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Remediation of Post Treated Fluorinated Photovoltaic Wastewater by Electrocoagulation

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The remediation of post treated fluorinated photovoltaic wastewater was carried out using electrocoagulation technique. The main objective of this study was to investigate the effects of DC on the removal of fluoride from wastewater using aluminum material as anode and cathode. To optimize the maximum removal efficiency, different parameters like number of electrodes, initial pH, and current density were studied. Fluoride adsorbed aluminum hydroxide coagulant was characterized by XRD, and FTIR.

1. Introduction

Fluoride removal receives much attention because its excessive intake may cause chronic diseases such as mottling of teeth and skeletal fluorosis (Barbier O et al.,). The contamination of fluoride in groundwater and surface water could come either from natural geological sources or from industries that use fluoride-containing compounds as raw materials such as glass production, semiconductor manufacturing and photovoltaic solar cells industry (Drouiche et al., 2008; Hu et al., 2008). This latter manufacturing process is characterized not only by the large volume of water required for various unit operations but also by the variety of chemicals used for various processes. Most commonly, fluoride is simultaneously present in wastewater effluents; therefore, wastewater effluents with high fluoride must be efficiently treated prior to discharge into the environment. The suitable level of fluoride in drinking water specified by the World Health Organization (WHO) is 1.5 mg/L (WHO, 2006). The discharge standard of fluoride in industrial wastewater is 15 mg/L in Algeria. Fluoride removal techniques fall into two main categories: physical and chemical. Physical methods have proven to be both too expensive, as in the case of electrodialysis and reverse osmosis. The common chemicals used for treatments are precipitation followed by lime aluminum sulfate and ferric chloride as coagulants. At present, chemical treatment is still used but has some disadvantages like high costs of maintenance, problems of sludge handling and its disposal (Bouamra et al., 2012; Drouiche et al., 2009b). Recent research has demonstrated that electrocoagulation offers an attractive alternative to above-mentioned traditional methods for treating wastewaters (Behloul et al 2012; Drouiche et al., 2010b).

The aim of this work is to study the efficiency of electrocoagulation as a treatment method for fluorinated wastewaters. The effect of main parameters such as effect of initial concentration, number of electrodes, initial pH, and effect of current density were studied. Obtained sludge was also characterized by XRD, and FTIR.

2. Materials and Methods

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.

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2.2. Electrocoagulation experiment

The electrolytic cell (Fig. 1) consisted of a 1.5-L Plexiglas vessel that was fitted with a poly-(vinyl chloride) (PVC) cell cover with slots to introduce the electrodes. Aluminum electrodes were used as the anode and cathode. Both electrodes were square in shape (100 mm side), each with a geometric area of 100 cm^2 and with an electrode gap of 10 mm. Aluminum electrodes were arranged in bipolar mode The fluorinated water is injected into the electrochemical reactor cell by means of the centrifugal Fontaine M7 feed pump, which allows flow rates of up to 460 L/h and maintain well mixing of the wastewater during the electrocoagulation process. The purity of the aluminum electrodes used was about 99.7 %. Electrodes were sanded and washed with HCl (1 N) before each experiment. Experiments were conducted at a temperature around 25 °C.

2.3. Chemical analysis

A selective ion sensor electrode [PF4L from Tacussel (Lyon,France)] was used to determine the fluoride concentration, according to the standard method given by American Public Health Association Greenberg et al.(1992). To prevent the interference from other ions (Al^{3+}, Fe^{3+}, Cu^{2+} and Ca^{2+}), TISAB II buffer solution containing CDTA (cyclohexylenediaminetetraacetic acid, Orion Research Inc.) was added to the samples.

