The Effect of the Electrode in Fluoride Removal from Drinking Water by Electro Coagulation Process

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***Abstract*—** Dental fluorosis appears in a very small percentage when fluoride in drinking water is in the range of 1-2 mg/L.Long-term intake of fluoride in concentrations higher than 4 mg/L may cause asymptomatic osteosclerosis in a small percentage of persons .Fluoride in drinking water above permissible level is responsible for human being affected by skeletal fluorosis. Electro coagulation process using iron and aluminum electrodes is proposed for removing fluoride from drinking water. Effects of different operating conditions such as treatment time, initial pH, applied voltage, type and number of electrodes and spaces between aluminum and iron electrodes and energy consumption in electro coagulation process were investigated in the batch reactor. Variable concentrations of fluoride solutions were prepared by mixing proper amount of sodium fluoride with deionizer water. Experimental results showed that aluminum electrode is more effective in fluoride removal than iron as in 40 min and initial pH of 7.5 at 20V, fluoride removal reached to 97.86%. The final recommendable limit of fluoride (1.5mgL-1) was obtained in 10 min at 20V with aluminum electrode. It can be concluded that in electro coagulation process with iron and aluminum electrodes, voltage rising, electrodes spaces decreasing, increasing number of electrodes and increase of reaction time increased fluoride removal efficiency in drinking water. In addition the effect of pH and initial concentration of fluoride varied to type of electrodes.

***Keywords*—** Electro coagulation, fluoride, drinking water, Iron and aluminum electrodes

I. Introduction

INORGANIC constituents, which may be presented in natural waters or in contaminated source waters, are found to become a major public health problem in drinking water. The presence of fluoride, as an inorganic ion, is serious more than limits in drinking water and it is a public health problem.[1] The suitable level of fluoride in drinking water specified by the World Health Organization (WHO) is ≤ 1.5mgL-1. [2,3,4]

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Different techniques like alum coagulation, bone char or calcite,[5] adsorption, membrane separation, ion-exchange, hybrid techniques and electro coagulation (EC) were reported for the removal of fluoride from drinking water [3,6]. Adsorption process using different adsorbents such as trimetal oxide [7], waste carbon slurry [8] and many low-cost materials were investigated for the removal of fluoride from aqueous medium. Membrane separation techniques were investigated for the effective separation of fluoride using electro dialysis [9], Donnan dialysis [10], nanofiltration [11] and anion-exchange membrane [12]. Garmes et.al [13] had performed defluoridation of ground water by a hybrid process combining adsorption and Donnan dialysis. Integrated biological and physicochemical treatment process for nitrate and fluoride removal was investigated by Kekkonen et.al [14]. Fluoride distribution in electro coagulation defluoridation process was investigated in order to explore the mechanism involved in fluoride removal process [15]. The kinetics was developed empirically in the removal process of fluoride using monopole electrode connection [5,16].

Electro-coagulation is a simple and efficient method to remove the flocculating agent generated by electro-oxidation of a sacrificial anode and generally made of iron or aluminum. In this process, the treatment is performed without adding any chemical coagulant or flocculants thus, reducing the amount of sludge which must be disposed. [17] On the other hand, electro coagulation is based on the in situ formation of the coagulant as the sacrificial anode corrodes due to an applied current, while the simultaneous evolution of hydrogen at the cathode allows for pollutant removal by flotation. This technique combines three main interdependent processes, operating synergistically to remove pollutants: electrochemistry, coagulation and hydrodynamics. An examination of the chemical reactions occurring in the electro coagulation process shows that the main reactions occurring at the electrodes (aluminum and iron electrodes) are:

$$Al \leftrightarrow Al^{3+} + 3e^{-} (anode)$$

$$3H_2O + 3e^{-} \leftrightarrow 3/2H_2 + 3OH^{-} (Cathode)$$

In addition, Al3+ and OH- ions generated at electrode surfaces react in the water to form aluminum hydroxide:

$$Al^{3+} + 3OH^{-} \leftrightarrow Al(OH)_3$$

Also the same chemical reactions occurring in the electro coagulation process using iron electrodes:

$$Fe(s) \leftrightarrow Fe^{3+}(aq) + 3e^{-} (anode)$$

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$$3H_2O+3e^- \leftrightarrow 3/2H_2(g)+3OH^-(aq)(cathode)$$
$$Fe^{3+}(aq)+3OH^-(aq) \leftrightarrow Fe(OH)_3$$

