APPLICATION OF ELECTROCHEMICAL PROCESSES FOR THE REMOVAL OF FLUORIDE FROM POLLUTED WATER

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INTRODUCTION

Fluoride in drinking water is an inorganic anion and is among the most important elements for human health.[1-3] The World Health Organization (WHO) guideline for the upper limit of fluoride in drinking water is between 1–1.5 $mg.L^{-1}$.[4] Excessive amounts result in various diseases such as dental and skeletal fluorosis, endocrine disruption, reproductive decline, immunopathy, decreasing intelligence quotient (IQ), and effects on hypertension as well as birth height and weight.[5-11] Fluoride is found in the wastes and air from various industrial activities, especially electroplating, and the production of semiconductors, glass, steel, ceramics, and chemical fertilizers.[12-16] In Iran, the fluoride concentration in several areas is higher than 1.5 $mg.L^{-1}$.[17,18] Various methods are used to reduce the fluoride level in water.[19-28] The electrochemical method has many advantages such as low operating cost, the capability of being used for a wide range of organic and inorganic pollutants, less contact time, and higher charged flocks entering the stilling basin.[29] As a result of the health risks of high fluoride concentrations in drinking water and the mentioned benefits of the electrochemical process, the current study aimed to investigate the application of electrochemical processes to fluoride removal from artificially polluted water.

MATERIALS AND METHODS

This is an experimental, bench scale study conducted in a batch reactor on synthetic wastewater samples containing 5, 12.5, and 20 $mg.L^{-1}$ fluoride. The total number of samples was 36.

In the electrochemical process, aluminum plates were used as electrodes. For this purpose, eight aluminum electrodes (four cathodes and four anodes which were fixed alternately 1 cm apart from each other) were applied. During the process, a magnetic stirrer was used at 300 rpm and a polyethylene container was applied as the reactor. The electrochemical process was used to determine the optimal current intensity. Four electrical current intensities (0.05, 0.1, 0.2, and 0.3 A) were studied for this purpose. Pure sodium fluoride (NaF) was used for the preparation of the fluoride stock solution (1000 $mg.L^{-1}$). Normal solutions of NaOH and H_2SO_4 were used for pH adjustment. In all runs, the pH value was set at pH 6, based on previous studies.[30,31]

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The residual fluoride was measured using a multi-meter (Mi 160 Milwaukee, Taiwan) along with a fluoride ion selective electrode (Sentek, England).

RESULTS

In this research, current intensity in the range of 0.05 to 0.3A was applied on three samples with different fluoride concentrations. Although other studies showed that there was an increase in fluoride removal by the electrochemical process with increasing run time,[32-34] in this work, the run time was fixed at 15 min. The lowest current intensity which could reduce the fluoride concentrations below the WHO guideline (1.5 $mg.L^{-1}$), was 0.05, 0.2, and 0.3 A for fluoride concentrations of 5, 12.5, and 20 $mg.L^{-1}$, respectively (Figure and Table 1). Increasing the current intensity is known to increase the removal efficiency of fluoride. Similar findings have been reported by other researchers.[35,36]

Table 2 shows the optimum conditions of the process for achieving a fluoride concentration level below the WHO guidelines (1.5$mg.L^{-1}$).

