

# An empirical model for defluoridation by batch monopolar electrocoagulation/flotation (ECF) process

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## Abstract

Excessive presence of fluoride concentration in community water supplies can cause fluorosis that affects the teeth and bones. Batch experiments with monopolar aluminium electrodes for fluoride removal were conducted and an empirical model is developed using critical parameters such as current concentration, electrode distance, and initial fluoride concentration. Fluoride ions were removed electrochemically from solution by electrocoagulation/flotation (ECF) process. The electrolytic dissolution of aluminium anodes in water produced aqueous  $Al^{3+}$  species and hydrogen bubbles at the aluminium cathodes. The fluoride removal efficiency increases steadily with increasing current values from 1 to 2.5 A. In the batch monopolar ECF process, the optimal detention time ( $d_{to}$ ) was found to be 55 min when the operational parameters including initial  $F^-$  concentration, current value, and inter electrode distance were respectively kept at 10 mg/l, 1.5 A, and 5 mm. The experimental results showed that the rate constant ( $K$ ) for defluoridation by monopolar ECF process depends on the current concentration ( $I/V$ ), electrode distance ( $d$ ) and initial fluoride concentration ( $C_0$ ). The  $Al^{3+}/F^-$  mass ratio is found to be not significantly different between monopolar and bipolar ECF systems. Overall, the results showed that the electrocoagulation technology is an effective process for defluoridation of water.

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**Keywords:** Water treatment; Monopolar electrocoagulation/flotation (ECF); Defluoridation efficiency; Empirical model; Aluminium electrodes

## 1. Introduction

Most groundwaters have low or acceptable concentration of fluoride (<1.5 mg/l) in the world [1]. In groundwater, the natural concentration of fluoride depends on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the soil and rocks, the temperature, the action of other chemical elements, and the depth of wells. Due to these variables, the fluoride concentrations in groundwater can range from less than 1 mg/l to more than 35 mg/l. In India and Kenya, concentrations up to 38.5 and 25 mg/l have been reported, respectively. The total number of people affected is not known, but an estimate would number in the tens of millions. In 1993, 15 of India's 32 states were identified as an endemic for fluorosis. A study by UNICEF shows that fluorosis is endemic in at least 27 countries across the globe [1]. These countries are: Algeria,

Argentina, Australia, Bangladesh, China, Egypt, Ethiopia, India, Iran, Iraq, Japan, Jordan, Kenya, Libya, Mexico, Morocco, New Zealand, Palestine, Pakistan, Senegal, Sri Lanka, Syria, Tanzania, Thailand, Turkey, Uganda, and United Arab Emirates. In Australia, the fluoride concentration was recorded 13 mg/l in a water bore near Indulkana region; however, it is not used for human consumption [2].

Health impacts from long-term use of drinking water with a high fluoride concentration have been summarized in Table 1 [3,4]. The maximum acceptable concentration of fluoride in water is 1.5 mg/l. Fluoride also can be found in industrial wastewaters, such as in glass manufacturing industries [5] and in high concentrations in semiconductor industries [6]. The discharge of these wastewaters without treatment into the natural environment may also contribute to groundwater contamination. To control fluoride concentrations in drinking water, several treatment options exist. A number of defluoridation processes, such as adsorption [7], chemical precipitation [8], electro dialysis [9], and electrochemical methods [10,11] have been tested globally. In the precipitation technology, alum or combination

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### Nomenclature

$A$	total electrode area ( $\text{m}^2$ )
$C_{\text{Al(T)}}$	theoretical concentration of $\text{Al}^{3+}$ ( $\text{mg/l}$ )
$C_0$	initial fluoride concentration ( $\text{mg/l}$ )
$C_t$	fluoride concentration at any time ( $\text{mg/l}$ )
$d$	distance between electrodes ( $\text{mm}$ )
$d_t$	detention time ( $\text{min}$ )
$d_{\text{to}}$	optimal detention time to achieve, $F^- = 1 \text{ mg/l}$ ( $\text{min}$ )
$\text{Ec}_i$	initial conductivity ( $\text{mS m}^{-1}$ )
$E_t$	defluoridation efficiency at any time (%)
$F$	Faraday constant, $F = 96487 \text{ (C mol}^{-1}\text{)}$
$i$	current density ( $\text{A/m}^2$ )
$I$	current (A)
$I/V$	current concentration ( $\text{A/m}^3$ )
$I_{\text{(T)}}$	total current (A)
$K$	kinetic constant ( $\text{min}^{-1}$ )
$K_{\text{exp}}$	experimental kinetic constant ( $\text{min}^{-1}$ )
$K_{\text{pre}}$	predicted kinetic constant ( $\text{min}^{-1}$ )
$M_{\text{Al}}$	molecular weight of aluminium ( $\text{g mol}^{-1}$ )
$n$	cells number
$t$	electrolysis time ( $\text{min}$ )
$T$	temperature ( $^{\circ}\text{C}$ )
$U_c$	electric potential in each cell (V)
$U_{\text{c(T)}}$	total electric potential (V)
$V$	volume ( $\text{m}^3$ )
$Z$	valence ( $Z = 3$ for aluminium)

of alum and lime are added to water with low and high concentrations of fluoride, respectively. Fluoride is then removed by flocculation, sedimentation and followed by filtration. Using chemical coagulants for precipitation is one of the most essential processes in conventional water and wastewater treatment. However, the generation of large volumes of sludge, the hazardous waste categorization of metal hydroxides, and high costs associated with chemical treatment have made chemical coagulation less acceptable compared to other processes.

