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Development of an empirical model for fluoride removal from photovoltaic wastewater by electrocoagulation process

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ABSTRACT

Electrocoagulation experiments were conducted with bipolar aluminium electrodes to determine the optimum conditions for the fluoride removal from synthetic photovoltaic wastewater. A high fluoride concentration in community water supplies can cause fluorosis which has a detrimental effect on human health in particular on teeth and bones. A full 2^3 factorial design of experiments was used to obtain the best conditions of fluoride removal from water solutions. The three factors considered were initial fluoride concentration, applied potential, and supporting electrolyte dosage. Two levels for each factor were used; supporting electrolyte (0 and 100), applied potential (10 and 30 V), and initial fluoride concentration (20 and 25 mg/L). Results showed that the optimum conditions for fluoride removal from photovoltaic wastewater containing an initial fluoride concentration of 20 mg/L were a supporting electrolyte dose of 100 mg/L and an applied potential of 30 V. These gave a residual fluoride concentration of 8.6 mg/L which was below the standard discharge limit. A mathematical equation showing the relation between residual fluoride concentration and the effective variables was also developed.

Keywords: Photovoltaic wastewater; Electrocoagulation; Fluoride; Experimental design; Empirical model

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1. Introduction

The total number of people adversely affected by high fluoride concentrations in drinking water is estimated in the tens of millions [1,2]. In 1993, for example, 15 of India's 32 states were identified as suffering from fluorosis (i.e. a pathological condition resulting from an excessive intake of fluorine, usually from drinking water). A study by UNICEF shows that fluorosis is widespread in at least 27 countries across the globe [1,2]. Recently, the World Health Organization (WHO) has set an acceptable maximum limit for fluoride in drinking water to be between 0.5 and 1 mg/L [3]. However, the natural concentration of fluoride in groundwater depends on the geological, chemical and physical characteristics of, for example, an aquifer, the porosity and acidity of the soil and rocks, the temperature, the action of other chemical elements, and the depth of wells. Due to these variables, the fluoride

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concentration in groundwater can range from less than 1 mg/L to more than 35 mg/L. In India and Kenya, for example, concentrations up to 38.5 and 25 mg/L have been reported, respectively [1].

Fluoride also can be found in industrial wastewater from, for example, the glass manufacturing and the semiconductor industries [4,5]. The discharge of these wastewaters without treatment into the natural environment contributes to groundwater contamination. Due to its high toxicity; industrial wastewater containing fluoride is strictly regulated. In Algeria, the standard limit for wastewater containing fluoride is 15 mg/L [6], which is well above the WHO recommended maximum of 1 mg/L. In an attempt to alleviate this problem, a number of defluoridation processes have been developed, such as adsorption [7], chemical precipitation [8], electrodialysis [9], and electrochemical [10]. In the precipitation technique, alum or a combination of alum and lime is added to water depending on the concentration of fluoride. Furthermore, there is a need to develop an effective process that produces less waste sludge and that could replace conventional chemical coagulation, and that can be retrofitted to existing facilities.

In recent years, electrocoagulation has been successfully tested to treat wastewater, in particular in the presence of suspended matter [11–16]. Dissolved ions can also be efficiently removed by electrocoagulation. This method was reported to be very effective when treating synthetic wastewater containing indium(III), boron and phosphate ions [17–20]. Cations generated by dissolution of sacrificial anodes induce flocculation of dispersed pollutants, by reduction in the zeta potential of the suspended entities. The metal hydroxide formed acts to coagulate the liquid impurities, and the hydrogen bubbles evolved at the cathode allow flotation of foam containing the organic matter. The aggregates created can be removed by decantation or flotation.

The difference between electrocoagulation and chemical coagulation is mainly in the way that aluminum ions are delivered. In electrocoagulation, coagulation and precipitation are not conducted by delivering chemicals to the system, but they occur as a result of electrode reactions in the reactor [21]. With electrocoagulation the stability of colloids, suspensions and emulsions is influenced by electric charges. Therefore, if additional electrical charges are supplied to the charged particles via appropriate electrodes, the surface charge of particles is neutralized and several particles combine into larger and separable agglomerates [22]. The electrode assembly is the heart of the treatment facility. Therefore, the appropriate selection of its materials is very important. The most common electrode materials for electrocoagulation are aluminum and iron. They are cheap, readily available, and proven effective [23].