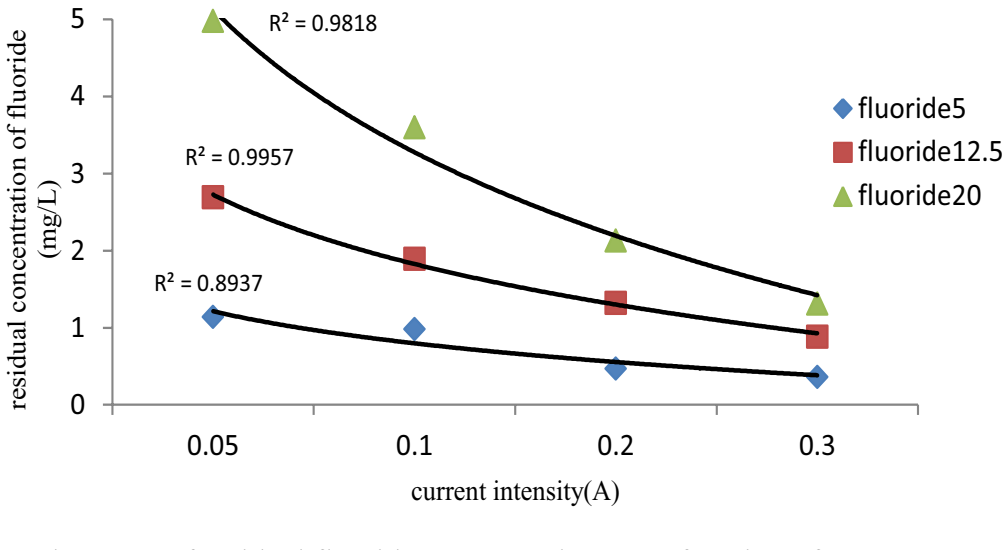

Figure. The mean of the residual fluoride concentrations as a function of current Intensity in the electrochemical process.

Table 1. The effect of different initial concentrations of fluoride on the the electrochemical process

Initial concentration of fluoride ($mg.L^{-1}$)	Number in sample	Current intensity (A)	Residual fluoride concentration ($mg.L^{-1}$)	The average removal efficiency (%)	P-value
5	12	0.05	1.13	77.28±0.94	<0.001
		0.1	0.98	80.32±3.14	
		0.2	0.47	90.66±0.77	
		0.3	0.35	92.88±0.77	
		Total	0.736	85.29±3.14	
12.5	12	0.05	2.68	78.51±7.04	<0.001
		0.1	1.89	84.85±1.78	
		0.2	1.32	89.40±0.74	
		0.3	0.88	92.95±0.54	
		Total	1.7	86.43±6.44	
20	12	0.05	4.97	75.11±4.78	<0.001
		0.1	3.60	81.98±2.5	
		0.2	2.13	89.36±0.48	
		0.3	1.31	93.43±0.80	
		Total	3.005	84.97±7.69	

Table 2. The optimum values of current intensity (A) of the reactions to achieve the fluoride standard level of below the WHO guideline of 1.5 $mg.L^{-1}$

Process	Initial concentration of fluoride ($mg.L^{-1}$)		
	5	12.5	20
Electrochemical	0.05 A (77.3% removal)	0.2 A (89.4% removal)	0.3 A (93.4% removal)

Regression analysis: According to the multiple linear regression model, to remove fluoride by the electrochemical process, the main effective factor on removing fluoride is the amount of electrical energy ($P<0.001$) while the initial concentration of fluoride has less effect on the removal efficacy ($P=0.184$). Equation 1 was developed to determine the fluoride removal efficiency as a function of electrical energy and initial fluoride concentration (Adjusted $R^2=0.847$).

$$R = 77.774 + 0.005\,E + 0.165\,F_{initial} \qquad \text{Equation 1}$$

Where:

R = Fluoride removal efficiency
E = electrical energy (kWh)
$F_{initial}$ = initial fluoride concentration (mg/L)

CONCLUSION

This research investigated the performance of the electrochemical process with aluminum electrodes, for fluoride removal from polluted water. The effects of various parameters like current intensity and the initial concentrations of fluoride were studied in detail. The electrochemical process was satisfactorily carried out on an initial fluoride concentration of 5.0–20.0 mg/L and 85% removal efficiency was obtained after 15 min. In the electrochemical process, for the initial fluoride concentrations of 5, 12.5, and 20 $mg.L^{-1}$, run time of 15 min, and pH 6, the optimum current intensity was 0.05 (0.156 mA/cm^2), 0.2 (0.63 mA/cm^2), and 0.3 A (0.937mA/cm^2), respectively. At these current densities, the initial fluoride concentration reduced from 5, 12.5, and 20 $mg.L^{-1}$ to 1.14, 1.32, and 1.31 $mg.L^{-1}$, respectively. Therefore, it can be concluded that the electrochemical process is an efficient process for fluoride removal from polluted water.

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