An effective process that produces less waste sludge and that could replace the conventional chemical coagulation, increase the process efficiency and that can be retrofitted to existing facilities would be highly desirable. A promising process is electrocoagulation/flotation (ECF), which is an electrochemical technique, in which a variety of unwanted dissolved particles and suspended matter can be effectively removed from an aqueous solution by electrolysis. More recently ECF has been sug-

Table 1  
Health impacts from long-term use of high fluoride concentration in drinking water (NHMRC and ARMCANZ, 2004 and WHO, 2004)

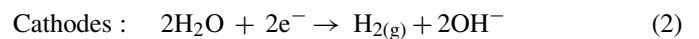
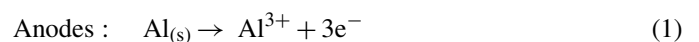
Fluoride concentration ( $\text{mg/l}$ )	Health effect
<0.5	Dental caries
0.5–1.5	Promotes dental health
1.5–4	Dental fluorosis
>4	Dental and skeletal fluorosis

gested as an alternative to conventional coagulation [12]. Some researchers [13–18] have in fact demonstrated that electrocoagulation using aluminium electrodes (as anodes) is effective for defluoridation in water and industrial wastewater treatment. It has been suggested that the electrocoagulation process for fluoride removal does not require a substantial investment [10]. Hu et al. [14] reported that defluoridation efficiency in the ECF system was almost 100% in solutions without co-existing anions ( $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^-$ ). Shen et al. [15] reported that the combination of electrocoagulation (EC) and electroflotation (EF) process was successfully applied in treating wastewater-containing fluoride.

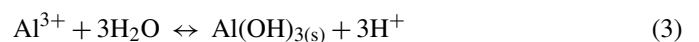
The previous results showed that the defluoridation process is more efficient when pH is kept constant between 6 and 8 during experiments. So, the pH was kept constant between this range in each run and the effects of initial pH have not been significant. Electrolysis time ( $t$ ) determines the rate of dissolution of  $\text{Al}^{3+}$  ions, as it strongly depends on the current value in the ECF process. Faraday's law can be used to describe the relationship between current value, volume of reactor and the amount of aluminium, which goes into solution. In batch ECF process, a minimum electrolysis time is required to reduce the fluorine concentration to the NHMRC and ARMCANZ [3], and WHO [4] drinking water guidelines ( $0.5 < F^- \leq 1.5 \text{ mg/l}$ ) and is called the detention time ( $d_t$ ). The main aim of this research is firstly to develop an empirical model using critical parameters such as current concentration ( $I/V$ ), electrode distance ( $d$ ), and initial fluoride concentration ( $C_0$ ) on evaluation of the rate constant ( $K$ ) for fluoride removal by a monopolar ECF process. Secondly, to determine the optimal detention time ( $d_{\text{to}}$ ) required to achieve a desirable fluoride concentration.

## 2. Fundamentals of ECF

In the ECF process, when aluminium electrodes are used, the aluminium dissolves at the anode (Eq. (1)) and hydrogen gas is released at the cathode (Eq. (2)). During the dissolution of Al at the anodes various aqueous aluminium species are produced, which depend on the solution chemistry. The aluminium species act as a coagulant by combining with the pollutants to form large size flocs. Interactions occurring within an electrocoagulation reactor are shown in Fig. 1. The electrolytic dissolution of Al anodes by oxidation in water produces aqueous  $\text{Al}^{3+}$  species [19] and the electrode reactions are outlined below:



The  $\text{H}_2$  bubbles float and hence drive the flotation process. The  $\text{Al}^{3+}$  ions further react as shown in Eq. (3) to form a solid  $\text{Al}(\text{OH})_3$  precipitate:



In its simplest form, an electrocoagulating reactor is made up of an electrolytic cell with one anode and one cathode [20]. There are two possible electrode connections in the electrocoagulation reactor, which are monopolar and bipolar connections. As shown in Fig. 2, in the case of monopolar electrode connections, an