The aluminum and iron hydroxide flocks normally act as adsorbents and/or traps for metal ions. Therefore, they would eliminate them from the solution. [17] The main purpose of this research was to investigation of the electro coagulation process efficiency for fluoride removal from aqueous environments with iron and aluminum electrodes and determination of the effects of voltage, pH, initial concentration of fluoride and reaction time on the removal efficiency. Finally electrical energy consumption for electro coagulation process using aluminum and iron electrodes was calculated. In surface water from rivers, average concentrations have been recorded at 0.2 mg/l (0-6.5 range):in groundwater's an average of 0.3-0.4 for limestone and dolomites ,shale's and clays ,as high as 8.7 average for alkali rocks, as low as 0.1 average for basaltic rocks, and 9.2 (all in mg/l) in granitic rocks. A research of literature in regard to epidemiological studies of high concentration of fluoride in natural water can be summarized as follows : Dental fluorosis appears in a very small percentage when fluoride in drinking water is in the range of 1-2 mg/L. Long-term intake of fluoride in concentrations higher than 4 mg/L may cause asymptomatic osteosclerosis in a small percentage of persons . Crippling fluorosis has been detected in individuals exposed to fluoride levels from 10-40mg/L

Sharply reduced dental caries formation has been determined when the fluoride level is at least 0.8 mg/L, reaching maximum benefits around 3 mg/L. USEPA, April 1986 Rule mentions the evaluation of BTGA (Best Technology Generally Available) in the following water treatment technology :

*Activated Alumina Adsorption
*Reverse Osmosis (RO)
*Modified lime softening
*Adsorption using bone Char and Tricalcium Phosphate
*Anion Exchange Resins
*Electrodialysis

II. Materials & Methods

A. Methods

An experimental study in laboratory scale was carried out and a glass made tank having dimensions of 0.15m x 0.15m x 0.15m with the effective volume of 3.375 was used to conduct the experiments. Aluminum and iron electrodes with total effective area of 120 cm2 were used. Thickness of aluminum and iron plates was 2 mm and inter electrode distance was maintained at 2 cm. Electrodes were connected to a DC power supply (PS-305D, 0-5A, 0-30V). The batch electro coagulation cell with bipolar electrode connection is shown in Fig.1. Magnetic stirring at 100 rpm maintained a homogeneous solution in the batch reactor. The temperature of each system was maintained at 25±1°C.

Samples were extracted from Izeh drinking water supply in Khuzestan Province of Iran. Characteristic of Izeh water supply is shown in table 1. Fluoride solutions were prepared synthetically by dissolving proper amounts of NaF. The fluoride concentration in each reaction increased to 5mgL-1. Three different initial fluoride concentrations (1, 3,5mg/L) were used to test the influence of the initial fluoride concentration. Three applied voltage (5, 10,20V) were used to examine the effect of the applied voltage. In order to achieve the desired pH , Sulfuric acid or NaOH (0.1N) were used. pH values were measured using pH meter. Experiments were carried out in the influent pH range of 4, 7.5,10 with AL-AL,Fe-Fe electrodes in various contact times (10,20,30,40min). The reacted fluoride samples were filtered before analysis. The residual fluoride concentrations were analyzed using a spectrophotometer (DR/2000,Hach) at 580 nm according to the standard methods.[18] Finally energy consumption of electro coagulation for per cubic meter treated water in 40 min detention time in various qualifications were calculated.

TABLE I
CHARACTERISTICS OF IZEH WATER SUPPLY

Item	concentration
(mg/l $CaCO_3$) Total	250
Alkalinity (mg/l $CaCO_3$)	105
Electrical Conductivity	450
Fluoride (mg/L)	0.46
pH	7.5

III. Results

Effect of applied voltage

Current density is the most important Parameter for controlling the reaction rate in most electro coagulation processes, because it determines the coagulant dosage rate, the bubble production rate and size and the flock growth, which can influence the treatment efficiency of the electro coagulation. [17, 19,20] The effect of current density or applied voltage on the fluoride removal was investigated. Figs.1,2 reveals the variation of fluoride concentration in the EC during the experiment using Aluminum and iron electrodes. As expected, it appears that for a given time, the removal efficiency increased with the increase in current density. It can be seen that in electro coagulation with four iron electrodes with 20 mm distances and initial pH, maximum fluoride removal was 29.6% the residual fluoride concentration in effluent at the end of reaction time (40 min) didn't reach to ⟨1.5 mg/l with iron electrodes. As it can be shown from Fig. 2, the highest electrical potential (20V) produced the quickest treatment with 97.86 reductions occurring after 10 min and the lowest fluoride removal efficiency occurred in the lowest electrical potential (5V). This is ascribed to the fact that at higher voltage the amount of aluminum oxidized increased, resulting in a greater amount of precipitate for the removal of pollutants. In addition, it was demonstrated that bubbles density increases and their size decreases with increasing current density, resulting in a greater upwards flux and a faster removal of pollutants and sludge flotation. [17,21]