The main aim of the research reported in this study was to develop an empirical model for fluoride removal by a bipolar electrocoagulation process using critical parameters such as initial fluoride and supporting electrolyte concentration as well as applied potential.

2. Theory

When aluminium is used as electrode material, the main reactions are as follows:

- At the anode:

$$\mathrm{Al} \rightarrow \mathrm{Al}^{3+} + 3e^{-} \tag{1}$$

- In the solution:

$$\mathrm{Al}^{3+}_{(aq)} + 3\mathrm{H_2O} \rightarrow \mathrm{Al(OH)_3} + 3\mathrm{H}^{+}_{(aq)} \tag{2}$$

- At the cathode:

$$3\mathrm{H_2O} + 3e^{-} \rightarrow 3/2\mathrm{H}_{2(g)} + 3\mathrm{OH}^{-} \tag{3}$$

As the direct electric current passes through the anodes, the aluminium metal is oxidized to aluminium ions following Eq. (1). Afterward, the aluminium ions are transformed to polymeric species or $\mathrm{Al(OH)_3}$ flocs [24]. The Al(OH)3 floc is believed to adsorb F^- strongly as shown by Eq. (4) [11].

$$\mathrm{Al(OH)_3} + x\mathrm{F}^{-} \rightarrow \mathrm{Al(OH)}_{3-x}\mathrm{F}_x + x\mathrm{OH}^{-} \tag{4}$$

Usually a wastewater photovoltaic process contains 200–2000 mg/L F^- [11]. Literature reports that by pH adjustment (i.e. hydroxide precipitation), the fluoride concentration in semiconductor fabrication can be reduced to 10–20 mg/L [24]. Generally additional processing in the form of coagulation/flocculation is required to achieve an acceptable discharge concentration. In addition, the applied potential determines the coagulant dosage rate, and also the bubble production rate and size [25,26]. Raising the applied potential causes a corresponding increase in the oxidized aluminium production from the electrodes. A drop in the residual F^- concentration is expected when the applied potential increases. Moreover, a high voltage produces more bubbles which are helpful to separate the sludge from treated water [10].

2. Methodology

2.1. Chemicals

In order to simulate photovoltaic wastewater, after calcium precipitation, the desired concentrations of F^- solution were prepared by mixing proper amount of sodium fluoride with water. Sodium chloride was used as a supporting electrolyte and the pH was adjusted by adding sodium hydroxide (1 N) or sulfuric acid (1 N). All chemicals were obtained from Prolabo, Paris, France.

2.2. Electrocoagulation experiments

A full 2^3 factorial design of experiments was used to obtain the best conditions for fluoride removal from water solution. The three factors considered were initial fluoride concentration, applied potential, and supporting electrolyte dosage. Two levels for each factor were used; supporting electrolyte (0 and 100 mg/L of NaCl), applied potential (10 and 30 V), and initial fluoride concentration (20 and 25 mg/L). The factorial experimental design, which involves changing all the variables from one experiment to the next, was chosen in order to estimate the influence of the different variables. Factorial designs are widely used to investigate the effects of experimental factors and the interactions between those factors, that is, how the effect of one factor varies with the level of the other factors in a response. The advantages of factorial experiments include the relatively low cost, a reduced number of experiments, and increased possibilities to evaluate interactions among the variables. The most popular first-order design is the two-level full (or fractional) factorial, in which each factor is experimentally studied at only two levels that are expressed in coded form: −1 for low level and +1 for high level. The full factorial design, as employed in this study, consists of a 2*k* experiment ($k = 3$ factors, each experiment at two levels), which is very useful for either preliminary studies or in initial optimization, while fractional designs are almost mandatory when the problem involves a large number of factors