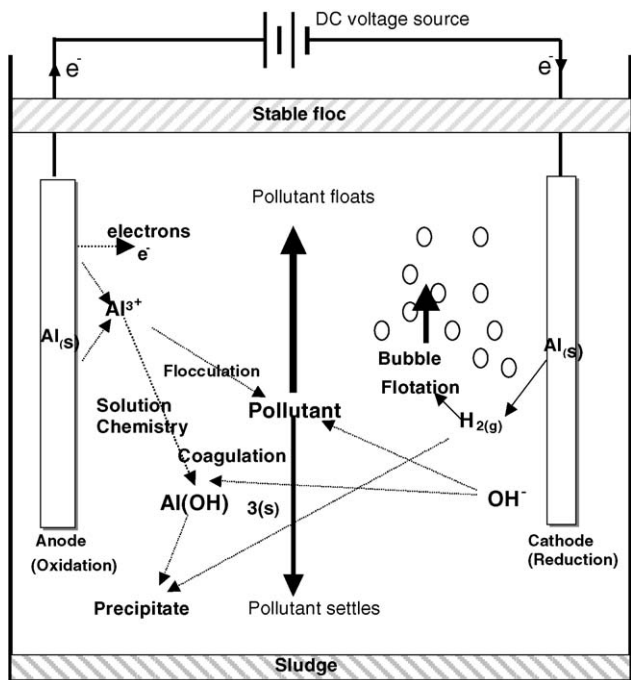


Fig. 1. Pollutant separation processes in an ECF reactor.

electric potential ( $V$ ) is connected between “ $n$ ” pairs of anodes and cathodes [21,22]. Parallel connections to each electrode will cause current ( $I_n$ ) to pass across each electrode and solution. By contrast, series connections to bipolar electrodes will cause the same current ( $I$ ) pass through “ $n$ ” electrode pairs.

3. Materials and methods

3.1. Bench scale batch ECF apparatus

A laboratory batch electrocoagulation reactor was designed and constructed to the dimensions shown in Fig. 3. In the electrochemical cell, five aluminium (purity of Al 95–97%, Ullrich Aluminium Company Ltd., Sydney) plate anodes and cathodes (dimension 250 mm × 100 mm × 3 mm) were used as electrodes. Fig. 3 also shows the electrode arrangement where three aluminium cathodes were interspersed with two aluminium anodes. The electrodes were connected using a monopolar configuration in the electrocoagulation reactor. In the case of monopolar electrodes, individual electrochemical cells can be combined in assemblies by parallel coupling. In the case of “parallel” coupling, the positive electrode of every cell is connected

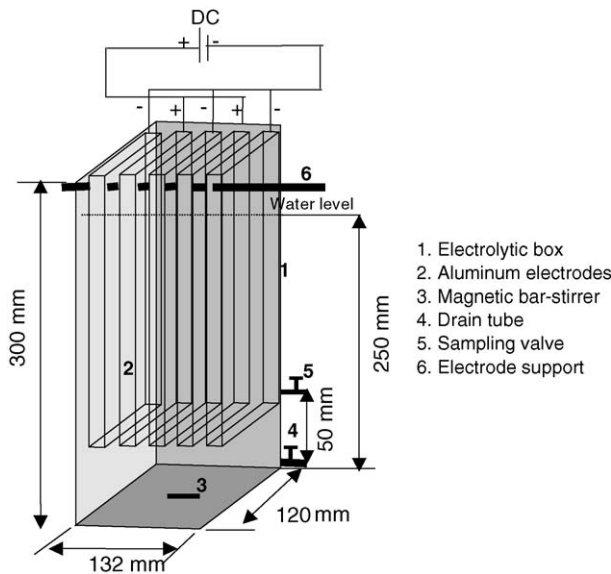


Fig. 3. Schematic diagram of the electrocoagulation reactor (ECF).

together, and the negative electrodes are connected separately, resulting in two external terminals. The voltage of every cell must be identical in parallel-coupled assemblies. The overall current passing through the assembly is the sum of the individual cell currents, while the assembly voltage is identical to the individual cell voltage. The electrodes were dipped 200 mm into an aqueous solution (net volume 3.66 l) in the Perspex reactor (dimension 300 mm × 132 mm × 120 mm). To attain uniform mixing, stirring was achieved using a magnetic bar placed between the bottom of the electrodes and the reactor. A draining tube was installed at the bottom of the box for cleaning. Samples of treated water or wastewater were collected from a port located 50 mm above the bottom of the reactor. The gaps between the two neighbouring electrode plates were varied from 5 to 15 mm for the experiments.