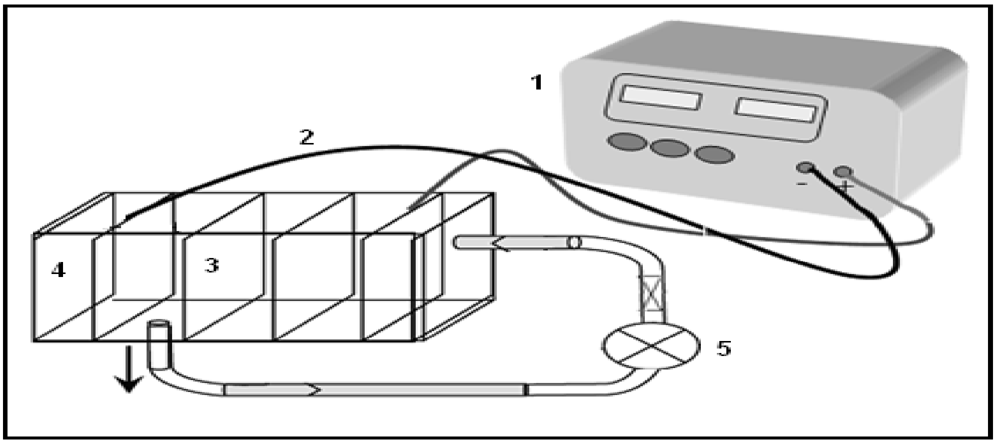

Figure 1 : Schematic diagram of the laboratory-scale electrocoagulation system. (1) Digital DC power, (2) electrical wires, (3) anode, (4) cathode, and (5) pump.

3. Results and discussion

3.1. Effect of current density

It is well known that the applied current determines not only the coagulant dosage rate but also the bubble production rate and the size and growth of flocs which can influence the treatment efficiency of the electrocoagulation (Drouiche et al., 2009c; Lemlikchi et al., 2012). To investigate the effect of current density on the removal efficiency of fluoride a series of experiments were carried out under the experimental conditions of applied current being varied from 50 to 200 mA for aluminum electrodes.

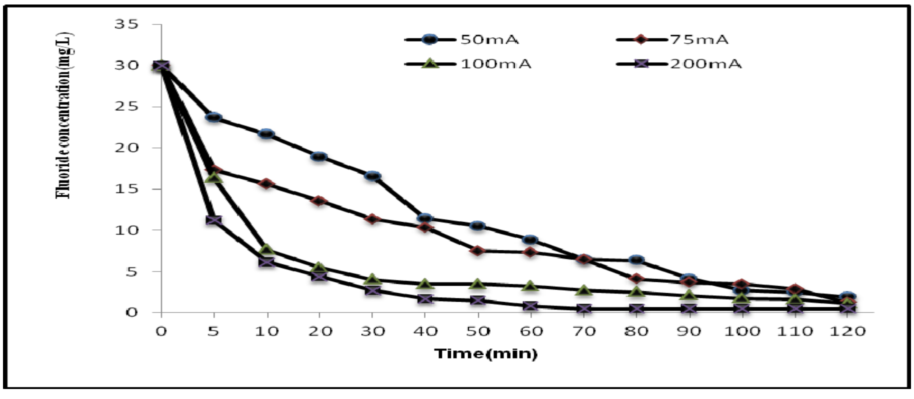

Figure 2 : Effect of current intensity on the removal of fluoride

Figure 2 illustrates that the fluoride removal efficiency increased quickly by increasing the applied current as the applied current density was increased from 9.25 to 37.03 A/m^2. The time needed to achieve standards limits discharge of fluoride for aluminum electrode decreased. The fluoride removal increased significantly and reaches standard limits in 5 and 10 minutes for applied current density of 37.03 A/m² and 18.51 A/m² respectively, whereas for 9.25 A/m^2 more time is needed to achieve the same results. This is mainly due to insufficient amount of electric power at lower voltages to produce aluminum coagulant species and gas bubbles responsible for defluoridation. According to Faraday's law, increasing the current density allows to a higher coagulant dosage per time unit.

3.2. Effect of pH

Initial pH is one of most important factors controlling EC performance (Drouiche et al., 2011a; Aoudj et al., 2010). In order to assess pH effect on fluoride ions removal, several solutions with pH values varying from 4 to 9 were studied. From figure .3, obtained results showed that the influence of pH is not negligible on the removal of the pollutant. However, it can be seen that the optimum results were obtained for pH values of 5 and 7. In fact, aluminum is mainly in the form of aluminum hydroxide in this pH range, the latter is suitable for fluoride removal. Similar results were obtained by Shen et al., (2003), when studying fluoride removal by aluminum electrodes from industrial wastewaters. Emamjomah and Sivakumar (2008) found that pH value of 7 gives the fastest removal kinetics in EC with aluminum electrodes.