The batch electrocoagulation experiments were conducted with use of two synthetic solutions consisting of 20 and 25 mg/L F^- concentration. The defluoridation process pH was kept constant between 6 and 8 during the experiments. The experimental setup is shown schematically in Fig. 1. The defluoridation apparatus consisted of an electrocoagulation reactor with an effective volume of 1 L. The reactor chamber had three aluminium electrodes, each 100 mm × 85 mm and with an effective area of 170 cm^2. The electrodes were installed vertically, and connected in a bipolar mode. The net spacing between the electrodes was 10 mm. The current input of the DC power supply was maintained at a steady value, (i.e. 10, 20 or 30 V by means of a precision DC power supply (P. Fontaine MC 3030C). The fluorinated water was injected into the electrochemical reactor cell by means of a centrifugal Fontaine M7 feed pump, which allowed flow rates of up to 460 L/h and maintained good mixing of the synthetic solution during the electrocoagulation process. The purity of the aluminium electrodes used was 99.8%. The electrodes were sanded and washed with dilute HCl before each experiment. All studies were conducted at a temperature of 20°C.

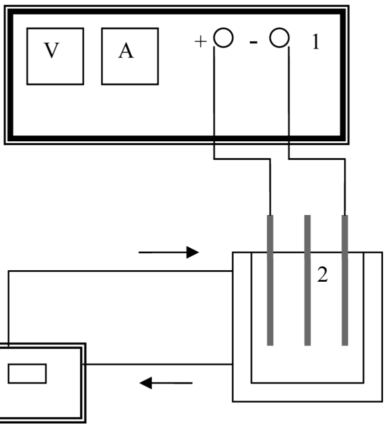

Fig. 1. Schematic diagram of the defluoridation apparatus experimental setup. 1. DC power supply; 2. EC reactor; 3. Feed pump.

The concentration of fluoride was measured using an ion meter Jenway 3205 equipped with a fluoride combination ion selective electrode. Total ionic strength adjustment buffer (TISAB) at pH 5.4 was used to maintain constant ionic strength and to prevent interference from other ions such as Al^{3+} and Ca^{2+}.

2.3. Determining the influence of initial fluoride concentration, applied potential and chloride concentration on the electrocoagulation process

The most commonly used way to remove fluoride ions from wastewater is to form calcium fluoride (CaF_2) by adding an excess of lime or other calcium salts. Adjustment of pH of the photovoltaic wastewater with lime could only reduce the F^- concentration down to 20–25 mg/l. This interval was used to determine the influence of the fluoride concentration on the electrocoagulation process with aluminum electrodes.

The investigation of the effect of changing the applied potential from 10, 20 to 30 V on fluoride removal was conducted under a fixed fluoride concentration of 25 mg/l. Ionic conductivity of the solution was enhanced with different doses of NaCl added as supporting electrolyte. A nominal value of NaCl concentration in the range from 0 to 100 mg/L was used for the present work for studying the effect of chloride on fluoride removal.

2.4. Optimization and model development

An experimental design method was employed to determine the simple and combined effects of operating variables such as initial concentration of fluoride, the dose of supporting electrolyte and applied potential on fluoride removal. Application of JMP Demo software [27] offers, on the basis of parameter estimation (Table 1), an empirical relationship between the residual fluoride concentration (*Y*) and independent variables; each having the lowest and highest levels designated by (–1) and (+1), respectively, that defined the domains of variation. The full factorial 2^3 design was employed for this particular study.

