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Fluoride removal from water and wastewater with a bach cylindrical electrode using electrocoagulation

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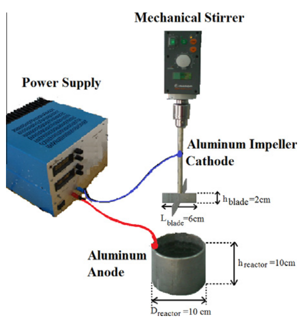
HIGHLIGHTS

- An electrochemical reactor in a unique design was used.
- 97.6% Fluoride removal was obtained using Al cylindrical anode and rotating impeller cathode.
- The recommended fluoride concentration of 1.2 mg/L by WHO was obtained within 5 min.
- The effects of anions, cations and all ions together were determined.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, an electrochemical reactor with a unique design was used for defluoridation. A rotating impeller aluminium cathode and a cylindrical aluminium anode, which until now have not been employed for fluoride removal in the literature, were used, and various operating parameters, such as the electrode material (aluminium and iron), the current density (in the range of 0.5–2 mA/cm^2), the duration of electrolysis, the supporting electrolyte dosage (in the range of 0.01–0.03 M Na_2SO_4), the initial pH (in the range of 4–8) and the presence of other ions(Ca^{2+}, Mg^{2+}, PO_4^{3-}, SO_4^{2-}), were examined to achieve optimal performance of the process. The experimental results revealed that the fluoride removal could be enhanced at pH 6, higher current density and higher electrocoagulation time using aluminium electrode. The presence of Ca^{2+} and Mg^{2+} ions also enhanced the removal efficiency while SO_4^{2-} and PO_4^{3-} ions effected adversely.

The fluoride concentration was reduced from the initial value of 5.0–0.12 mg/L, with a removal efficiency of 97.6% after 30 min treatment at the current density of 2 mA/cm^2, pH_i of 6 and presence of 0.01 M Na_2SO_4. The required electrocoagulation time to reach the WHO-recommended fluoride limit of 1.2 mg/L at 0.5 mA/cm^2 was 5 min, with an energy consumption of 0.47 kW h/m^3. The obtained results show that this specially designed electrochemical reactor is an efficient alternative for the defluoridation of the water and the wastewater.

1. Introduction

Fluorine is a natural element which occurs in geochemical deposits, minerals, and natural water systems and gets involved in food chains with drinking water, plants and cereals. Although the fluoride present in drinking water is essential for human health, an excessive intake of fluoride causes severe dental or skeletal fluorosis [1]. Therefore, the 1984 World Health Organization (WHO) guidelines suggested an optimal fluoride concentration level in the range of 1–1.2 mg/L [2].

The literature reports that many countries have regions where the water contains more than 1.5 mg/L of fluoride. A study conducted by UNICEF showed that fluorosis was endemic in at least

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27 countries across the globe [3]. In the several regions of the world especially in Africa, Asia and Turkey, groundwater contains high fluoride levels [2]. In some parts of Turkey surface and ground water contains high fluoride concentration in the range of 1.5 and 13.70 mg/L. For example, in the Isparta Province situated south-west of Turkey, the fluoride content is between 1.5 and 6.0 mg/L. In the Dogubeyazit area, the fluoride concentration is between 6.5 and 12.5 mg/L, and in the areas south and north of the Tendurek Volcano, the fluoride content ranges from 5.7 to 15.2 mg/L and from 10.3 to 12.5 mg/L, respectively [4]. For these reasons, the removal of excess fluoride from water and wastewater is important for protecting public health and the environment.

Several methods have been attempted to remove fluorides from water, namely, adsorption [5], precipitation [6], and ion exchange [7]. Recent investigations have revealed that electrocoagulation (EC) is an effective alternative for fluoride removal, both in drinking water [3] and in industrial wastewater [8]. On the other hand, the deficiency of electrocoagulation studies is the lack of the variety in reactor design. A survey of the literature showed that most EC studies have been carried out with parallel plate monopolar or bipolar electrode configuration systems. For example, fluoride removal has been performed using bipolar connections with two aluminium electrodes [9,10], three aluminium electrodes [11], four aluminium electrodes [12], seven aluminium electrodes [13,14], and nine aluminium electrodes [15] and using two monopolar aluminium electrodes [16]. An external-loop airlift reactor was also used for fluoride removal [17,18]. In summary, all the works mentioned focused mainly on the design of the electrodes.