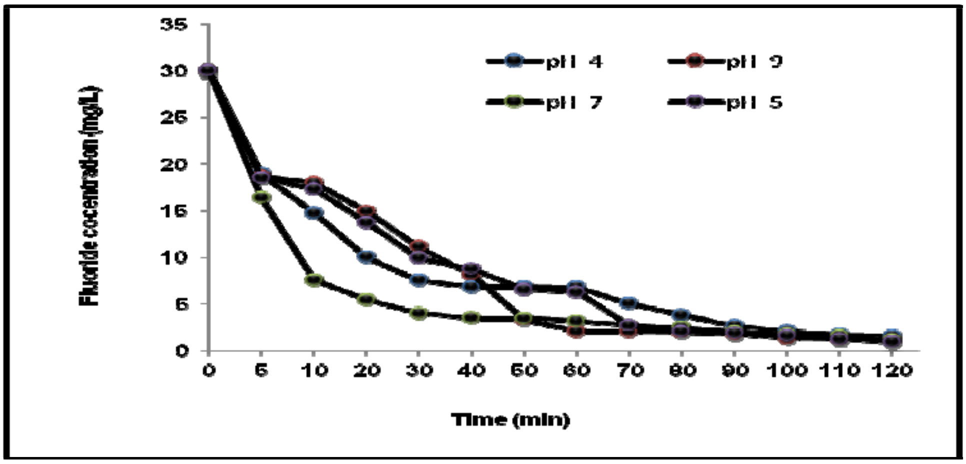

Figure 3: Effect of pH on the fluoride removal during electrocoagulation process

3.4. Effect of electrodes number

In order to optimize the most important factors influencing EC operation in bipolar mode, the effect electrodes number was studied. From figure.4, it may be observed that the different values present the same behavior, where, almost 90 % removal efficiency is obtained within 50 minutes for electrodes number 3, 4, 5 and 6. Whereas, it was only 62 % when 2 electrodes are used. However, it had been observed that the best result was obtained with the number 3 because only 10 are necessary to reach the discharge limit.

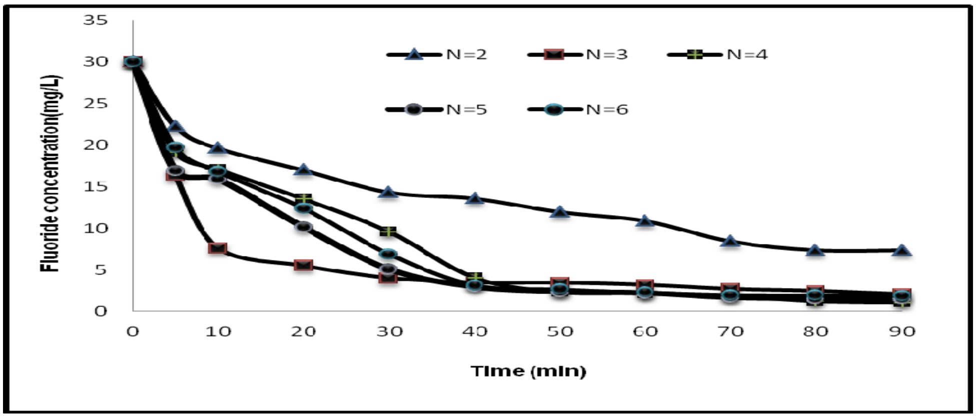

Figure 4 : Effect of number of electrodes on the removal of fluoride

3.5. Defluoridation of photovoltaic wastewater

Fluoride removal in a bipolar batch reactor, previously presented, was performed with real wastewater obtained from semiconductor research center (CRTSE) — Algiers, fluoride concentration of this wastewater after precipitation with lime is 30 mg L (Table 1).Figure 5 shows that discharge standards of fluoride are obtained in similar time than using synthetic solution. This similarity can be explained by the fact that no co-existing anions are present in the fluoride containing wastewater.

