Journal of Mines, Metals and Fuels, *73(8): 2539-2545; 2025. DOI: 10.18311/jmmf/2025/49307*

Print ISSN : 0022-2755

Journal of Mines, Metals and Fuels

**Contents available at:** *www.informaticsjournals.co.in/index.php/jmmf*

# Optimisation of Fluoride Removal in Synthetic Water Using Electrocoagulation Techniques

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## Abstract

*The presence of excess fluoride in water bodies presents significant environmental concerns due to its harmful effects on human health. Medical studies indicate that fluoride concentrations exceeding 1.5 mg/L can result in various health issues, including skeletal fluorosis, dental fluorosis, and several other health complications. The electrocoagulation (EC) method offers a practical solution for removing various contaminants from water and wastewater. This study focuses on reducing the fluoride concentration in an aqueous solution using electrochemical (EC) methods, starting with an initial concentration of 10 mg/L, and explores different electrode combinations, including aluminium-aluminium, aluminium-copper, aluminium-lead, and aluminium-zinc. Further research was conducted by adjusting the electrode spacing to determine the optimal fluoride removal efficiency in the electrolytic cell. The findings reveal that with 2 cm electrode spacing, the aluminium-copper combination achieved a fluoride removal efficiency of 73% after 120 minutes. Meanwhile, a 4 cm electrode spacing with the aluminium-copper combination also resulted in an 80% removal efficiency at a reaction period of 120 minutes. Thus, the study shows that the combination of aluminium–copper electrodes gives the best results concerning fluoride removal from aqueous solutions at higher concentrations.*

**Keywords:** *Current Density, Electrocoagulation, Electrode, Fluoride, Removal Efficiency, pH*

## 1.0 Introduction

Fluorides are significant inorganic contaminants found in drinking water, primarily originating from the Earth's crust[1]. The increasing use of groundwater for domestic, industrial, and agricultural purposes due to global population growth has led to the depletion of groundwater reserves, impaired water quality, and higher extraction costs[2]. The presence of fluoride in water is a growing health concern when consumed in either very low or excessively high concentrations. The World Health Organization (WHO) recommends a fluoride concentration of 1.5 mg/L for drinking water[3]. Fluoride at a concentration of around 1 mg/L can be beneficial for preventing dental decay[4]. However, when fluoride concentrations exceed 4 mg/L, they can cause serious health issues, including dental and skeletal fluorosis, osteoporosis, kidney, liver, lung, and muscle problems, neural damage, brain injuries in children, and even structural DNA alterations. On the other hand, low fluoride concentrations can promote osteoblast *activity*[5].

Various methods, such as chemical coagulation, have been employed for removing fluoride and other contaminants like arsenic from water. These techniques include reduction, ion exchange, electrodialysis, electrochemical precipitation, evaporation, solvent extraction, reverse osmosis, chemical precipitation, and absorption[6]. Electrocoagulation (EC), a promising electrochemical technique, has been found effective for fluoride removal with moderate energy consumption.

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The EC process involves generating trivalent metal species from sacrificial electrodes, such as aluminium or iron, along with water electrolysis at the cathode to produce hydrogen and hydroxide ions. These ions then interact with the fluoride, leading to its removal. With aluminium electrodes, the trivalent aluminium species predominantly form solid $Al(OH)_3$ between pH 5 and 8, which polymerizes into large solid Al oxo-hydroxy networks (flocs) with various complex hydration states and molecular structures[7]. During EC, several processes occur, including electrolytic reactions on the electrode surfaces, coagulant formation in the water, and adsorption of pollutants facilitated by the coagulant, which are then separated through flotation or sedimentation aided by small hydrogen bubbles generated at the cathode[8]. EC involves electrolytic oxidation on the anode, and the reactor consists of an electrolytic cell with an anode and cathode[9]. In EC, secondary reactions occur, including hydrogen ($H_2$) and oxygen ($O_2$) gas production.