Table 1
Data for optimization operation

Variable	Factor	Level (–1)	Level (+1)
x_1	Initial fluoride concentration, mg/L	20	25
X_2	NaCl concentration, mg/L	0	100
x_3	Applied potential, V	10	30

3. Results and discussion

3.1. Influence of initial fluoride concentration on electrocoagulation process

Initial precipitation experiments with a calcium concentration of 217 mg/L could reduce the concentration of F^- down to 20–25 mg/L, which was, however, still above the industrial wastewater discharge limit of 15 mg/L. Hence batch electrocoagulation experiments were conducted using 20 and 25 mg/L F^-. The results presented in Fig. 2 show that for a fixed applied potential of 10 V the increase in fluoride concentration from 20 mg/L to 25 mg/L resulted in an increase in the required electrocoagulation time. It was noted, for example, that for 20 mg/L the standard (i.e. maximum) allowable discharge concentration limit for fluoride (i.e. 15 mg/L) was obtained in 40 min while for an initial fluoride concentration of 25 mg/L it took 60 min. Aluminium hydroxide $Al(OH)_3$ was the main component responsible for fluoride removal, because $Al(OH)_3$ floc is believed to adsorb F^- ions strongly [11]. For a given amount of electrogenerated coagulant, adsorption capacity increases at lower fluoride concentrations because there are more available adsorption sites for fluoride ions. Similar results were obtained by Yilmaz et al. [19] who found that increasing initial boron concentration in an electrocoagulant reactor increased the amount of residual ions in solution. The same trend was also found in the study of Chou et al. [17] who concluded that when the initial concentration of indium ions was increased, the electrocoagulation removal efficiency for indium ions decreased.

3.2. Influence of the applied potential on electrocoagulation

Dependence of fluoride removal on applied potential is presented in Fig. 3. The results revealed at 10 V it took 80 min to bring the fluoride concentration down to an acceptable discharge standard of 15 mg/L while at 30 V only 60 min were needed. In the electrocoagulation process, the number of the aluminium ions produced is proportional to the potential supplied. Therefore, it affects the F^- removal significantly [10]. This behavior was similar to that found in literature. For example, when treating dye wastewater, Xiong et al. [28] reported that both color and COD removal increased with increasing cell voltage. Identical results were observed by Chou et al. [18] who reported that as the applied voltage increased, the removal efficiency of indium(III) ions increased.

3.3. Effect of chloride concentration on the electrocoagulation procedure

Chloride ions decrease the passivity of the electrodes by removing the oxide layer formed on the electrode surface, due to its catalytic action. This increases coagulant generation [29]. Increasing the solution conductivity by using NaCl resulted in a reduction in cell voltage which caused a decrease in electrical energy consumption [11]. In addition, NaCl salt has other advantages; chloride ions could significantly reduce the adverse effects of other anions, such as HCO_3^- and SO_4^{2-}. The existence of the carbonate ion would lead to the precipitation of Ca^{2+} ions that form an insulating layer on the surface of the cathode [Eqs. (5) and (6)]. These will increase the ohmic resistance of the electrochemical cell.

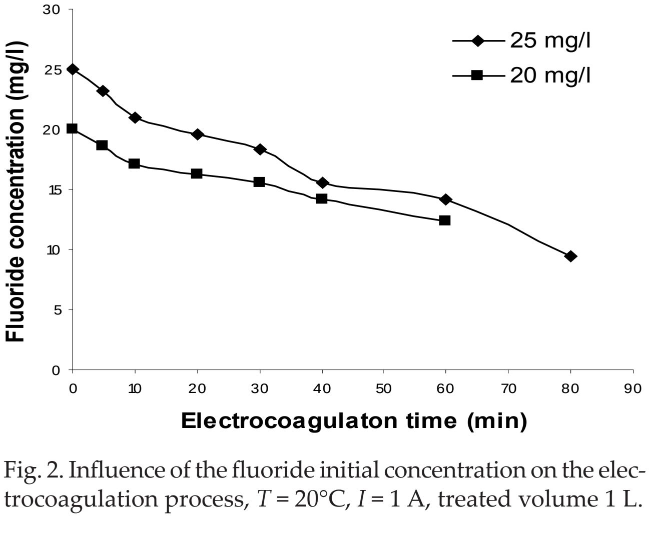

Fig. 2. Influence of the fluoride initial concentration on the electrocoagulation process, $T = 20°C$, $I = 1$ A, treated volume 1 L.

