



DEFLUORIDATION OF SEPTENTRIONAL SAHARA WATER OF NORTH AFRICA BY ELECTROCOAGULATION PROCESS USING BIPOLAR ALUMINIUM ELECTRODES

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Abstract—The purpose of this paper is to suggest an efficient defluorination process which does not require a big investment. For this, the electrocoagulation process with aluminium bipolar electrodes was used. In the first step, the influence of parameters such as inter-electrode distance, fluoride concentration, temperature and the pH of the solution, were investigated and optimized with synthetic water in batch mode. In the second step, the optimization process was continued with Oued Souf water (south Algeria) where the influence of the current density and the area/volume ratio on the defluorination process was evaluated. The electrocoagulation process with aluminium bipolar electrodes permitted the defluorination of Sahara water without adding soluble salts to the treated water. The aluminium-fluoride weight ratio attained 17/1. © 1998 Elsevier Science Ltd. All rights reserved

Key words—defluorination, drinking water, electrocoagulation, aluminium bipolar electrodes

NOMENCLATURE

A	=	Total electrode area (m^2)
A_o	=	Electrode area (m^2)
A/V	=	Area/volume ratio ($m^2 m^{-3}$)
B	=	Constant (A·s)
C	=	The fluoride concentration ($kg m^{-3}$)
C_E	=	Current efficiency (%)
C_f	=	Final fluoride concentration ($kg m^{-3}$)
C_o	=	Initial fluoride concentration ($kg m^{-3}$)
d	=	Inter-electrode distance (cm)
F	=	Faraday number ($C mol^{-1}$)
i	=	Current density ($A m^{-2}$)
K_i	=	Kinetic constant (s^{-1})
m^*	=	Theoretical aluminium mass (kg)
m_e	=	Initial mass of the electrode before treatment (kg)
m_{exp}	=	Difference mass between initial and final masses (kg)
M_i	=	Aluminium molar mass (kg)
m_t	=	Final mass of the electrode after treatment (kg)
N	=	Cells number
n	=	Aluminium valence
NTU	=	Nephelometric Turbidity Units
pH _i	=	Initial pH
t	=	Experiment time (s)
T	=	Temperature ($^{\circ}C$)
t_N	=	Necessary time to decrease the fluoride concentration at 0.8 mg/l (s)
U_c	=	Electric potential (V)

INTRODUCTION

The chronic toxic effects of the excessive intake of fluoride or fluorosis are usually skeletal abnorm-

alities or damage. Dental fluorosis is characterized by an increasing molting of the teeth as the fluoride levels increase (Lynch, 1987; Mansson and Withford, 1990; Yam *et al.*, 1995).

It is generally agreed that the optimum fluoride concentration in water for the production of stony teeth of the children is about 1.0 mg/l. If fluoride concentration is substantially greater than 1.5 mg/l, dental fluorosis or molting enamel will result (Jinadasa *et al.*, 1988; Yam *et al.*, 1995).

Many countries have regions where the drinking water contains more than 1.0 mg/l, for example: U.S.A. (South California, Oakley and Idaho), Australia, South Africa, Kenya, India, China and north Africa (septentrional Sahara).

The septentrional Sahara people are exposed to excess of the fluoride in drinking water of the region. The percentage disease attaining Oued Souf people (South Algeria) is more than 80%. This situation incited different research teams to quantify fluoride in drinking water, fruits and vegetables of the regions having this problem (Lebza, 1986; Achour, 1990).

Earlier studies have applied different defluorination techniques to wastewater and/or natural water. The technique based on the precipitation with aluminium salt or calcium dihydroxyde was used previously (Borruf, 1934; Rabosky and Miller, 1974; Mazounie and Mouchet, 1984; Zhang and Liang, 1992), however, it was not efficient economically

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compared to other techniques (Bulusu and Nawlakhe, 1988; Diéye *et al.*, 1994). Indeed, the defluorination technique based on the adsorption phenomena was more efficient. The most usual adsorbents were activated alumina (Shoeman and Leach, 1987; Bulusu and Nawlakhe, 1990; Diéye *et al.*, 1994, Lee *et al.*, 1995) and calcium phosphate (Nawlakhe and Paramasvam, 1993; Saha, 1993; Abou Elela *et al.*, 1995). A new process based on electrochemical methods using an aluminium anode was developed (Ming *et al.*, 1983; Cheng, 1985; Prisyazhnyuk, 1992; Walsh and Mills, 1994). All teams worked with monopolar aluminium electrodes and met a passivation problem. The efficiency of this defluorination process was considerably reduced. To prevent the formation of the alumina layer on the anode, chloride salt was added to the solution (Ming *et al.*, 1983; Cheng, 1985) or an alternative current was used (Prisyazhnyuk, 1992).