In this study, a cylindrical aluminium reactor with a rotating impeller aluminium cathode designed in a unique manner from those reported in the literature was used. In our previous studies [3], design performance was evaluated using iron as the electrode material for fluoride removal. However, further investigations for fluoride removal were considered using aluminium electrodes because of their superior performance in a similarly designed reactor. Therefore, improving the performance of aluminium electrodes was the purpose of this study, although iron electrodes were used for comparison. The effects of the electrode material, the current density (i; mA/cm^2), the duration of electrolysis, the supporting electrolyte dosage ($C_{Na_2SO_4}$), the initial pH (pH$_i$), and the presence of other ions on the performance of the reactor for fluoride removal from synthetic solutions were investigated, and the electrical energy consumptions were determined.

2. Experimental

2.1. Electrochemical reactor

In this study, a cylindrical aluminium reactor with a rotating impeller cathode was used for the defluoridation of drinking water. The anode was a cylindrical aluminium reactor with a diameter of 10 cm, and the effective wet area of the electrode was 238 cm^2. The rotating impeller cathode had two aluminium blades with the width of 2 cm and the length of 6 cm, and it was located in the center of the reactor. It mechanically stirred the solution at 100 rpm to maintain homogeneity of the solution and prevent the particles in the reactor from settling during the electrocoagulation. The experimental system used in this study is shown in Fig. 1. The experiments were carried out batch-wise.

2.2. Materials and methods

The synthetic solutions were prepared by mixing stoichiometric amounts of sodium fluoride with deionised water. The initial fluoride concentration in the synthetic solution was 5 mg/L. The conductivity of the solutions was adjusted by adding Na_2SO_4 salt as an electrolyte. The conductivity measurements were carried out using an Inolab conductivity meter (level 1). The solution pH was measured by a pH meter (Orion 420 A).The initial pH values of the solutions were adjusted with diluted H_2SO_4 or NaOH solutions. The chemicals used in the experiments were of analytical degree.

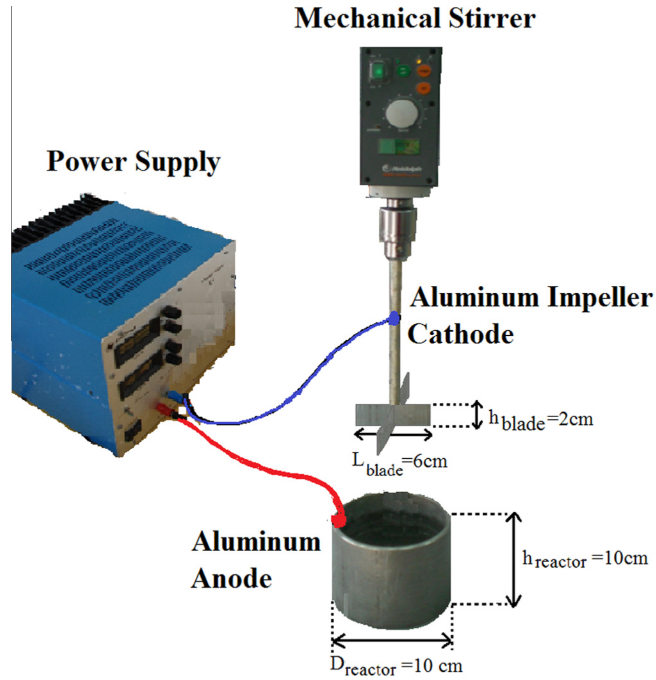

Fig. 1. Experimental set-up.

2.3. Experimental protocol

A sample solution of 400 mL was placed into the reactor for each test run. The rotating aluminium impellers (Heidolph RZR 2102) were placed in the reactor and stirred the mixture at 100 rpm while simultaneously operating as the cathode. The electrodes were connected to the power supply (Statron T-25), and a constant current was applied for 30 min for each run. The temperature and pH of the solution were not controlled but were monitored during the experiments. To follow the performance of the electrocoagulation process, samples were taken from the reactor at several time intervals during the course of electrocoagulation and were centrifuged. The supernatant liquid was analysed for fluoride content. The fluoride concentrations in the sample were determined by the SPANDS method from Standards Methods. All analyses were performed twice, and an further measurements were conducted when necessary.

The removal efficiency (RE%) was calculated using the following equation:

$$\mathrm{RE\%} = \frac{C_0 - C}{C_0} \times 100 \quad (1)$$

where C_0 and C are the concentrations of fluoride before and after the treatment, respectively, in mg/L.

3. Results and discussion

3.1. Effect of the electrode material

Aluminium and iron are the most widely used materials as sacrificial anodes in electrocoagulation studies. For comparative