CHEMICAL ENGINEERING TRANSACTIONS Volume 24, 2011
Editor Sauro Pierucci
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DOI: 10.3303/CET1124202

Experimental Design for the Elimination of Fluoride from Pretreated Photovoltaic Wastewater by Electrocoagulation

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An electrocoagulation system using aluminium electrodes was studied for the removal of fluoride from pre treated photovoltaic wastewater with lime. In this regard, a full factorial experimental design was performed in order to study the main variables affecting the degradation treatment process as well as their most significant interactions. The interactions between initial concentration of fluoride and the dose of supporting electrolyte, as well as between supporting electrolyte and applied potential were no significant for fluoride removal, whereas interaction between initial fluoride concentration and applied potential had a strong positive effect. Results showed that the optimum conditions for fluoride removal from pre treated photovoltaic wastewater containing an initial fluoride concentration in the range of 20-25 mg/L were a supporting electrolyte dose of 100 mg/L and an applied potential of 30 V.

Introduction

Excessive fluoride intake over a long period of time may result in a serious public health problem called fluorosis, which is characterized by dental mottling and skeletal manifestations such as crippling deformities, osteoporosis, and osteosclerosis (WHO, 2006).An appropriate concentration of fluoride in drinking water for general good health set by WHO is considered to be less than 1.5mg/L (WHO, 1993). Fluoride also can be found in industrial wastewaters such as photovoltaic industry which uses a large amount of hydrofluoric acid in wafer etching and quartz cleaning operations (Drouiche et al., 2009a, Drouiche et al., 2009b). The traditional method of removing high F^- contents from wastewater is liming followed by precipitation (Emamjomeh et al., 2006). However this method is not suitable for wastewater containing high concentration, because the lower limit for F^- reduction is only about 20-25 mg/L. Therefore, alternative process is needed to lower fluoride concentration after calcium precipitation. Electrocoagulation is an electrolytic process that generates metallic hydroxide flocs via electro-dissolution of a soluble sacrificial anode immersed in wastewater. The electrochemically generated metallic ions hydrolyze near the anode to form a series of metal hydroxides that act to destabilize dispersed particles present in the wastewater to be treated (Chou et al.,2010). In this study, fluoride removal using electrocoagulation method was investigated. Factors affecting electrocoagulation, such as supporting electrolyte concentration, applied potential and initial fluoride concentration were also

Please cite this article as: Drouiche N., Aoudj S., Hecini M. and Ouslimane T., 2011, Experimental design for the elimination of fluoride from pretreated photovoltaic wastewater by electrocoagulation, Chemical Engineering Transactions, 24, 1207-1212
DOI: 10.3303/CET1124202

investigated. The experimental work is carried out using a 2^3 factorial design in order to examine the main factors affecting the fluoride removal and their interactions.

2. Experimentation

Initial trial calcium precipitation experiments with initial concentration of 217mg/L could reduce concentration of F^- between 20 to 25 mg/L, which is above the discharge limit which is 15 mg/L. Hence the batch EC experiments were conducted with 20 and 25 mg/L F^- concentration. Furthermore, the previous results showed that the defluoridation process is more efficient when pH is kept constant between 6 and 8 during experiments.

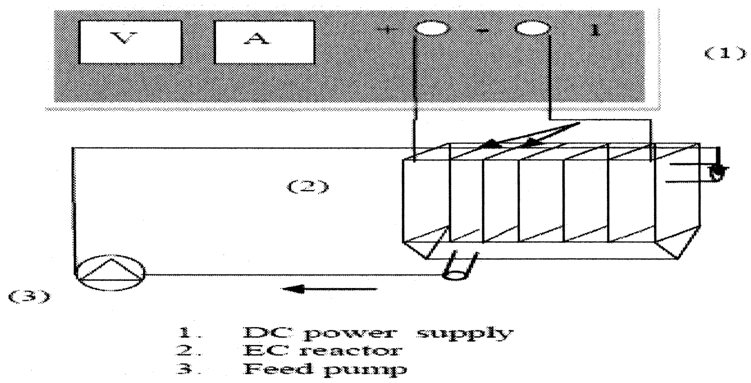

Fig.1. Schematic diagram of the experimental setup

2.1. Chemical analysis

The concentration of fluoride was measured using an ion meter Jenway 3205 equipped with Jenway fluoride combination ion selective electrode.