This findings are in line with the result of Malakootian study in 2009 in relation to hardness removal by electro coagulation and the results of Bazrafshan et al study in 2007 in relation to the capability of electro coagulation method with aluminum and iron electrodes in fluoride removal and Ghosh et al study in 2008 in relation to fluoride treatment with electro coagulation using monopolar and bipolar electrode connections. [6,17,21] As the current density decreases, the time needed to achieve similar efficiencies increased and the results of this research confirm this that the treatment efficiency was mainly affected by charge loading (Q=It), as reported by Bazrafshan et al [17]. This fact has also been confirmed by Ghernaout in his study in 2008 on Escherichia coli removal from surface water by electro coagulation method [22]. Hence, based on the results of the present study and previous studies electro coagulation process can act as a pH moderator. [17,23]

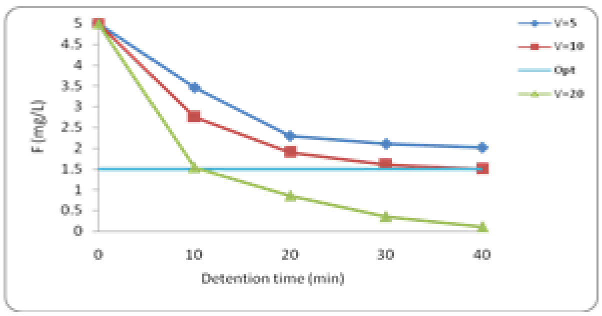

Fig. 1 Effect of applied voltage on Fluoride residue (4 Aluminum electrodes, electrode distances= 20mm)

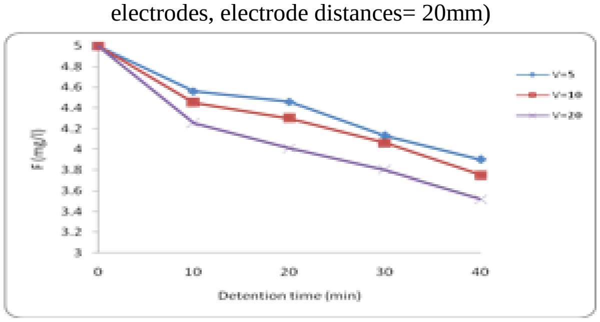

Fig. 2 Effect of applied voltage on Fluoride residue (4 Iron electrodes, electrode distances= 20mm)

The effect of distance between the electrodes

A set of experiments was performed with different electrode distances to determine fluoride removal efficiency during electro coagulation. The results obtained at 20, 30,40 mm electrode distances showed that maximum fluoride removal achieved at minimum distance between electrodes for iron and aluminum electrodes. As it is seen in Figs. 3,4 fluoride removal efficiency decrease with increase in distance between the electrodes so maximum fluoride removal achieved in 20 mm distance between electrodes in 40 min detention time for both aluminum and iron electrodes. This fact has also been confirmed by Escobar in his study in 2006 on copper, lead and cadmium removal from natural waters by electro coagulation process and the results of Ghosh study in 2008 on treatment of fluoride containing drinking water by electro coagulation that optimum interelectrode distance was 5 mm. [6,24].

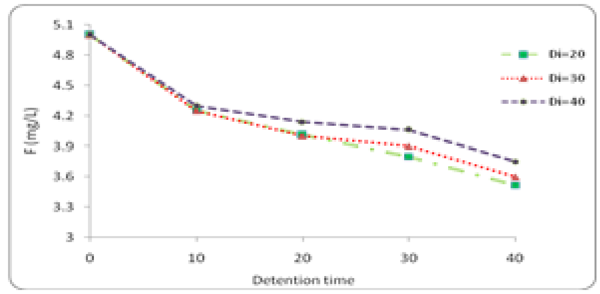

Fig. 3 Effect of distance between the electrodes on Fluoride residue (4 Aluminum electrodes, voltage=20V)

