride removal efficiency (\%)} = \frac{y_f - y_i}{y_i} * 100\% \quad (1)$$

where y_f is the final fluoride concentration in the solution while y_i is the starting fluoride concentration in mg/l.

2.4. The influence of the experiment parameters

The influence of 4 parameters namely electrical current density, pH level, the spacing between plates and treatment duration on the removal efficiency of the fluoride ions from the water was investigated in the research [7]. First, the influence of the pH level was studied by maintaining 2 mA/cm² electrical current, 5 mm spacing between electrodes, and initial fluoride concentration of 10 mg/l for 20 minutes treatment duration while the pH of the solution changed from alkaline (9), neutral (7), and acidic (4). Second, the electrical current influence was studied using three values that are 1, 2 and 4 mA/cm² while maintaining the pH a 7, the spacing at 5mm, the initial fluoride concentration at 10mg/l. Third, the treatment duration impact was analyzed by changing the treatment duration from 0 to 35 minutes while maintaining the remaining variables to achieve ideal fluoride removal. finally, the impact of plate spacing was analyzed by changing the plated spacing from 5 to 15 and maintaining the remaining variables.

3. Results and discussions

3.1. Solution pH level influence

Khalid *et al.*, [7] stated that the pH level plays a vital role in the electrocoagulation process for the treatment of any pollutant. The influence of pH level on the removal efficiency of fluoride has been investigated by treating 500 ml water using the predefined reactor for 20 minutes at pH level values of 4, 7, and 9 using 5 mm spacing and 2 mA/cm² electrical current. Figure 2 provides a graphic presentation

on the impact of pH on the removal efficiency of fluoride at different pH levels. The removal efficiency of both pollutants increased with the decrease in the wastewater acidity. The best removal efficiency was reached in acidic levels (pH = 4). When the wastewater becomes alkaline (pH = 9), the removal efficiency significantly decreases with the increase in water pH level. The variation in the elimination efficiency of the fluoride indicates an acidic environment is desirable when fluoride is removed using electrocoagulation. Researchers highlighted that the acidic environment (pH is between 4 and 6) is preferred to achieve higher fluoride removal efficiency.

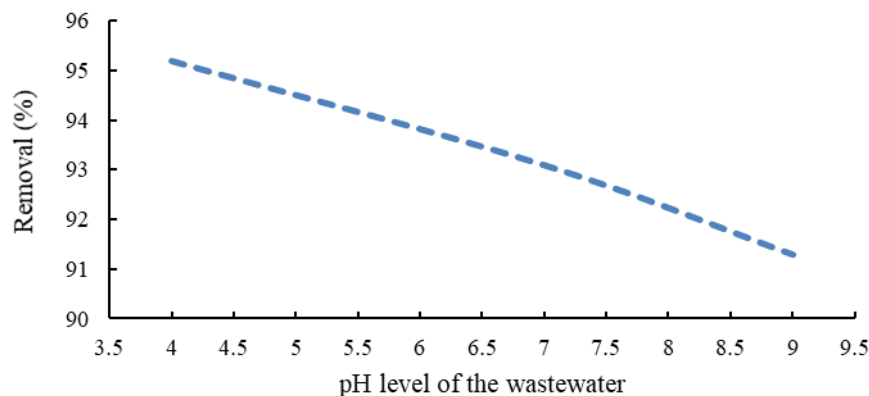


Figure 2. The influence of solution pH on the fluoride removal efficiency.

3.2. *The influence of electrical current*

The electrical current strangely influences the removal of fluoride as it regulates bubbles size and floc production. Thus, the influence of the electrical current on the efficiency of fluoride removal was studied in this investigation using three different currents ranged from 1 to 4 mA/cm² while maintaining the pH level at 7, the initial fluoride concentration at 10 mg/l and the spacing of the plates at 5 mm. The treatment duration was also varied between 5 to 20 minutes. As illustrated in figure 3, the removal of fluoride increased with higher electrical current. For instance, after 10 minutes of treatment, the removal efficiency was about 78%, 83% and 86.5% for the electrical current of 1, 2 and 4 mA/cm², respectively. Besides, detention time also has a positive impact on the removal efficiency of fluoride from water; for 1 mA/cm² current, the efficiency was about 89%, 84%, 77%, and 65% at detention time of 20, 15, 10 and 5 minutes, respectively. It can be said that the removal efficiency using a 4 mA/cm² electrical current is 1.5 the removal efficiency using 2 mA/cm² electrical current. This confirms other scholars statement that supplying higher current directly speeds up the reaction that results in Al species in charge of the coagulation and speeds up floc creation to enhance the removal efficiency [12, 25].

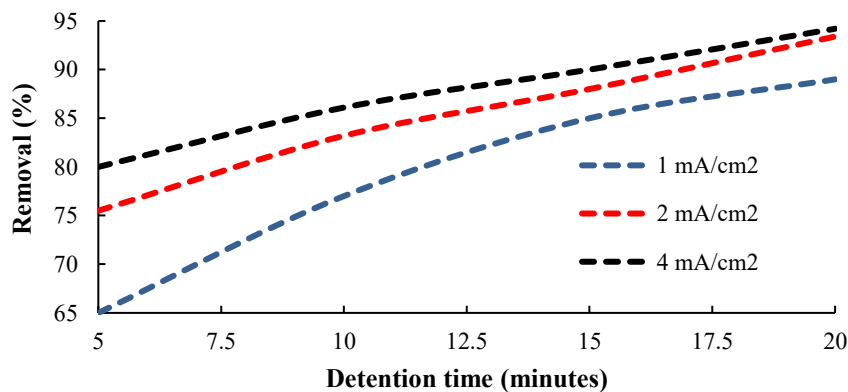


Figure 3. The influence of electrical current on the fluoride removal efficiency.