2.2. Electrocoagulation experiment

The experimental setup is schematically shown in Fig.1. The defluoridation apparatus consisted of an EC reactor (1 L volume). The EC chamber had three aluminum electrodes, each with a dimension of 100 mm × 85 mm and an effective area of 170 cm^2. The aluminum electrodes were installed vertically, and connected in a bipolar mode. The net spacing between the electrodes was 10 mm. The current was maintained constant, by means of a precision DC power supply. It was turned on with a voltage kept at a desired value of 10, 20 and 30 V. The fluorinated water is injected into the electrochemical reactor cell by means of the centrifugal feed pump, which allows flow rates of up to 460 L/h and maintains well mixing of the synthetic solution during electrocoagulation process.

3. Results and Discussions

3.1. Influence of the fluoride initial concentration on the electrocoagulation process

The interval of 20–25 mg/L. was used to determine the influence of the fluoride concentration on the electrocoagulation process with aluminum electrodes. The results presented in figure 2 show for the fixed applied potential of 10 V that the concentration increase induces the electrocoagulation time increase. The concentration of fluoride dropped from 25 to 14.2 mg/L in 80 min of treatment and dropped further to about 11.7 mg/L in 100 min, well below the wastewater discharge standard limit. The same trend was observed with initial concentration of 20 mg/L of fluoride. We can note that the standard discharge limit of fluoride was obtained in less time for 20 than for 25 mg/L.

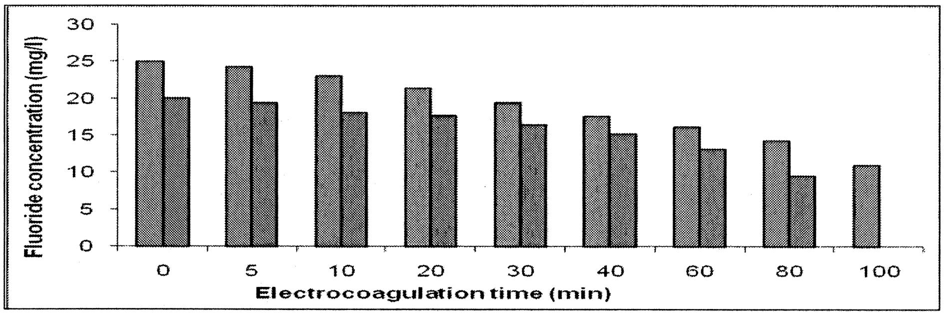

Fig.2. Influence of the fluoride initial concentration on the electrocoagulation process. T = 20°C, I = 1A, and volume treated 1 l.

3.2. Influence of the applied potential on the electrocoagulation process

The investigation of the effect of applied potential on fluoride removal was conducted under a fluoride concentration of 25 mg/L. Fig. 3, reveals that good fluoride removal was achieved in 60 and 80 min of electrocoagulation for 30 and 10 V and was under the discharges standards. The fluoride concentration decreases with increasing applied potential.

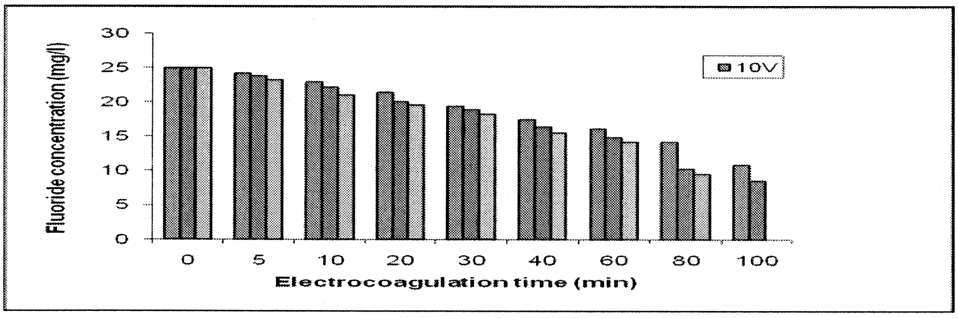

Fig.3. Influence of the applied potential on the electrocoagulation process. T = 20°C, I = 1A, and volume treated 1 l.

3.3. Effect of chloride concentration on the electrocoagulation process

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 the fluoride removal. The obtained results in Fig.4 showed that the increase of Cl^- slightly increases the fluoride removal. This can be explained by the pitting corrosion phenomenon at the EC electrode.