3.2. Experimental protocol

Electrocoagulation experiments were performed for 60 min for each run and samples were taken every 5 min interval from the drain tube section in the electrocoagulator. All experiments were conducted at a room temperature of about 25 °C with different initial  $F^-$  concentration from 10 to 25 mg/l. Current was varied over the range 1–2.5 A, however, it was held constant for each run. Based on net volume of electrobox (3.66 l),

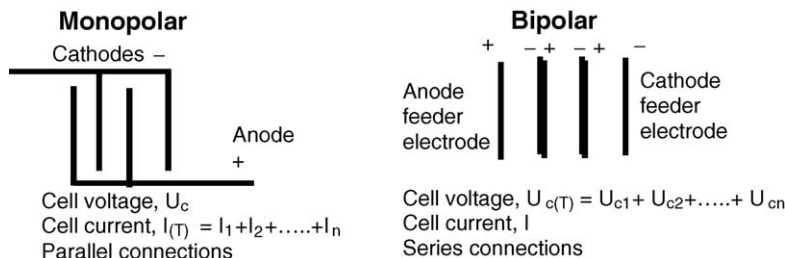


Fig. 2. Monopolar and bipolar electrode connections in the electrocoagulation reactor.

Table 2  
Characteristics of water quality bore sample used for defluoridation by ECF process

Sample	Concentration (mg/l)						
	Ions						
	Ca <sup>2+</sup>	Mg <sup>2+</sup>	K <sup>+</sup>	Cl <sup>-</sup>	pH	F <sup>-</sup>	Ec (mS/m)
Bore water sample from Central Australia	8	21	28	628	7.82	13	422

the current to volume ratio ( $I/V$ ) was varied between range of 273–683 A/m<sup>3</sup>. The pH of the feed and product water were measured for each experiment as the pH was kept constant between 6 and 8 during experiments by adding either sodium hydroxide or hydrochloric acid solutions.

### 3.3. Solution chemistry

The influence of the experimental design parameters on the defluoridation process was achieved with “synthetic” water (distilled water + NaF salt + NaCl + NaHCO<sub>3</sub>). Fluoride solutions (e.g. 10 mg/l as F<sup>-</sup>) were prepared by mixing sodium fluoride (5.2 × 10<sup>-4</sup> M) in deionized water. Sodium chloride (0.01 M) was added to the aqueous solution to increase conductivity. 1 M Sodium hydroxide and 1:5 hydrochloric acid solutions were added for final pH adjustment. Sodium bicarbonate was added to maintain alkalinity. The defluoridation efficiency was also studied using bore water from Central Australia (see Table 2).

### 3.4. Analytical techniques

Fluoride concentration was determined using the ionometric standard method [23] with a fluoride selective electrode (Metrohm ion analysis, Fluoride ISE 6.0502.150, Switzerland). To prevent interference from the Al<sup>3+</sup> ion, TISAB buffer [58 g of NaCl, 57 ml of glacial acetic acid, 4 g 1,2-cyclohexylene diamine tetraacetic (CDTA), 125 ml 6N NaOH were dissolved in 1000 ml distilled water with stirring until pH 5.3–5.5 was reached] was added to the samples. Direct current from a d.c. power supply (0 18 V, 0 20 A, ISO-TECH, IPS-1820D) was passed through the solution via the five electrodes. Cell voltage and current were readily monitored using a digital power display. Conductivity and pH were measured using conductivity meter and a pH meter, respectively. Total aluminium concentration was determined based on standard method [23] by acidifying samples with concentrated nitric acid (HNO<sub>3</sub>) to pH < 2. The samples were transferred to a beaker, 5 ml of concentrated acid HNO<sub>3</sub> was added and it was boiled without allowing sample to dry. The samples were cooled, filtered and then they were transferred to a 100 ml volumetric flask and distilled water was added to make up to the 100 mL mark. Once samples are prepared, total aluminium concentration was determined using atomic absorption spectrophotometer.

## 4. Results

### 4.1. Effect of $t$ and $I/V$

In most electrochemical processes, current ( $I$ ) and electrolysis time ( $t$ ) are the most important parameters for controlling the reaction rate in the reactor. Current not only determines the coagulant dosage but also the mixing rate within electrocoagulation. Electrolysis time ( $t$ ) determines the rate of dissolution of Al<sup>3+</sup> ions, as it strongly depends on the current value [10,13]. It is important to determine the Al<sup>3+</sup> dose achieved within the EC process. The higher the aluminium dose the quicker the defluoridation process will occur. Faraday’s law can be used to describe the relationship between current value, volume of reactor and the amount of aluminium, which goes into solution [24]:

$$C_{\text{Al(T)}} = \frac{ItM_{(\text{Al})}}{Z F V} \quad (4)$$

where  $C_{\text{Al(T)}}$ ,  $Z$ ,  $F$ ,  $V$ , and  $M_{(\text{Al})}$  are the theoretical concentration of Al<sup>3+</sup> (g/m<sup>3</sup>), aluminium valance, Faraday constant (96,500 C/mol), volume of electrobox (m<sup>3</sup>), and molecular weight of aluminium (g/mol), respectively. Based on Faraday’s formula, it is clear that electrolysis time and current concentration ( $I/V$ ) are two important parameters that determine Al dissolution in water. Current concentration is the ratio between of the current flowing through a compartment of an electrochemical cell and the volume of that compartment.