At the cathode and cathode:

$$nH_2 + ne^- \rightarrow (n/2)\, H_{2\,(g)} + nOH^-_{(aq)} \quad (1)$$

At anode:

$$2H_2O \rightarrow 4H^+_{(aq)} + O_{2(g)} + 4e^- \quad (2)$$

$$M_{(s)} \rightarrow M^{\,n+}_{(aq)} + ne^- \quad (3)$$

The $H_2$ gas assists in lifting flocs to the solution surface, a process known as electro-floatation, which enables their removal through skimming. Oxygen generated at the anode forms hydrogen peroxide, an intermediate that aids in the oxidation of toxic and non-toxic species. Heavier flocs tend to sink and form sludge, which can be efficiently removed by various methods[10]. The adaptability, efficiency, affordability, and environmental friendliness of this electrochemical technology have made it a viable option[11]. The potential for metal electrode corrosion and other pollutants to leak into the water, the requirement for frequent electrode replacement and maintenance, and the possibility of excessive energy consumption, if the procedure is not optimized, are some of the drawbacks of electrocoagulation[12].

## 2.0 Materials and Methods

An electrochemical reactor, made from Plexiglas, was designed with dimensions of 22.5 cm in height, 2.5 cm in length, and 6.5 cm in width. For the EC process, electrodes of aluminium, lead, zinc, and copper with 12 mm circular cross-sections were used. The reactor setup is depicted in Figure 1. In the reactor, a 1300 mL aqueous sodium solution was prepared, and the aluminium, zinc, lead, and copper electrodes were employed as cathodes and anodes, with inter-electrode distances set at 2 cm and 4 cm, respectively, mounted on a supporting plate. A constant current was supplied using a DC power source (60V). The fluoride concentrations before and after treatment were analyzed using a UV-VIS spectrophotometer, and the results were documented in the data from Tables 1 to 4.

## 3.0 Influence of pH on Electrocoagulation

The pH of the water samples tested was in the range of 9.1 to 9.6 (alkaline side). Hence to bring down this pH value to neutral or ideal pH requirement for satisfactory functioning of the process (7.2 to 7.8) concentrated sulphuric acid was added at precise dosage.

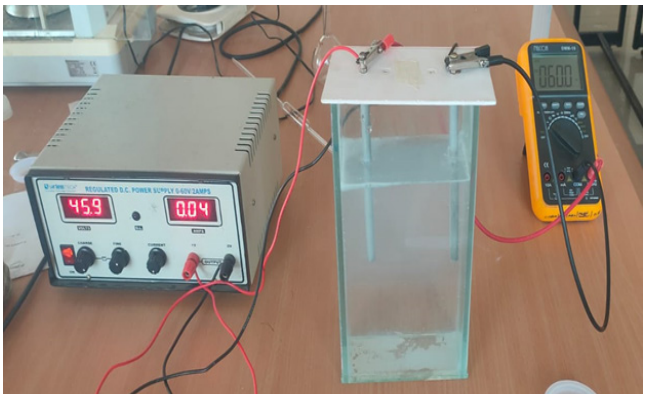



**Figure 1.** Schematic representation of an EC cell with pollutant fluoride removal mechanism.

## 4.0 Results and Discussion

The electrocoagulation process is quite complex and may be affected by several operating parameters,