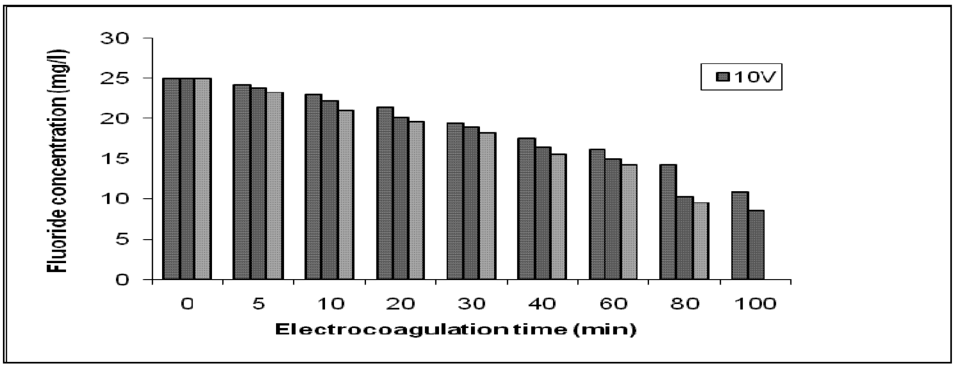

Fig.4. Influence of supporting electrolyte on electrocoagulation process. T = 20°C, I = 1A, and volume treated 1 l.

3.4. Optimization

An experimental design method was used to determine the simple and combined effects of operating variables such as initial concentration, the dose of supporting electrolyte and applied potential on fluoride removal using electrocoagulation process with aluminium plates. Application of JMP Demo software offers, on the basis of parameter estimation (Table 1), empirical relationship between the residual fluoride concentration (*Y*) and independent studied 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 appeared to be most appropriate 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

By using the least squares method, the following empirical relationship between the residual fluoride concentration (Y) and independent studied variables is obtained. The column vector of coefficients $\hat{a}$ was calculated by Eq. (3):

$$\hat{a} = (X^t.\, X)^{-1}.\, X^t.y \qquad (3)$$

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

As can be seen from Table 1, 2, x_1, x_2 and x_3 show the levels of initial fluoride concentration NaCl concentration and applied potential, respectively. X_1, X_2 and X_3 represent the coded forms of initial fluoride concentration NaCl concentration and applied potential as previous one. 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 noticeable that when the effect of a factor is negative, an increase in the value of the fluoride concentration is observed when the factor changes from low to high level. In contrast, if the effect is positive, a reduction in the concentration occurs for the high level of the same factor.

Table 2: Matrix for calculation of effects for aluminium anodes

Trial n°	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

The complete factorial model that can be used to fit the data in Table 2 is:

$$Y= 10.237 + 1.212X_1 - 0.4375X_2 -1.1625X_3 + 0.0375X_1X_2 - 1.137X_1X_3 + 0.0125X_2X_3 + 0.0375X_1X_2X_3 \qquad (4)$$

This equation reveals the effect of individual variables and interactional effects for residual fluoride concentration from photovoltaic wastewater. As can be seen from Eq. (4), supporting electrolyte and applied potential has a positive effect, while the initial concentration of fluoride has a negative effect on the removal of fluoride from photovoltaic wastewater in the range of variation of each variable selected for our work. The effect of initial fluoride concentration has a negative value, indicating that the amount of fluoride concentration decreased while this factor increased which is in good agreement with the study of N. Mameri et *al.* who concluded that increasing fluoride initial concentration decreases the fluoride removal by electrocoagulation (Mameri et al., 1998). It is known that the larger the coefficient, the larger is the effect of related parameter. On the one hand, the greatest effect on fluoride removal was applied potential. On the other hand, the supporting electrolyte has the least effect. All of the parameters have an influence on the fluoride removal by EC. In addition, no significant interaction between initial concentration of fluoride and the dose of supporting electrolyte or between supporting electrolyte and applied potential were observed, whereas interaction between initial fluoride concentration and applied potential has a strong positive effect. Also, the interaction between the three parameters has no effect.

4. Conclusion

This study demonstrated the applicability of electrocoagulation method with aluminium electrodes for fluoride removal from photovoltaic wastewater. Under optimal values of process parameters, concentration below standard limit was obtained.

In order to detect the main factors influencing the process and also the interactions between the main factors, a full 2^3 factorial design was used. From the obtained mathematical model, it is determined that the dose of supporting electrolyte NaCl and applied voltage have a positive effect, whereas initial fluoride concentration exhibited a negative effect on fluoride removal. It is also showed that the most significant effect among studied parameters is attributed to the applied voltage. On the one hand, the interaction between the initial fluoride concentration and applied potential contributes to fluoride removal from the aqueous solution. On the other hand, the other interactions were not effective on fluoride removal.

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