The influence of electrolysis time on the defluoridation process at different current concentrations is shown in Fig. 4. It appears that the lower the current concentration, the less aluminium is released from the anode and hence the fluoride reduction is low. The rate of change of F<sup>-</sup> concentration can be expressed as a first order kinetic model, as follows:

$$\frac{dC_t}{dt} = -KC_t \quad (5)$$

Eq. (5) can be arranged by simple integration to give:

$$C_t = C_0 e^{-Kt} \quad (6)$$

where  $C_t$ ,  $C_0$ , and  $K$  are the fluoride concentration at any time  $t$ , initial fluoride concentration, and kinetic constant, respectively. In Fig. 5, plots of  $-\ln(C_t/C_0)$  with time is shown for various current concentrations when the initial fluoride concentration

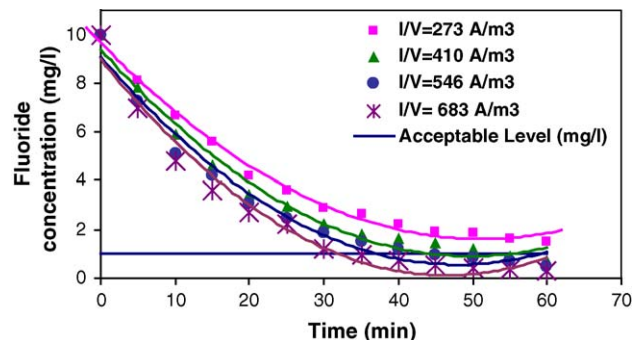


Fig. 4. Variation of fluoride concentration with time at different current concentrations ( $d = 5$  mm,  $T = 25$  °C, and  $Ec_1 = 10$  mS/m).

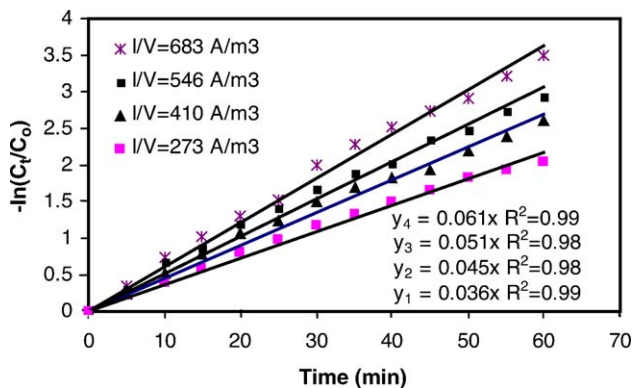


Fig. 5. Determination of the kinetic constants for the defluoridation by ECF process at different current concentrations ( $C_0 = 10$  mg/l,  $T = 25^\circ\text{C}$ ,  $d = 5$  mm, and  $E_c = 10$  mS/m).

was 10 mg/l. It appears that the reaction rate increases from 0.036 to 0.061  $\text{min}^{-1}$  when current concentration is increased from 273 to 683  $\text{A/m}^3$  in the solution, respectively.

From Fig. 5, the linear relation for each current value confirms the fact that the kinetics of defluoridation follows the exponential law with time. This result is in agreement with the results obtained by Mameri et al. [11] in a bipolar electrocoagulation reactor.

#### 4.2. Effect of $C_0$ and $d$

The experiments were conducted by changing initial fluoride concentration from 10 to 25 mg/l and the electrodes distancing ( $d$ ) between range of 5–15 mm. The effects of different initial fluoride concentrations and the electrode distance on the defluoridation process at various current concentrations are shown in Fig. 6(a and b) and Fig. 7(a and b), respectively. At the same current concentration, it is clear that the residual fluoride concentration increases when the initial fluoride concentrations and the electrode distance are increased. This is possibly due to insufficient of aluminium hydroxide complex formation on both cases.

In Fig. 6(a and b), plots of  $-\ln(C_t/C_0)$  with time are shown for initial fluoride concentrations of 15 and 25 mg/l at various current concentrations. It appears that the reaction rate decreased when initial fluoride concentration is increased in the solution. In Fig. 7(a and b), plots of  $-\ln(C_t/C_0)$  with time are shown for the electrode distances of 10 and 15 mm at different current concentrations. The results show that when the distance between the cathodes and the anodes is increased from 5 to 15 mm, the resistance between electrodes is also increased and the fluoride removal efficiency decreases.

