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Cost effectiveness ratio in fluoride removal from water using electrochemical, coagulation, and combined electro-coagulation processes

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ABSTRACT

Different methods are used to remove a high concentration of fluoride or decrease it to the standard level. In this study, the cost-effectiveness ratio of fluoride removal (F-ion removal) by coagulation, electrochemical, and co-application of electro-coagulation processes was investigated. The initial F-ion concentration, alum dosage, and electrical current intensity were controlled as independent variables and the F-ion removal efficiency was considered as a dependent variable. The research was done in a batch system on model solution samples containing 5, 12.5, and 20 mg L^{-1} of fluoride. First, the optimum condition to reach the Iran standard for fluoride in drinking water was determined. Then the cost for each gram of removed fluoride was calculated in US$ and compared. For solutions containing an initial concentration of 5 mg L^{-1} of fluoride, the ratio of cost to eliminated fluoride in electrochemical, coagulation–electrochemical, and chemical coagulation processes was 0.0026, 0.1178, and 1.4815 US$ g^{-1}, respectively. However, these ratios were greatly reduced at higher initial concentrations. The study showed that the electrochemical process is cheaper than others while combining coagulation and electrochemical processes is more efficient. Therefore, in water and wastewater treatment plants where F-ion removal is performed using the coagulation process, adding an electrochemical process to it can reduce the cost of F-ion removal and increase the efficiency.

Keywords: Electrochemical process; Fluoride removal cost; Water treatment

1. Introduction

The contamination of water resources by fluoride is one of the global concerns. Fluoride can be found in different environments such as air, water, food, and so on [1–5]. Where the fluoride in drinking water is more than the standard level leads to various diseases, such as dental and skeletal fluorosis, immunopathy, and decreasing intelligence quotient [6–11]. Fluoride may be present in concentrations from 1 to 30 mg L^{-1} in different groundwater aquifers, in which high concentrations may be caused by the discharge of industrial wastewater or dissolution of mineral rocks [12–15]. According to the World Health Organization (WHO) guideline the fluoride concentration in drinking water should be between 1 and 1.5 mg L^{-1} [16,17]. Many processes can be used for efficient removal of fluoride ion

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such as coagulation–sedimentation, ion exchange, electrocoagulation, electrochemical process, membrane filtration, and absorption [18–26]. The feasibility of implementing any purification process in a full scale depends on the operating costs of the process. Costs of chemicals, electricity, electrodes replacement, sludge disposal, fixed costs, etc., have been considered as operating costs in many studies. Because the cost-effectiveness ratio for F-ion removal (adsorption) by these processes has not been yet investigated. The present study evaluates the cost-effectiveness of F-ion removal by coagulation, electrochemical, and combined electrocoagulation methods.

2. Materials and methods

This study was conducted in a batch system on model solution containing 5, 12.5, or 20 mg L^{-1} of fluoride. Three processes including coagulation, electrochemical alone, and combined electro-coagulation were done on every model solution concentration and the optimum conditions were investigated to reach a standard level of fluoride. F-ion removal was determined using a multi-meter device (Mi 160Milwakie, Taiwan) and fluoride ion-selective electrode (SENTEK, England) by measuring the F-ion before and after each process.

First, In the study of the coagulation process, to determine the optimum values of alum ($Al_2(SO_4)_3$, $18H_2O$) dosage and reaction time, six doses of alum (50, 100, 200, 400, 700, and 1,200 mg L^{-1}) and three reaction time (15, 30, and 45 min), was investigated. The goal was to reduce the concentration of fluoride ion to the standard fluoride level in drinking water.

The chemical coagulation process with alum was conducted by jar-test to F-ion removal. For this purpose, 15 cm^3 × 15 cm^3 × 20 cm^3 polyethylene containers were used. Rapid mixing at 100 rpm for 1 min and slow mixing at 40 rpm at 15, 30, and 45 min were done using a polyethylene-coated magnetic stirrer.

Next, in the electrochemical process, the optimal current intensity was investigated. In this stage, four current density (0.156, 0.31, 0.63, and 0.94 mA cm^{-2}) were studied. For this purpose, eight aluminum electrodes (four cathodes and four anodes 1.5 cm apart from each other) with 15 cm length, 2 cm width, and 0.1 cm thickness were used that, submerged to a depth of 10 cm in the model solution. During this process, a magnetic stirrer coated by polyethylene with a speed of 300 rpm was used.

After that, the effect of co-application of coagulation and electrochemical processes was investigated. The only difference between the last two steps was the addition of alum in optimum dosage obtained in the first step to the electrochemical process. The other specifications of this process were quite similar to the second step. All of the processes were done at pH: 6 and run time: 15 min.

Since electricity and chemical prices are the main operating parameters that affect the total costs, their prices in the industrialized countries were used to the cost estimation and comparisons. The rates for electricity and alum were 0.13 US\$ kWh^{-1} and 63.75 US\$ kg^{-1}, respectively. The operating costs were calculated in US\$ g^{-1} of fluoride removed in coagulation, electrochemical, and combined processes. The cost of electricity was calculated using the following formula:

$$P.C. = T \times V \times I \times C \times R \quad (1)$$

where P.C. is the power cost in US\$, T is the reaction time in h, V is the applied voltage, I is the power intensity in A, C is the kW W^{-1} coefficient (10^{-3}), and R is the power rate in US\$ kWh^{-1}.