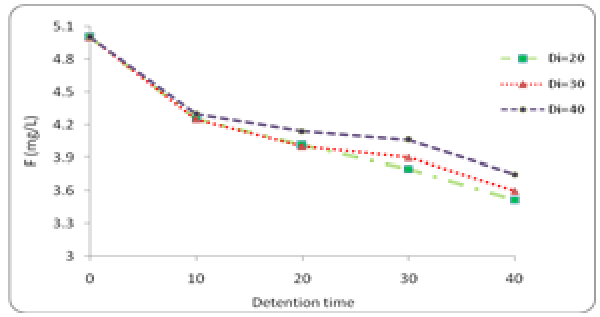

Fig. 4 Effect of distance between the electrodes on Fluoride residue (4 Iron electrodes, voltage=20V)

The effect of number of electrode:

The number of electrodes dependence of fluoride removal by electro coagulation method in equal conditions in 2, 4,6 numbers of plates is shown in Figs 5,6. It can be seen from the figures that up to 99% fluoride removal achieved with 6 aluminum electrodes and 36% removal obtained with 6 iron electrodes. With increase in number of electrodes, electricity consumption increases that lead to the increase of flock production consequently causes increasing removal efficiency. [25]

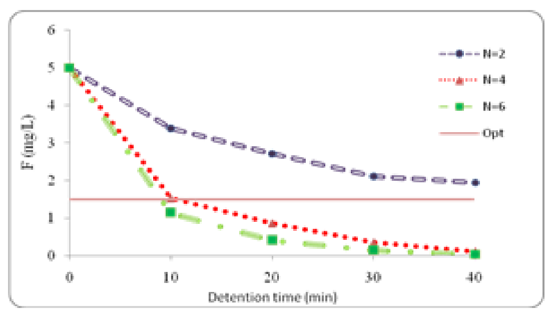

Fig. 5 Effect of number of electrode on Fluoride residue (Aluminum electrode distances=20mm, voltage=20V)

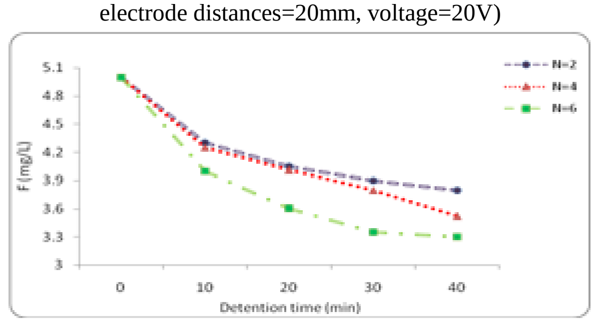

Fig.6 Effect of number of electrode on Fluoride residue (Iron electrode distances=20mm, voltage=20V)

Effect of initial pH

It has been established in previous studies that initial pH has a considerable effect on the efficiency of the electro coagulation process. Also as observed by other investigators the pH of the medium changed during the process depending on the type of electrode material and initial pH. [17] In this study, the pH was varied in the range 4,7.5,10 in an attempt to investigate the influence of this parameter on the removal of fluoride. Removal efficiencies of fluoride as a function of initial pH with iron and aluminum electrodes are presented in figs 7,8. As observed in the figures maximum fluoride removal for aluminum electrode was achieved in pH=4 and for iron electrode in pH=10. These findings are in line with the results of Bayramoglu et al, 2006 in relation to the analysis of Benzoquinone solution treatment by electro coagulation process and the results of Malakootian et al study in 2009 in relation to the hardness removal by iron electrodes in electro coagulation method. [21,26].

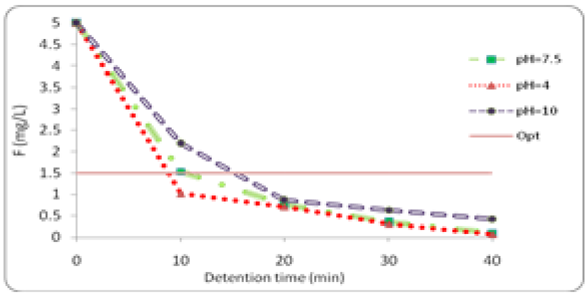

Fig. 7 Effect of initial pH on Fluoride residue (4 Aluminum electrode with 20mm distances, voltage=20V)