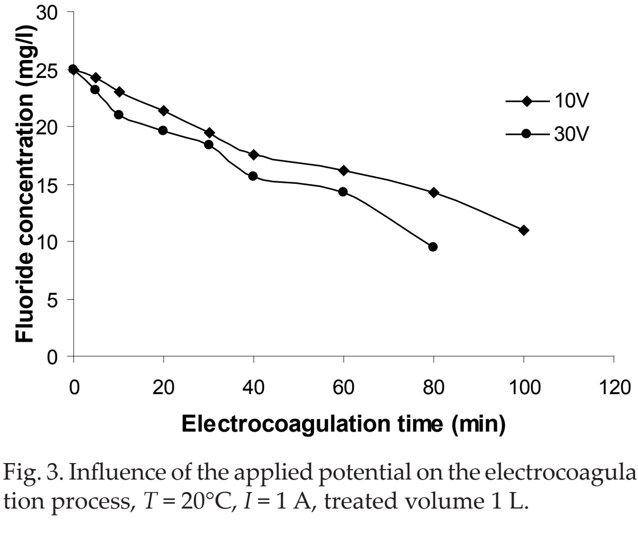

Fig. 3. Influence of the applied potential on the electrocoagulation process, $T = 20°C$, $I = 1$ A, treated volume 1 L.

$$HCO_3^- + OH^- \rightarrow CO_3^{2-} + H_2O \tag{5}$$

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \tag{6}$$

The results shown in Fig. 4 indicated that a higher concentration of Cl^- slightly increases the fluoride removal rate. This increase can be explained by the pitting corrosion phenomenon at the EC electrode due to presence of chloride ions. The reactions are as follows:

$$Al + 3HCl \rightarrow AlCl_3 + H_2 \tag{7}$$

$$AlCl_3 + 3H_2O \rightarrow Al(OH)_3 + 3HCl \tag{8}$$

Pitting corrosion can produce more aluminum hydroxide $Al(OH)_3$ flocs which are responsible for fluoride removal [10]. This behavior is similar to that found in the literature by Yilmaz et al. [19] who reported that boron removal by electrocoagulation increased with increasing dose of supporting electrolyte The same trend was found by Chou et al. [17] who concluded that when the concentration of supporting electrolyte in the reactor increased from 0 to 200 mg/L, the removal efficiency improved markedly from 22 to 80%.

3.4. Optimization and model development

A simple empirical model was developed to predict the removal of fluoride from wastewater using sacrificial Al anodes. The model was able to describe the effects of initial fluoride concentration, NaCl concentration and applied potential on the fluoride elimination efficiency from wastewater treatment. By using the least squares method, the following empirical relationship between the residual fluoride concentration (Y) and independent studied variables was obtained. The column vector of coefficients $\hat{a}$ was calculated by Eq. (9):

$$\hat{a} = \left(X^t \cdot X\right)^{-1} \cdot X^t \cdot y \tag{9}$$

where X is the calculation matrix and y is the column vector of response (see Table 2), that is, the residual concentration of fluoride.

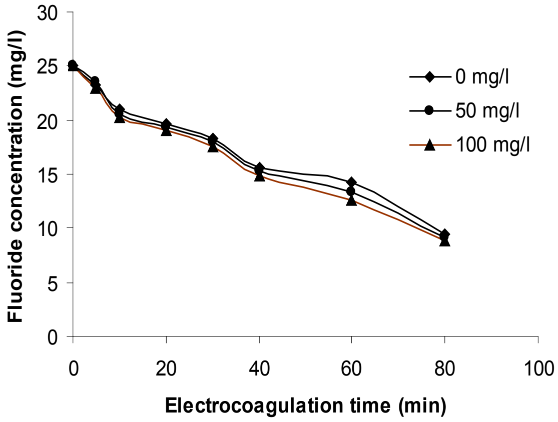

Fig. 4. Influence of supporting electrolyte concentration on electrocoagulation process, $T = 20°C$, $I = 1A$, treated volume 1 L.