Bipolar electrodes

A conductive plate placed between two electrodes having opposite charges without any electric connection will transform the two neutral sides of the plate to charged sides which have an opposite charge compared to the parallel beside the electrode (Fig. 1). This plate is commonly called the bipolar electrode. Thus, on the positive side it undergoes anodic reactions while on the negative side cathodic reactions were encountered. Industrial electrochemical reactors equipped with serial parallel bipolar electrodes where only the outer electrodes were connected to the electric generator presented more efficiency than the current processes. Indeed several advantages were observed such as more efficient current distribution and less important drop electric potential occurred with the electric connections. Nevertheless, the electrochemical processes are more expensive than chemical processes (Chaussard, 1991).

The defluorination of water by electrolysis processes using aluminium electrodes was described by Ming *et al.* (1983). The mechanism proposed was that the dissolution aluminium anode produces Al^{3+} ions which at appropriate pH are transformed, in a first step, to aluminium hydroxide $\text{Al}(\text{OH})_3$ and finally polymerized to $\text{Al}_n(\text{OH})_{3n}$ (equations 1), (4) and (5). The latter compound has strong affinity to fix the fluoride ions. The second hypothesis was that the Al^{3+} ions under the high fluoride concentration encountered near the anode may be induced to the formation of AlF_6^{3-} which could be transformed to insoluble salt Na_3AlF_6 by sodium ions (equations 2) and (3).

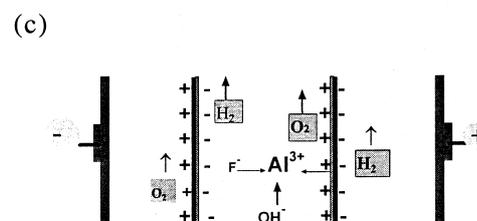
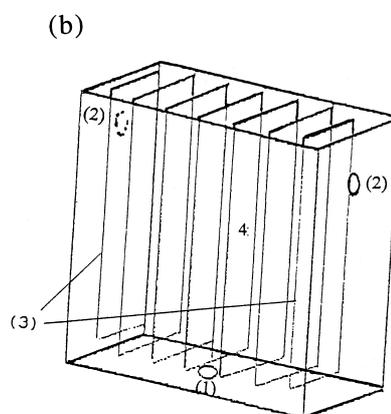
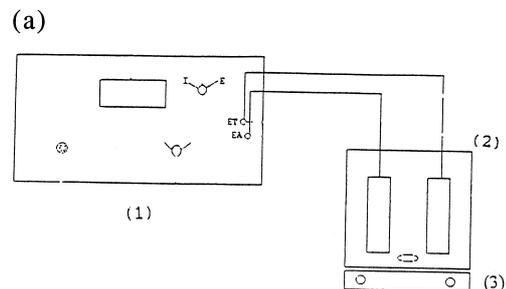
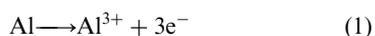
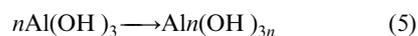
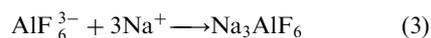
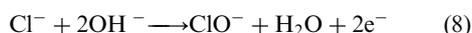
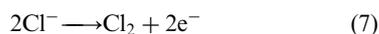


Fig. 1. Electrocoagulation functioning anodes and electrocoagulation cell. (a) Electrocoagulation apparatus: (1) Potentiostat *TACUSSEL PRT 20-2X*, (2) electrochemical reactor, (3) Magnetic stirrer. (b) Electrocoagulation cell: (1) leading tube, (2) deliver tube, (3) outer electrodes (4) internal electrode. (c) Schematic representation of the bipolar electrode.

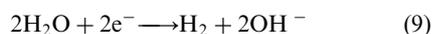


Other reactions may be also encountered in anodic compartment:

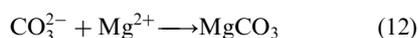




The main reaction encountered in cathodic compartment was as follows:



Other reactions may be observed at high pH (pH > 8) near the cathode and provoked the precipitation of the carbonate salt on this electrode (equations (10–12)). These later reactions are not interesting for the electrocoagulation process because they increased the ohmic resistance of the electrochemical cell.