## 5. Discussion

### 5.1. Analysis of data

Based on the experimental results, presented in Fig. 5, Fig. 6(a and b), and Fig. 7(a and b), the rate constant ( $K_{\text{exp}}$ ) may depend on the current concentration ( $I/V$ ), initial fluoride concentration ( $C_0$ ), and electrodes distance ( $d$ ) for a constant

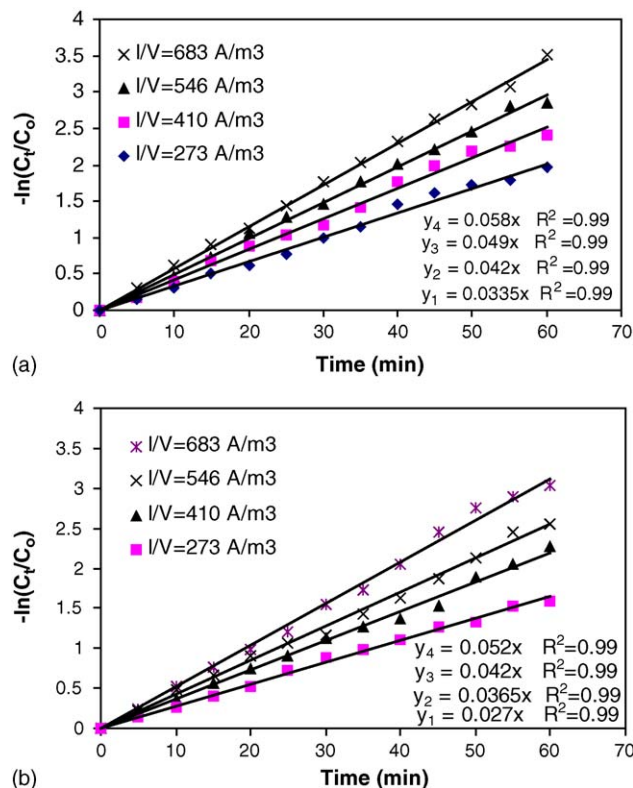


Fig. 6. Determination of the kinetic constants for the defluoridation by ECF process at different current concentrations ( $d = 5$  mm,  $T = 25^\circ\text{C}$ , and  $E_c = 10$  mS/m): (a)  $C_0 = 15$  mg/l, (b)  $C_0 = 25$  mg/l.

temperature and pH. The  $K_{\text{exp}}$  can be expressed as:

$$K_{\text{exp}} = f(I/V, C_0, d) \quad (7)$$

A multiple regression analysis is performed using the SPSS statistical package. The results are shown in Table 3 and present a high degree of correlation ( $R^2 = 0.99$ ) for the following equation:

$$K_{\text{pre}} = 10^{-5}[5.9(I/V) - 37.1(C_0) - 82.1(d) + 2746.4] \quad (8)$$

A linear relation between  $K_{\text{exp}}$  and  $K_{\text{pre}}$ , is illustrated in Fig. 8, showing that there is no significant difference between measured and predicted rate constants. Eq. (8) can now be substituted into Eq. (6) to provide Eq. (9) as:

$$C_t = C_0 e^{-10^{-5}[5.9(I/V) - 37.1(C_0) - 82.1(d) + 2746.4]t} \quad (9)$$

Eq. (10) defines the defluoridation efficiency ( $E_t$ ), where  $C_t$  is the fluoride concentration at any time  $t$  and  $C_0$  is the initial fluoride

Table 3  
Summary of statistical results for the predictive equation

Variable	B	SE B	Bata	T	Sig T
I/V	5.92E-05	1.2281E-06	0.906807	48.257	0.0000
$C_0$	-0.000371	3.0076E-05	-0.231693	-12.33	0.0000
$d$	-0.000821	4.5942E-05	-0.335738	-17.867	0.0000
Constant	0.027464	0.000918		29.928	0.0000

$R^2 = 0.99$ .

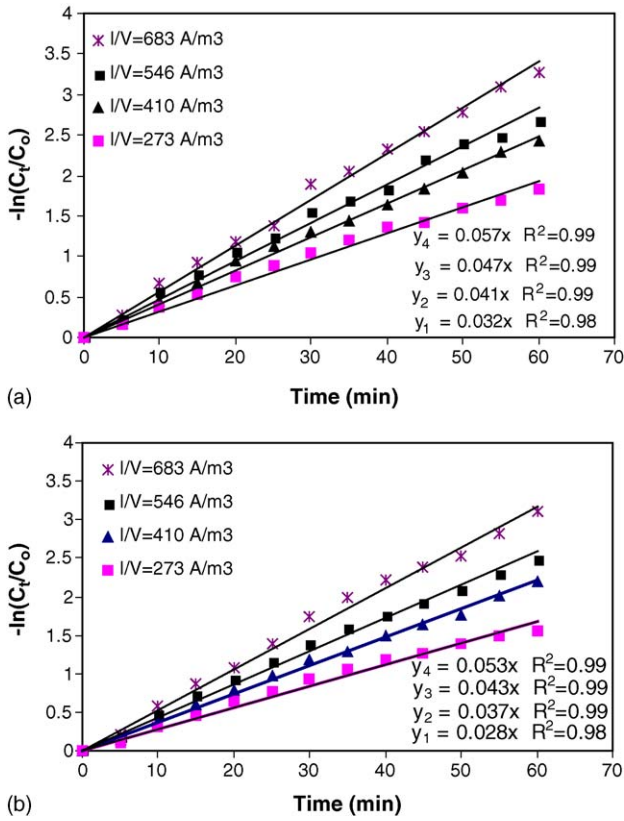


Fig. 7. Determination of the kinetic constants for the defluoridation by ECF process at different current concentrations ( $C_0 = 10$  mg/l,  $T = 25^\circ\text{C}$ , and  $Ec_i = 10$  mS/m): (a)  $d = 10$  mm, (b)  $d = 15$  mm.