As can be seen from Tables 1 and 2, x_1, x_2 and x_3 show the levels of initial fluoride concentration, NaCl concentration and applied potential respectively. While X_1, X_2 and X_3 represent the coded forms of initial fluoride concentration, NaCl concentration and applied potential respectively. The coefficients of the first-order terms indicate the effects, and those of second order express the interactions among the studied parameters while the third order coefficient represents the interacting effect of all three variables.

It should be noticed that when the effect of a factor is positive, an increase in the value of the fluoride concentration was observed when the factor changed from a low to high level. In contrast, if the effect was negative, a reduction in the concentration occurred for the high level of the same factor. The complete factorial model that could be used to fit the data in Table 2 was determined to be:

$$\begin{aligned} Y = {} & 10.237 + 1.212X_1 - 0.4275X_2 - 1.1625X_3 \\ & + 0.0375X_1X_2 - 1.137X_1X_3 + 0.0125X_2X_3 \\ & + 0.0375X_1X_2X_3 \end{aligned} \tag{10}$$

Eq. (10) is an empirical model which reveals the effect of individual variables and interactional effects for resid-

Table 2
Matrix for calculating effects for fluoride removal by EC

Trial no.	Mean	X_1	X_2	X_3	X_1X_2	X_2X_3	X_1X_3	$X_1X_2X_3$	Y: Residual fluoride concentration (mg/L)
1	1	–1	–1	–1	1	1	1	–1	9.5
2	1	1	–1	–1	–1	1	–1	1	14.2
3	1	–1	1	–1	–1	–1	1	1	8.6
4	1	1	1	–1	1	–1	–1	–1	13.3
5	1	–1	–1	1	1	–1	–1	1	9.5
6	1	1	–1	1	–1	–1	1	–1	9.5
7	1	–1	1	1	–1	–1	–1	–1	8.5
8	1	1	1	1	1	1	1	1	8.8

ual fluoride concentration from photovoltaic wastewater. As can be seen from this equation, supporting electrolyte and applied potential had a negative effect, while the initial concentration of fluoride had a positive effect on the residual fluoride found in photovoltaic wastewater. The effect of supporting electrolyte and applied potential had a negative effect, which means that the residual fluoride concentration dropped when these parameters increased. A similar conclusion was made by Trompette et al. [30] who found that both voltage and chloride ions were beneficial in the breakdown of the passive film for aluminum corrosion and consequently ion removal.

The effect of initial fluoride concentration had a positive value, indicating that the amount of final fluoride concentration increased while this factor increased. This is in good agreement with the study of Mameri et al. [14], who concluded that increasing the initial fluoride concentration results in a decrease in the fluoride removal rate by electrocoagulation. The greatest effect on fluoride removal was the applied potential as shown by Eq. (9). On the other hand, the supporting electrolyte has the least effect. All of the parameters, however, had an influence on fluoride removal. In addition, no significant interaction between initial concentration of fluoride and the dose of supporting electrolyte or between supporting electrolyte and applied potential was observed, whereas interaction between initial fluoride concentration and applied potential had a strong positive effect. Also, the interaction between the three parameters had no effect.

Despite the multitude of work on water and wastewater treatment by electrocoagulation, there is still a scarcity of models that best describe the process. This is probably due to the complexity of the electrocoagulation process and large number of variables. Design of experiments is a powerful tool for modeling and analyzing the influence of process parameters on each specific variable. The most important aspect of a design of experiment lies in the selection of the controlling factors.

4. Conclusions

The removal of pollutants from effluents by electrocoagulation has become an attractive method in recent years. This study demonstrated the applicability of the electrocoagulation method with aluminium electrodes for fluoride removal from photovoltaic wastewater. The removal efficiency was found to be dependent on the initial fluoride concentration, the applied potential, and the electrolyte concentration. A mathematical model showing the relationship between residual fluoride concentration and the effective variables was developed. From the model, it was concluded that the amount of supporting electrolyte NaCl and the applied voltage have a negative effect, while initial fluoride concentration exhibited a positive effect on final fluoride concentration. It was also shown that the most significant effect among the studied parameters was attributed to the applied voltage.

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