Table 1: Characteristics of photovoltaic wastewater after precipitation with lime

[F-](mg/L)	30.00
pH	1.38
Conductivity (ms/cm)	1430
COD, (mgO_2/L)	2850
Aspect	Colorless

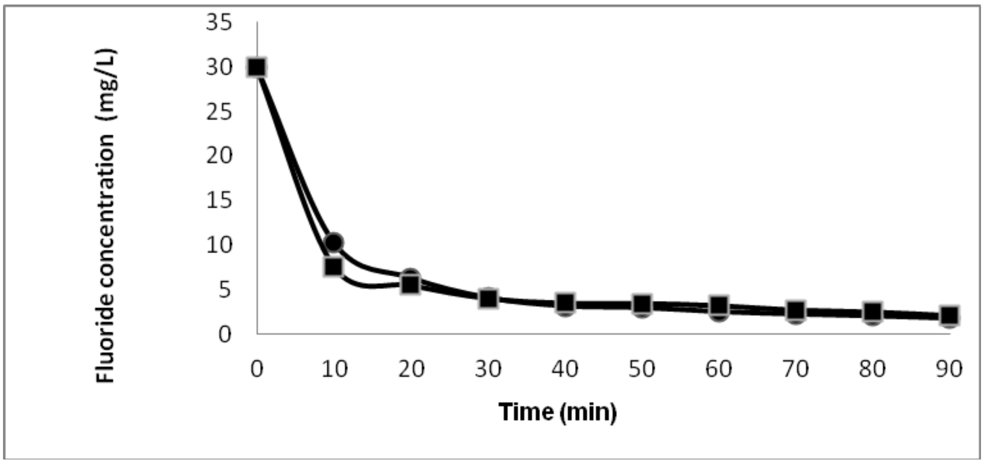

Figure 5 : Comparison ***i****n the fluoride removal of synthetic and real wastewater*

4. Characterization of the by-products

Characterization of precipitate was carried out using FTIR (Thermo- Nicolet model Nexus 670), and DRX (Philips X'Pert), respectively.Fig. 6 represents XRD patterns of the aluminum sludge, after adsorption of fluoride ions. The XRD patterns of the electrode by-product after defluoridation exhibit the characteristic of Bragg reflections possessing very broad humps and low intensity indicate that the analyzed phase possesses a short-range order, i.e., amorphous or very poorly crystalline in nature. It showed very broad and shallow diffraction peaks demonstrating most likely amorphous/poorly crystalline phases for aluminum hydroxide/oxyhydroxides. Further, the XRD analysis shows several peaks which were identified to be aluminum (B), aluminum oxyhydroxide diaspore (AlO(OH)) (C), aluminum hydride (AlH_3) (D),and fluoride aluminum hydroxide (Al(OH)F) (A).FTIR spectroscopy was useful for investigating characteristics of amorphous oxides and flocs. Fig. 7 shows the FTIR spectra of the Al electrode by-product. The broad band around 3727 cm^{-1} and 2781 cm^{-1} is attributed to hydrogen bonding from –OH of the precipitate. The band at 1640 cm^{-1} corresponds to the O–H deformation. The peaks in the 400–1300 cm^{-1} region corresponded to the stretching and bending modes of Al – O. The peaks in the 400–1300 cm^{-1} region corresponded to the stretching and bending modes of Al – O. Peaks at 612 and broad band at cm^{-1} 1067 cm^{-1} corresponded to vibrations of Al – O and Al – F bands, respectively.