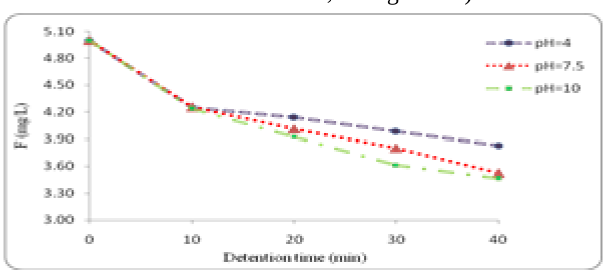

Fig. 8 Effect of initial pH on Fluoride residue (4 Iron electrode with 20mm distances, voltage=20V)

The effect of contact time

The effect of residence time was investigated on the defluoridation process in the electro coagulation. Fig 9 shows the effect of detention time in the electro coagulation process at initial pH and voltage when initial fluoride concentration is 5 mg/L. Theatrically, based on Faraday's law, current and duration of electrolysis should affect the quantity of aluminum and iron electrodes. As it can be seen from the figure up to 69% of the initial fluoride concentration decreased within 10 min of processing and the residual fluoride concentration in the effluent of the end of reaction time (40 min) reached to ⟨1 mgL^{-1} for aluminum electrodes. But as it shown in Fig 9, the residual concentration of fluoride with iron electrodes didn't reached to ⟨1 mgL^{-1} so it can't be discharge to the environment in safety. For both aluminum and iron electrodes maximum removal occurred in first 10 min. Emamjomeh investigated the fluoride removal by a continuous flow electro coagulation reactor with aluminum electrodes and reported that maximum fluoride removal occurred in initial fluoride concentration of 5mgL^{-1} in 200 ml/min flow rate and current density of 12.5A/m^2 in 40 min residence time. [20] The effect of reaction time for finding optimum fluoride concentration in the effluent was investigated. Results showed that reaction time increased when current density decreased. With increasing distances between the electrodes, reaction time increased as in 30 mm interior electrode distances, reaction time for achieving optimum fluoride removal was 20 min and it increased into 30 min when distances between electrodes increased to 40 mm.

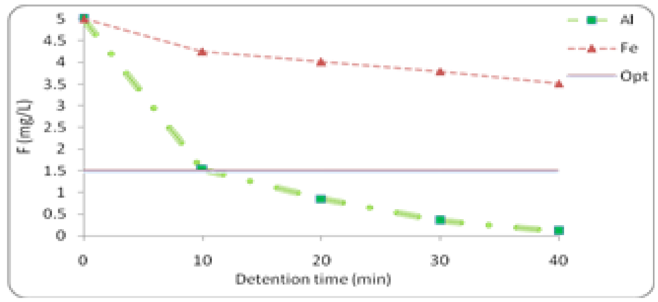

Fig. 9 Effect of contact time on Fluoride residue (4 electrodes with 20mm distances, voltage=20V)

The effect of number of electrodes on reaction time showed that with increasing number of electrodes, detention time was decreased. Initial concentration of fluoride and initial pH have direct effect on reaction time as with increasing fluoride concentration and pH, detention time was increased. The effect of kind and material of electrodes on residence time revealed optimum fluoride concentration for similar electrolysis condition obtained in 10 min for aluminum electrode and 40 min for iron electrode.

The effect of initial concentration of fluoride ion

Preliminary laboratory testing of the electrolysis cell involved determining the effect of different initial concentrations of fluoride on the efficiency of fluoride removal.

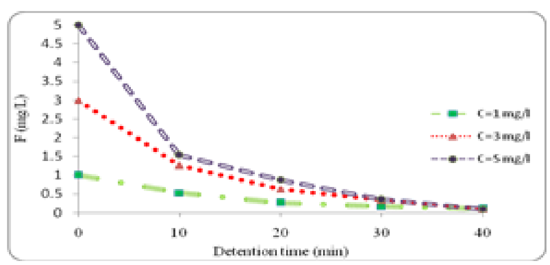

Fig. 10 Effect of initial concentration of fluoride ion on Fluoride residue (4 Aluminum electrodes with 20mm distances, voltage=20V)

From Fig.10, it appears that for various concentrations of fluoride, residual fluoride concentration was increased with increasing fluoride from 1 mgL^{-1} to 5mgL^{-1}. It is clear from Fig.11 those higher concentrations of fluoride, leads to lower fluoride removal efficiency for electrocoagulation with iron electrodes. As it is shown maximum fluoride removal was achieved in 1mgL^{-1} up to 43% and then decreased to 30% with increasing fluoride to 5mgL^{-1}. Vasudevan et al studies in 2009 on an Mg-Al-Zn Alloy as an anode for the removal of fluoride in electrocoagulation process reveal fluoride removal efficiency reduction from 96 to 20% with increasing fluoride concentration from 5 to 50 mgL^{-1}. [27]