concentration:

$$E_t = 1 - C_t/C_0 \quad (10)$$

Eq. (10) can be rearranged to provide an equation for removal efficiency as:

$$E_t = 1 - e^{-10^{-5}[5.9(I/V) - 37.1(C_0) - 82.1(d) + 2746.4]t} \quad (11)$$

The efficiency calculated by the empirical equation is plotted against the experimental fluoride removal efficiency in Fig. 9. As expected the match is very good with high confidence inter-

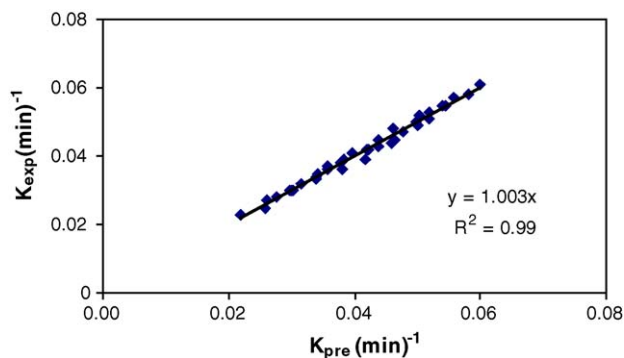


Fig. 8. Relationship between predicted rate constant ( $K_{pre}$ ) and experimentally determined rate constant ( $K_{exp}$ ) for defluoridation by ECF process ( $I = 1-2.5$  A,  $V = 3.66$  l,  $C_0 = 10-25$  mg/l,  $d = 5-15$  mm,  $T = 25^\circ\text{C}$ ,  $Ec_i = 10$  mS/m, and pH 6–8).

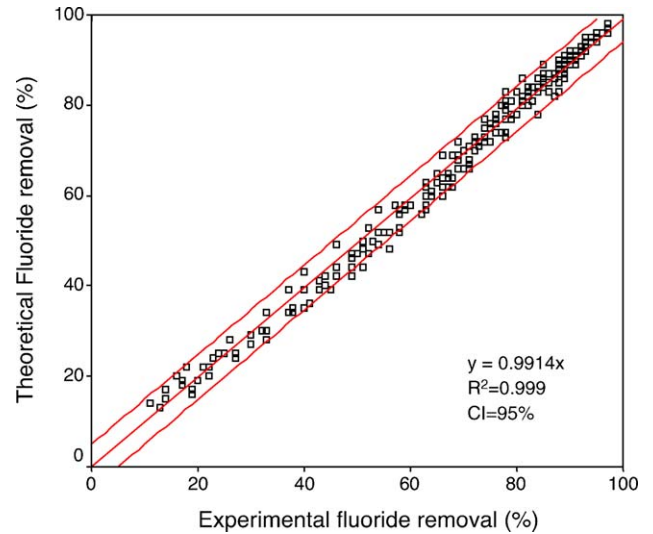


Fig. 9. Relationship between theoretical fluoride removal efficiency and the experimental defluoridation efficiency by ECF process at different operational parameters ( $I = 1-2.5$  A,  $V = 3.66$  l,  $C_0 = 10-25$  mg/l,  $d = 5-15$  mm,  $Ec_i = 10$  mS/m, pH 6–8, and  $T = 25^\circ\text{C}$ ).

val. The highest current (2.5 A) produced the quickest fluoride removal due to the ready availability of  $\text{Al}^{3+}$  ions in the solution. However at higher currents, more coagulant (aluminium) is available per unit time, which may be unnecessary, because not only excess residual aluminium is unsafe for drinking water but also high current is also uneconomic in terms of energy consumption. In this batch ECF process, the minimum electrolysis time required to reduce the fluoride concentration to the NHMRC and ARMCANZ [3], and WHO [4] drinking water guidelines ( $0.5 < F^- \leq 1.5$  mg/l) is defined as the detention time ( $d_t$ ). The detention time was experimentally determined to achieve the desirable fluoride concentration range in the electrocoagulator. From Eq. (6), the detention time ( $d_t$ ) can be expressed as:

$$d_t = \frac{1}{K} \ln\left(\frac{C_0}{C_t}\right) \quad (12)$$

When the fluoride concentration ( $C_t$ ) reaches to 1 mg/l, the optimal detention time ( $d_{to}$ ) can be expressed as Eq. (13):

$$d_{to} = \frac{1}{K} \ln(C_0) \quad (13)$$

Eq. (8) can be substituted into Eq. (13) to provide:

$$d_{to} = \frac{1}{10^{-5}[5.9(I/V) - 37.1(C_0) - 82.1(d) + 2746.4]} \ln(C_0) \quad (14)$$