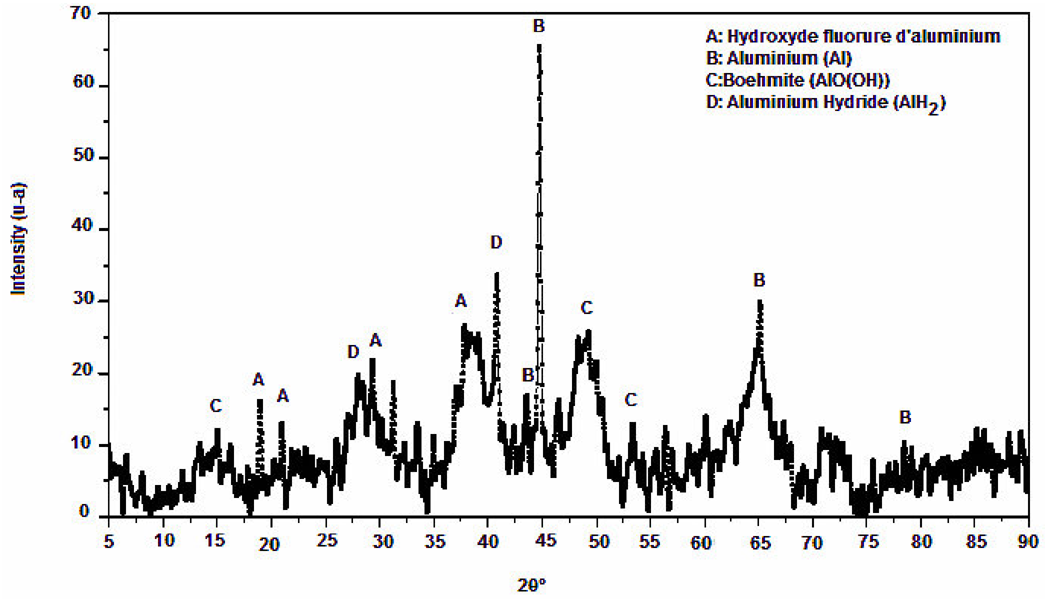

Figure 6: *XRD diagram for aluminum sludge produced in the electrocoagulation process*

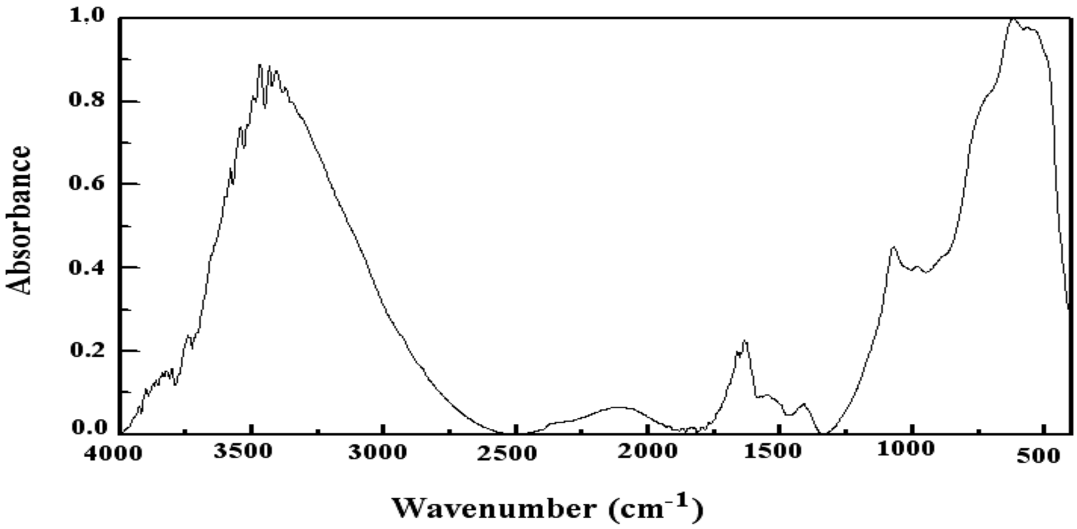

Figure 7 : FT-IR spectrum of the sludge produced in the electrocoagulation process.

5. Conclusion

The present study attempted to investigate the applicability of an electrocoagulation technique in the treatment of fluorinated photovoltaic wastewater. In this work the removal of fluoride from synthetic aqueous solution contains a concentration of fluoride of 30 mg/L by EC cell using Al as sacrificial electrodes. The removal efficiency was found to be dependent on the initial pH, the applied current density, and the number of electrodes. It was observed that these variables significantly affected the fluoride removal. The optimum fluoride removal (95 %) was obtained with typical operating conditions: current density, 18.51 A/m²; number of electrodes of 3, and initial pH, 7. The treated wastewater was found to satisfy wastewater discharge legislation and the electrogenerated sludge was discovered to be neutral and non-toxic.

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