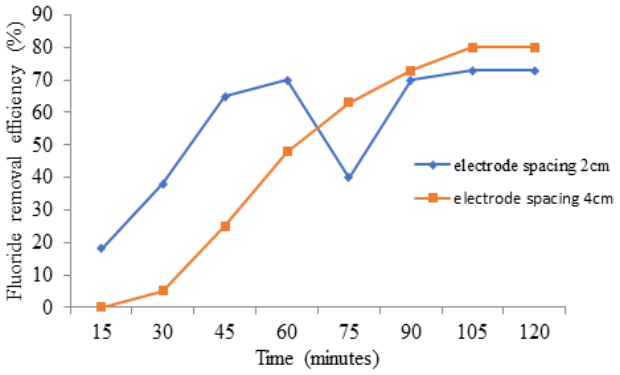



**Figure 2.** Fluoride removal efficiency using aluminium-copper Electrodes.

such as pollutant concentrations, initial pH, electrical potential (voltage), and type of electrode[13]. The primary objectives of the present were to determine the effects of voltage, pH, initial concentration, and reaction time on the removal efficiency of fluoride from the aqueous environment using an electrocoagulation process with electrodes made of iron, copper, zinc and aluminium electrodes. Different combinations of electrodes (aluminium – copper, aluminium – zinc, aluminium-lead, aluminium- aluminium) were used in the study to evaluate the efficiency of fluoride removal in 10 mg/L initial concentration of fluoride by varying the combinations of electrode material and electrode with a spacing of 2 cm and 4 cm respectively. During the working cycle, the electrodes function without any drop in their performance efficiency for their complete experimental duration. However, the useful life of these electrodes is around a week in the laboratory study and its shelf life is around 3 months in the industrial sector and as per pilot scale study.

Figure 2 shows the Fluoride removal efficiency using aluminium-copper Electrodes. From the analysis, it is found that the maximum fluoride removal efficiency obtained was 73% and 80% at 120 minutes from the time of electrode reaction using aluminium-copper electrodes with an electrode spacing of 2cm and 4cm. The initial fluoride removal was 8% at 15min duration with an electrode spacing of 2cm and 0% fluoride removal at 15min duration with an electrode spacing of 4cm and the percentage removal of fluoride gradually increased as the reaction period increased, there is a drop in the fluoride reduction at 75 minutes and in 60 min with a spacing of 2 cm and 4 cm, this is due to low or high pH, formation of flocs may reduce which hinder the removal efficiency. The same is presented in Table 1.

Using an aluminium-lead electrode, the fluoride removal efficiency was 65 % at 60 minutes with an electrode of 2 cm and 70 % with 4 cm electrode spacing at 90 minutes with an initial concentration of 10 mg/L which is shown in Figure 3. The fluoride removal in

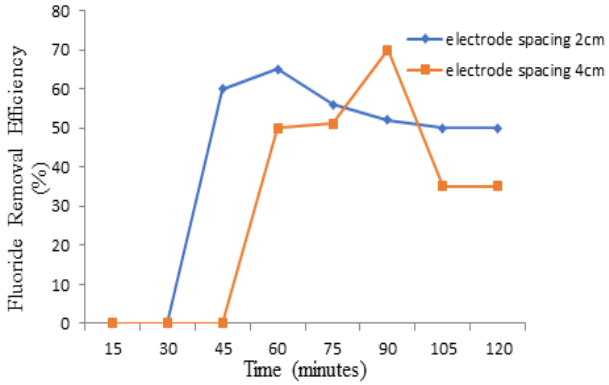



**Figure 3.** Fluoride removal efficiency using aluminium-lead electrodes.

**Table 1.** Fluoride removal efficiency using aluminium-copper electrodes

| Time (min) | 15 | 30 | 45 | 60 | 75 | 90 | 105 | 120 |
|---|---|---|---|---|---|---|---|---|
| Efficiency of fluoride removal (%) with 2cm spacing | 18 | 38 | 65 | 70 | 40 | 70 | 73 | 73 |
| Efficiency of fluoride removal (%) with 4cm spacing | 0 | 5 | 25 | 48 | 63 | 73 | 80 | 80 |

**Table 2.** Fluoride removal efficiency using aluminium-lead electrodes

| Time (min) | 15 | 30 | 45 | 60 | 75 | 90 | 105 | 120 |
|---|---|---|---|---|---|---|---|---|
| Efficiency of fluoride removal (%) with 2cm spacing | 0 | 0 | 60 | 65 | 56 | 52 | 50 | 50 |
| Efficiency of fluoride removal (%) with 4cm spacing | 0 | 0 | 0 | 50 | 51 | 70 | 35 | 35 |