From Eq. (14), the optimal detention time for fluoride removal using monopolar ECF process is calculated for various operational parameters including current values (1–2.5 A), electrodes distance (5–15 mm), and initial fluoride concentrations (10–25 mg/l). Defluoridation experiments were also conducted using bore water sample taken from Central Australia (see Table 2). This bore is currently not used for human consumption. A similar research by authors [25] showed that anion such as  $\text{Cl}^-$

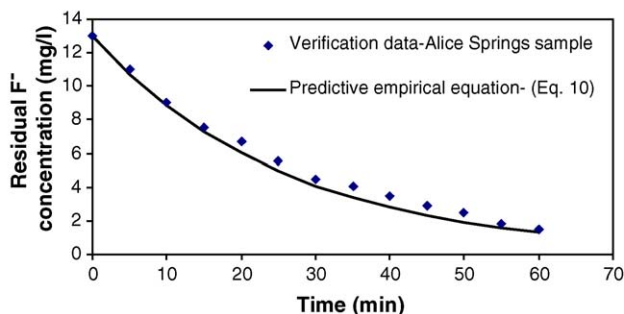


Fig. 10. Comparison between predicted residual fluoride concentrations with electrolysis time for the different experimentally measured data ( $I=1.3$  A,  $V=3.66$  l,  $C_0=13$  mg/l,  $d=7$  mm,  $E_c=422$  mS/m, pH 7.8).

had no significant effect on the defluoridation efficiency. For the same operational conditions, Fig. 10 presents a comparison between the predicted and measured residual fluoride concentration for the Central Australian bore sample for the monopolar ECF system. The result shows good agreement between the experimentally measured data (independent data) and the predictive Eq. (9).

### 5.2. Monopolar and bipolar processes

The influences of selected parameters on defluoridation by monopolar ECF process obtained are compared with results obtained by other researchers in bipolar electrocoagulation process. Earlier monopolar ECF experiments showed that the mass ratio  $Al^{3+}/F^-$  was between 13 and 17.5 for various initial fluoride concentrations from 10 to 25 mg/l and for the pH range 6–8 [25–26]. This result is in agreement with the results obtained by Mameri et al. [11] observation in the bipolar electrocoagulation process. It can be concluded there is no significant difference in  $Al^{3+}/F^-$  mass ratio between monopolar and bipolar ECF system.

Previous research by the authors [26] showed that the  $Ca^{2+}$  ion competition effect on defluoridation process is very significant. Thus, a comparison is also made on the residual fluoride concentrations between the authors' data and Mameri data [11] as shown in Fig. 11. It is clear that there is no significant difference between the residual fluoride concentration in monopolar

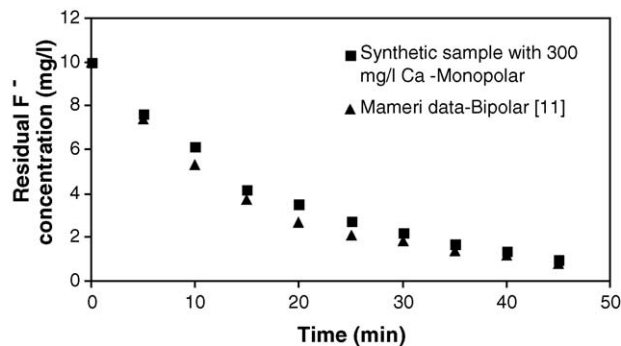


Fig. 11. Comparison between monopolar and bipolar systems for defluoridation by ECF process for the same concentration of  $Ca^{2+}$  and the same operational parameters ( $i=10.2$  A/m<sup>2</sup>,  $C_0=10$  mg/l,  $d=20$  mm, and pH 6–8).

and bipolar systems for the same water quality when the batch monopolar experiments were conducted to investigate the effects of the concentration of  $Ca^{2+}$  ion (300 mg/l) on the defluoridation efficiency.

## 6. Conclusion

An empirical model is developed to relate the critical parameters such as current concentration, electrode distance and initial fluoride concentration with the rate constant ( $K$ ) for fluoride removal using monopolar ECF process. Based on the operational parameters, an empirical equation is given to calculate the optimal detention time for fluoride removal. The results show good agreement between the experimental data and the predictive equation. It is also found that there is no significant difference in  $Al^{3+}/F^-$  mass ratio and the residual fluoride concentration between monopolar and bipolar ECF systems when the same operational parameters are used. It is concluded that the electrocoagulation technology using aluminium electrodes is a viable process for defluoridation of water supplies that contains excess fluorides.

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