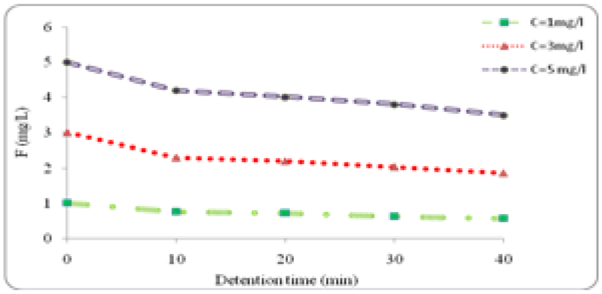

Fig. 11 Effect of initial concentration of fluoride ion on Fluoride residue (4 Iron electrodes with 20mm distances, voltage=20V)

Effect of electrode material

As it can be shown in Fig.9, aluminum electrodes are more effective in fluoride ion removal than iron electrodes. This is in complete agreement with the results obtained previously by Essadki et.al in 2009, and Bazrafshan et.al studies in fluoride removal. [17, 28]

Energy consumption

The relation between the electrical energy consumption and fluoride concentration are presented in Table 2 3 for iron and aluminum electrodes. In any electrical process, cost is incurred due to electrical energy demand, which affects the operating cost. For EC process the operating cost includes material, mainly electrodes and electrical energy costs, as well as lab our, maintenance, sludge dewatering and disposal and fixed costs.

In this investigation energy consumption was taken into account as major cost item in the calculation of the operating cost. As the table represents the energy consumption for aluminum electrodes is dramatically less than iron electrodes. As in equal condition of initial fluoride concentration, number and distances between electrodes, voltage and 40 min detention time, energy consumption for aluminum and iron electrodes was respectively 1435 kwh/kgF^- and 5590 kwh/kgF^-. Also energy consumption increased with increase in voltage and decreased with increasing distances between electrodes.

TABLE II

ELECTRICAL ENERGY CONSUMPTION DURING ELECTRO COAGULATION PROCESS USING ALUMINIUM & IRON ELECTRODES

Variation	Value	Removal efficiency		Energy consumption			
		Aluminum	Iron	Aluminum		Iron	
				kWh/m^3	kWh/kgF^-	kWh/m^3	kWh/kgF^-
Current density (volt)	5	59.6	22	0.29	97	0.32	293
	10	70	25	1.56	445	1.87	1493
	20	97.86	29.58	7.02	1435	8.27	5590
Electrode distances (mm)	20	97.86	29.58	7.02	1435	8.27	5590
	30	93	28	5.33	1147	4.8	3429
	40	86.96	25	4	920	4	3200
Number of electrodes	2	61.02	24	2.04	670	2.53	2111
	4	97.86	29.58	7.02	1435	8.27	5590
	6	99.35	34	11.29	2273	12.22	7190
pH	4	98.44	23.44	7.02	1427	8.27	7054
	7.5	97.86	29.58	7.02	1435	8.27	5590
	10	91.56	30.77	7.02	1534	8.27	5373
Initial concentrat ion	1	89	43.09	4.89	5493	7.69	17844
	3	96.67	38	6	2069	8	7018
	5	98	30	7.02	1435	8.27	5590

V. CONCLUSIONS

The present research attempted to investigate the applicability of an electrocoagulation process using aluminum and iron electrodes in fluoride removal from aqueous solutions. The influence of various variables such as pH, reaction time, voltage, fluoride concentration, material and distances between electrodes was investigated. The results showed that electrocoagulation process using iron and aluminum electrodes could successfully remove fluoride from the aqueous solutions. The results obtained with synthetic solutions revealed that the increase of detention time, voltage and number of electrodes enhanced the treatment rate for both iron and aluminum electrodes. The contact time for achieving optimum residual fluoride increased with increase in distances between electrodes and initial fluoride concentration and decreased with voltage and number of electrodes. The fluoride removal efficiency according to the study results decreased with increasing distances between electrodes. The energy consumption with aluminum electrodes is less than iron electrodes, so electrocoagulation with aluminum electrodes in fluoride removal process is more effective than iron electrodes.

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