Later on, descriptive statistical analysis was conducted to determine the mean and standard deviation (SD) of the results. The normality of data was studied by Kolmogorov–Smirnov test. The variance analysis with repeated measurements was conducted to investigate the effect of alum concentration and current density with regard to run time. A linear multiple regression model was developed based on the affecting factors.

3. Results and discussion

In the coagulation process, the optimal dosage of alum were 50, 100, and 200 mg L^{-1} for initial concentrations of 5, 12.5, and 20 mg L^{-1} fluoride, respectively (Fig. 1). Aoudj et al. [27]

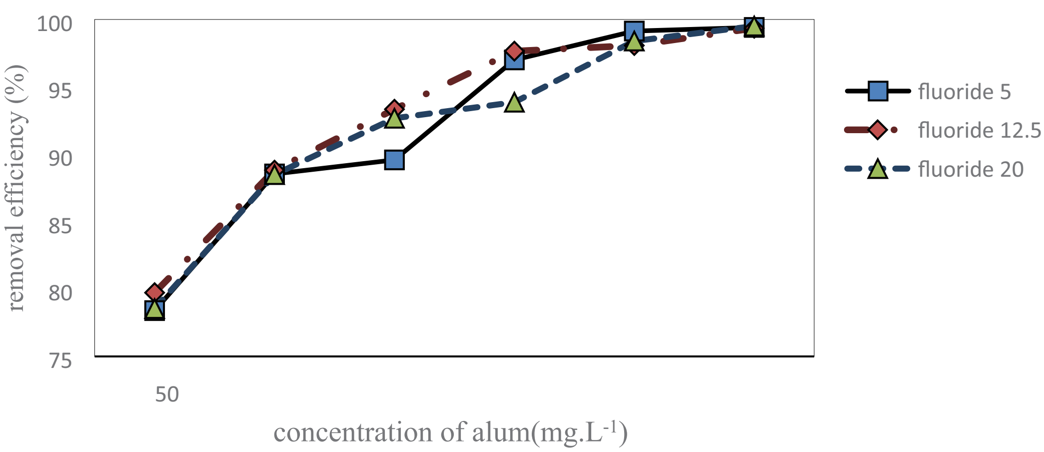

Fig. 1. Mean of F-ion removal efficiency as a function of alum dosage in the coagulation process, pH 6, and run-time = 15 min.

found 97% efficiency for 160 mg L^{-1} coagulant dose to remove fluoride [27].

In the electrochemical process, optimal current density for initial concentrations of 5, 12.5, and 20 mg L^{-1} of fluoride were 0.156, 0.63, and 0.94 mA cm^{-2}, respectively (Fig. 2). Aoudj's et al. [27] study also showed that increasing the current intensity from 100 to 150 mA increased the efficiency from 86% to 97%. The weight of electrodes were measured at the beginning and end of the process, and it was showed that the total weight loss of eight electrode was less than 1 g.

In electro-coagulation process, the optimal findings of the coagulation and electrochemical processes were combined. This method increased the efficiency of F-ion removal by 3%–6% (Table 1). Aoudj's et al. [27] study found coagulation–electroflotation method is an efficient process for wastewater treatment.

The average cost of removing 1 kg of fluoride is provided by various processes in Table 2.

The average cost of alum required in the coagulation process to remove 1 kg fluoride is the US $3,800.

This average cost in electrochemical process alone and in the simultaneous application of coagulation and electrochemical are US$ 5 and US$ 313, respectively. In the electrochemical process, the main cost was electricity consumption.

Akbay et al. [28] study showed that the operating cost of electro-coagulation process for phosphate removal was 0.94 $ m^{-3} also, claimed that this method is an economically sustainable treatment technique.

Studies by Chibani et al. [29] have shown that the low-energy electro-coagulation method reduces the residual concentration of fluoride in the WHO guidelines for F in drinking water [29].

Table 3 shows the operating costs ($ m^{-3}) to meet the WHO guidelines for fluoride in drinking water (1.5 mg L^{-1}) for different concentrations of primary fluoride. As can be seen, the operating cost in the chemical coagulation process was very high. Whereas, in the electrochemical process is significantly reduced. Electrochemical method in several studies has been considered environmentally as a cost-effective process [30–32].

Dalvand's et al. [33] findings showed that in the electro-coagulation process the dye and COD removal efficiencies were 98.6% and 84%, respectively, and the operating cost was 0.256 US$ m^{-3} that is very low at the treatment of wastewater.

Table 4 shows the cost ratio of each fluoride unit removed at different initial concentrations of fluoride (US$ g^{-1}). Clearly, the electrochemical process is efficient than the others. For these three processes, the ratio has been decreased with increasing initial concentration of fluoride. The operating cost of F-ion removal in the electrochemical process is lower than 0.38 US$ m^{-3} that has been stated by Ghosh et al. [34], Hashim et al. [35], and Thakur and Modal [36]. Also, in several works, it has been emphasized that electro-coagulation is a very effective, promising, and cost-benefit process. Also, it is much more reliable than ion exchange and membrane processes in F-ion removal [37–39].