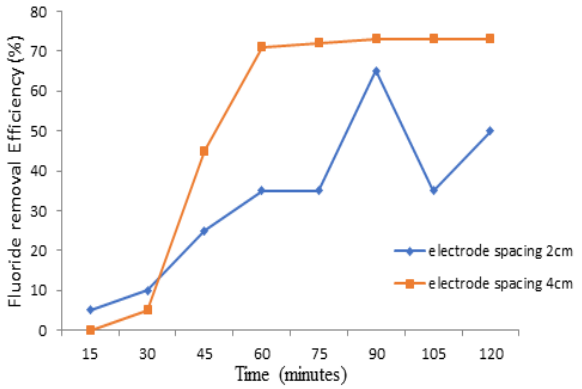



**Figure 4.** Fluoride removal efficiency using aluminium-zinc electrodes.

the initial stage is nil i.e., 0-30 minutes in both the electrode spacing. There is a gradual decrease in the reduction of fluoride in the reactor after 60 minutes in the electrode spacing of 2cm, this is because of cell reaction. Table 2 represents the fluoride efficiency removal with an electrode combination of aluminium-zinc combination.

Figure 4 shows the combination of aluminium-zinc electrodes where initially the fluoride reduction is slow as the removal rate decreases with increasing Inter-Electrode Distance (IED). The fluoride removal using aluminium-zinc combination was achieved at 90 minutes for both the electrode spacing with maximum efficiency

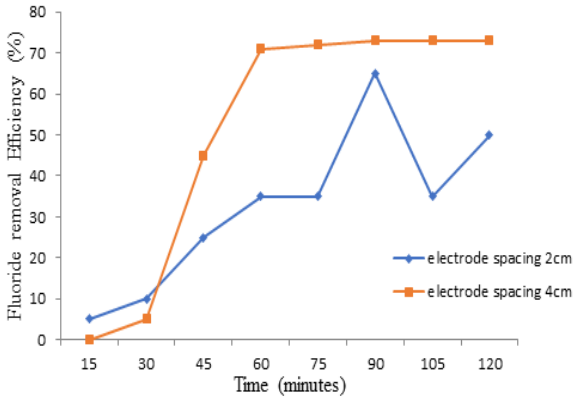



**Figure 5.** Fluoride removal efficiency using aluminium-aluminium electrodes.

**Table 3.** Fluoride removal efficiency using aluminium-zinc electrodes

| Time (min) | 15 | 30 | 45 | 60 | 75 | 90 | 105 | 120 |
|---|---|---|---|---|---|---|---|---|
| Efficiency of fluoride removal (%) with 2cm spacing | 5 | 10 | 25 | 35 | 35 | 65 | 35 | 50 |
| Efficiency of fluoride removal (%) with 4cm spacing | 0 | 5 | 45 | 71 | 72 | 73 | 73 | 73 |

**Table 4.** Fluoride removal efficiency using aluminium- aluminium electrodes

| Time (min) | 15 | 30 | 45 | 60 | 75 | 90 | 105 | 120 |
|---|---|---|---|---|---|---|---|---|
| Efficiency of fluoride removal (%) with 2cm spacing | 5 | 10 | 25 | 35 | 35 | 65 | 35 | 50 |
| Efficiency of fluoride removal (%) with 4cm spacing | 0 | 5 | 45 | 71 | 72 | 73 | 75 | 73 |

of 65% and 73% with the initial concentration of 10 mg/L. Table 3 shows the efficiency of fluoride removal using aluminium–zinc electrodes.

Figure 5 shows the fluoride removal efficiency using an aluminium-aluminium electrode combination, the electrolytic action in the cell started in the initial stages itself compared to other electrode combinations. The electrolysis with Al electrodes acts as pH neutralization of the electrolytic medium taking into account. The advantage of electrode spacing observed during the electrolysis process and thus removing maximum fluoride in synthetic water at 90 minutes with an electrode spacing of 2cm and 75% efficiency obtained with 4cm spacing at 105 minutes and the same is represented in Table 4. In neutral electrolyte solutions, aluminium tends to passivate, yielding very low dissolving current densities over a wide range of potentials. It was shown that as the electrode potentials become more positive, the thickness of the passive layer increases linearly[14].