3.3. Impact of electrodes spacing

Researchers, [49, 50], highlighted that the removal performance of the electrocoagulation unit is highly influenced by the spacing of the electrodes. Thus, the influence of electrodes spacing on the removal performance of the electrocoagulation unit were analyzed by treating 500 ml water with an initial fluoride concentration of 10 mg/l, a pH level of 7, and an electrical current of 2 mA/cm² while changing spacing distances; 5 to 15 mm. Besides, the detention time also changed from 5 to 20 minutes. The outcome showed in figure 4 highlighted that electrodes spacing negatively influencing the removal efficiency of the fluoride from the water while the detention time still positively impact on the removal efficiency of the pollutant. For instance, after 20 minutes of treatment, the removal increased from 74% to 93% as the spacing between electrodes decreased from 15 mm to 5 mm. Researchers attributed the change in the fluoride removal efficiency to the electrical resistance between anodes and cathodes. At a relatively remote distance, electrical resistance increases which enlarges the passive layer around the cathodes and minimizes the overall removal efficiency of the fluoride in the electrocoagulation method [49, 50].

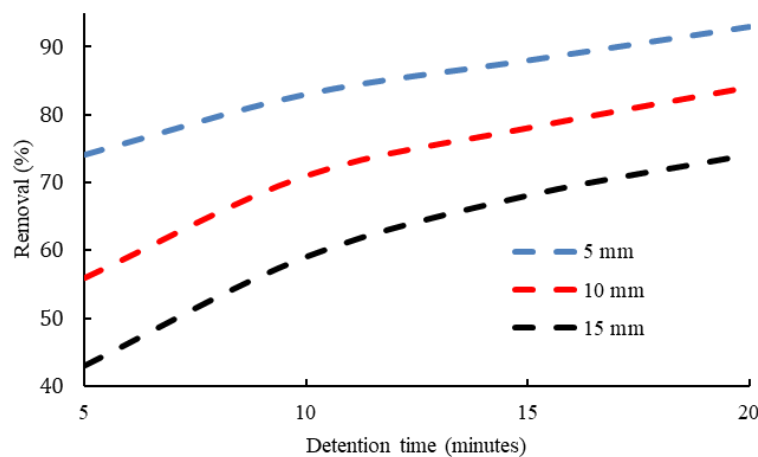


Figure 4. The influence of the spacing on the fluoride removal efficiency.

3.4. The influence of the detention time

Detention time significantly influences the overall removal efficiency of the fluoride from wastewater using electrocoagulation as it affects the generation of Aluminum ions [48]. The influence of the detention time on fluoride removal was investigated in this study by treating 500 ml of 7 pH level using the electrical current of 2 mA/cm² between electrodes of 5mm spacing at several times which were 5, 10, 15, 20, 25, 30, and 35. It was deduced from the outcome that the fluoride removal efficiency increases with the increase of detention time (Figure 5). The removal efficiency achieved at 5 minutes was 75% due to the availability of free fluoride in the solution which can be easily coagulated and removed. The increase in fluoride removal efficiency increased nonlinearly after 5 minutes of detention time to about 97% after 25 detention time. This agrees with the results of many researchers whom stated that the treatment progress using the electrocoagulation technique is proportion to the detention time [46, 47].

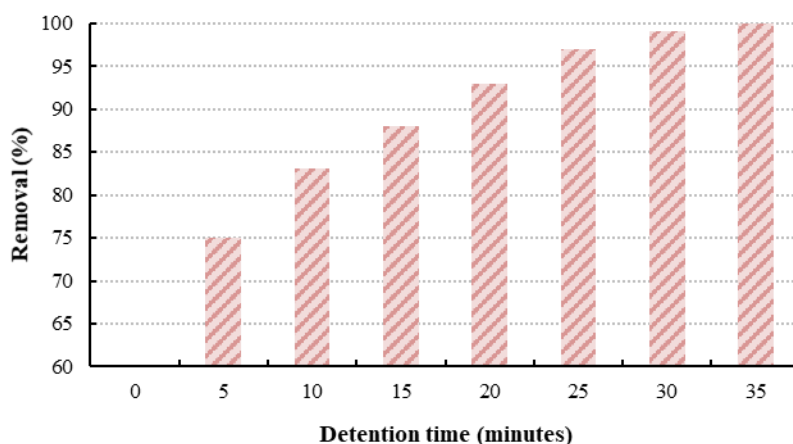


Figure 5. The influence of detention time on fluoride removal efficiency.

For future studies, basing on the literature that confirmed that the climate change has seriously increased water demands [51, 52], water pollution [53, 54] and nature of the cities [55-58], researchers should investigate the relationship between fluoride concentrations in surface water and climate changes.

4. Conclusion

The current study investigated the effectiveness of rectangular electrocoagulation cells with perforated Aluminum electrodes and the impact of main operating components namely pH level, electrical current, the spacing between electrodes, and detention time duration on the removal of the fluoride from water and wastewater. According to the outcome of this experimental study, it can be said that this cell can be used to remove the fluoride from wastewater. Besides, the main operating components are highly influencing the removal efficiency of the fluoride. The pH level plays important role in the removal of the fluoride from the wastewater as a higher removal efficiency can be achieved in the acidic setting (pH level is lower than 5). The removal efficiency is positively related to the electrical current and the detention time as it enhances the generation of the coagulates and increases the contact time. On the other hand, the electrode spacing has a negative impact on the removal efficiency of the fluoride from the wastewater. The removal efficiency of 93% was achieved after 20 minutes of treatment times using 2 mA/cm² electrical current and 5 mm electrodes gap at neutral pH level.

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