The cost of F-ion removal in the electrochemical and electro-coagulation processes was 5 and 313 US$ kg^{-1} respectively. We found that the operating cost for F-ion removal by the combined electro-coagulation process was more than

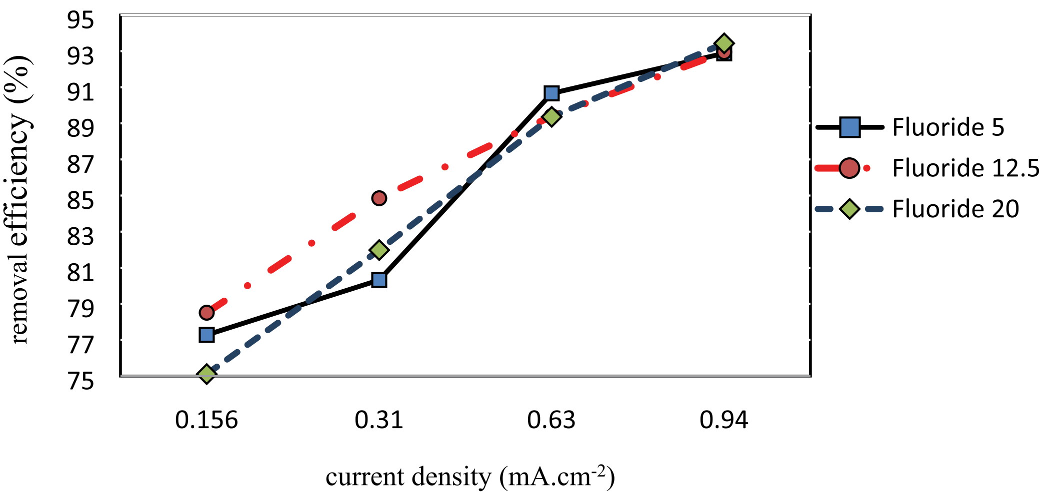

Fig. 2. Mean of F-ion removal efficiency as a function of current density in the electrochemical process, pH 6, and run-time = 15 min.

Table 1
Effect of combined electro-coagulation process on removal efficiency of fluoride

Initial concentration of fluoride (mg L^{-1})	Alum concentration (mg L^{-1})	Current density (mA cm^{-2})	Average removal efficiency (%)	*P*-value
5	10	0.03	75.04	<0.001
12.5	100	0.63	94.53	
20	200	0.94	96.14	

Table 2
Average costs to remove 1 kg of fluoride by the different processes

Process	Utilized energy or substance		Final cost US$ kg^{-1}
	Alum	Electricity	
Electrochemical	–	+	5
Electro-coagulation	+	+	313
Chemical coagulation	+	–	3,800

*Power demand in the chemical coagulation process is negligible.

Table 3
Comparison of operating costs (US$ m^{-3}) to reduce F-ion below the WHO guideline at a different initial concentration of fluoride

Process	Initial concentration of fluoride (g L^{-1})		
	0.005	0.0125	0.02
Electrochemical	0.01	0.004	0.002
Electro-coagulation	0.443	0.245	0.209
Chemical coagulation	5.8	2.3	1.4

*Power demand in the chemical coagulation process is negligible.

Table 4
US$ g^{-1} ratio fluoride-ion removed from the solution with different initial F-ion concentrations

Process	Initial fluoride concentration (mg L^{-1})		
	5	12.5	20
Electrochemical	0.0026	0.0004	0.0001
Electro-coagulation	0.1178	0.0218	0.0111
Chemical coagulation	1.4815	0.2083	0.0755

the electrochemical process and by far less than the chemical coagulation process alone. In the three mentioned methods, the operating cost were meaningfully decreased with increasing the initial fluoride concentration ($P = 0.034$).

4. Conclusion

In this study, the optimal conditions for the elimination of fluoride by coagulation, electrochemical, and hybrid processes at a fixed reaction time of 15 min, initial pH 6, on different initial concentrations of 5, 12.5, and 20 mg L^{-1} of fluoride were investigated.

Our findings showed that the electrochemical process is economically better than coagulation and combined electro-coagulation processes. Adding alum to the electrochemical process can improve the removal efficiency by about 3%–6%, or reduce the reaction time if the WHO drinking water guideline is targeted for fluoride concentrations.

The mass of removed fluoride per US$ in solutions containing initial fluoride 5 mg L^{-1} by electrochemical, electro-coagulation, and chemical coagulation processes were 387, 8.49, and 0.675 g US$$^{-1}$, respectively. However, these ratios are much higher at higher initial concentrations. Wherever F-ion removal is carried out using the coagulation process, the addition of an electrochemical process to it can reduce the cost of F-ion removal and increase efficiency. We believe that our experimental work with an economic orientation will help the societies that suffer from high F–ion concentration in their drinking water. Also, it has good comments for water and wastewater engineers engaged in F-ion removal. Low needed DC power can be supplied by photocell even in small communities.

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