## 5.0 Effects of Operating Conditions on Fluoride Removal Efficiency

### 5.1 Influence of Electrode Type

In this study, copper, lead, zinc and aluminium electrodes were used as anode and cathode during the electrolysis time and their performance in fluoride removal was assessed during the whole experiment. Table 5 shows the experimental results of maximum fluoride removal with different electrode combinations. The aluminium copper electrodes give a maximum efficiency of 80% with an electrode spacing of 4cm at 120 minutes from the initial electrode reaction in a DC supply of 60V. The aluminium-lead electrode combination achieves 60% fluoride at 120 minutes of the reaction period removal which is the least fluoride removal compared to other electrode combinations and this may be due to the electrolytic reaction in the cell.

**Table 5.** Fluoride removal efficiency with different electrode combinations

| Electrode Type | 2 cm Electrode Spacing | | 4 cm Electrode Spacing | |
|---|---|---|---|---|
| | Efficiency (%) | Time (minutes) | Efficiency (%) | Time (minutes) |
| Aluminium-Copper | 73 | 120 | 80 | 120 |
| Aluminium-Lead | 65 | 60 | 70 | 90 |
| Aluminium-Zinc | 65 | 90 | 73 | 120 |
| Aluminium- Aluminium | 65 | 90 | 73 | 120 |

### 5.2 Effect of Current Density

Current densities were studied for a concentration of 10 mg/l of fluoride; the current density was 60A/$m^2$ using bipolar electrode connections. Research indicates that fluoride elimination increased in tandem with an increase in current density. This is because anodic oxidation happens more quickly at higher current densities than at lower ones. This helps to form enough amorphous aluminium hydroxides both in the bulk and close to the electrode, which leads to the formation of a gelatinous layer over the electrode[15].

### 5.3 Influence of Electrode Spacing

The obtained results in the aluminium-aluminium, aluminium-zinc, aluminium-lead- lead and aluminium-copper electrodes with an electrode spacing of 2cm, show less percentage removal of fluoride[16]. This is due to the shortening of the anode-cathode distance is directly influenced by the gap between the electrodes. Short distances between the electrodes can, however, result in reduced removal efficiency of the pollutants from water due to the possibility of numerous phenomena (e.g., coagulation, flocculation, precipitation, electro-flotation, etc.) being affected. Because the high electrostatic effect prevents particles from colliding, these effects have an impact on the production of flocs and their precipitation, preventing the formation of aggregates. On the other hand, a large distance between electrodes greatly reduces the likelihood of floc formation[17].

### 5.4 Influence of pH

The pH in the study did not affect the electrolysis process; a pH of 7.2 to 7.8 was maintained throughout the study. One of the key factors influencing the removal of fluoride in electrocoagulation, as well as the execution of other electrochemical processes and chemical coagulation operations, is the pH of a solution. However, because of the defluorination reaction, it is exceedingly difficult to control the pH of the solution during the electrocoagulation process. Furthermore, it is not particularly helpful to alter the electrocoagulation system's initial pH to change the final pH[18]. Due to aluminium hydroxide's buffering ability, the electrocoagulation process's final pH and residual fluoride concentration did not significantly alter within the initial pH range of 4–8.

## 6.0 Conclusion

The electrocoagulation method effectively removed fluoride from synthetic water with high fluoride concentrations. The study's results indicate that the electrolysis efficiency is primarily determined by the quantity of coagulant ($Al(OH)_3$) produced in the water, which is influenced by factors such as electrolysis duration, applied voltage, and electrode spacing. A shorter electrode distance hindered ion diffusion by limiting the free flow of the electrolyte. The highest fluoride removal efficiency of 75% was achieved using an aluminium-aluminium electrode combination, with a maximum treatment time of 105 minutes. Additionally, an electrode spacing of 2cm resulted in a fluoride removal efficiency of 73% with the aluminium-copper combination over 120 minutes, while a 4 cm electrode spacing with an aluminium-aluminium combination reached 76% removal efficiency over the same duration. Overall, the study highlights that aluminium electrodes are the most effective for maximizing fluoride removal efficiency.

## 7.0 Acknowledgement

The authors are extremely thankful to Dr. M.G.R Educational and Research Institute, Chennai for providing the laboratory and library facilities during this research work.

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