The purpose of this work was to focus on an economical defluorination process based on the electrocoagulation using aluminium bipolar electrodes where different operator parameters were optimized.

MATERIALS AND METHODS

Materials

The experiments were achieved in a batch mode which is presented in the Fig. 1, the cell used is shown too. The fluorinated water enters by the leading tube (1) and gets out from the deliver tube (2).

The current density was maintained constant by means of a Tacussel PRT 20-2X potentiostat used in intensiostat mode. The aluminium electrode purity was about 99.5% and its dimensions were 125 mm × 60 mm. The dimensions of the outer electrodes in the cell were reduced to prevent an electric field dispersion. In case of the batch mode, magnetic stirring (40 rpm) was used to have a homogeneous solution in the electrochemical reactor.

Methods

The influence of different parameters during the defluorination experiments was studied by varying only one parameter. For each value of this parameter, the fluoride concentration was followed with time. In first step, the influence of parameters such as inter-electrode distance, fluoride concentration, temperature and the pH of the solution, were investigated and optimized with synthetic water in batch mode. In the second step, the optimization process was continued with Oued Souf water (south Algeria) where the influence of the current density and the area/volume ratio on the defluorination process was evaluated. The experiment was stopped when the fluoride concentration reached the concentration $C = 0.8$ mg/l, a value recommended by the World Health Organization

(W.H.O.) monograph 59 for tropical countries (Bell and Ludwig, 1970).

The current efficiency (C_E) was calculated using the equation:

$$C_E (\%) = \frac{1 - m_e}{m_t} \times 100 \quad (13)$$

where the theoretical mass (m_t) was obtained by using the Faraday relation. The experimental mass (m_e) was obtained by the electrode mass difference before and after electrocoagulation process. All the electrodes were weighed after having been soaked in an alkali solution (0.1 mol/l NaOH) during twenty seconds.

Chemical analysis

The fluoride analysis has been achieved using the ionometric method with specific fluoride electrode PF4L from Tacussel. To prevent interfering ions (Al^{3+} , Fe^{3+} , Cu^{2+} , Ca^{2+} , ...), TISAB buffer at pH = 5.5 was added to the samples. The calcium and magnesium analysis were achieved by means of Unicam-Philips SP19 atomic absorption. The sodium Na^+ and potassium K^+ concentrations were determined using Perkin Elmer Coleman 59 Ca flame photometry while the chloride concentration was obtained by specific chloride electrode with the ionometric method. The bicarbonate HCO_3^- concentrations were determined by the volumetric method (Rodier, 1984) using the chloride acid (HCl N/35). The X-rays spectrum was recorded by means of a Philips PW 1710 spectrometer.

RESULTS AND DISCUSSION

First, the aluminium electrode behavior was studied with synthetic solution having an average characteristic of the drinking water found in the Oued Souf region (see Table 1). The results presented in Table 2 obtained for different current densities show that a current efficiency is superior to 100% for the batch mode. This may be explained by the dissolution of aluminium as a result of pitting corrosion caused by a strong chloride concentration present in the treated solution (700 mg/l) which is erroneously calculated as dissolved aluminium by the electrochemical process. Damien (1992) has also observed a current efficiency greater than 100%. This suggests that there was no formation of passive film on the surface electrode and may explain the efficiency of the electrocoagulation process.

Second, the influence of the experimental parameters on the defluorination process was achieved with synthetic water in batch mode. This study was realized with monopolar electrodes. The first parameter was the inter-electrode distance (d) which was chosen in the range of 0.5 to 3 cm corresponding to the values used in the electrochemical industry (Damien, 1992). The results, presented in Fig. 2, show that the necessary time (t_N) to attain the flu-

Table 1. Characteristics of synthetic water

Ions	Ca^{2+}	Mg^{2+}	K^+	Na^+	Cl^-	HCO_3^-	F^-	SO_4^{2-}
Concentration (mg/l)	280	200	20	200	700	100	3	942

Table 2. Aluminium electrodes behavior during the electrocoagulation test at different current densities

i ($A\ m^{-2}$)	U_c (V)	m^* (mg)	m_{exp} (mg)	C_E (%)
3.30	1.04	24.36	26.50	109
8.33	1.25	28.10	30.00	106
17.39	1.64	32.00	33.00	103
25.00	2.00	68.41	72.00	105
40.00	2.4	45.02	55.00	122

oride concentration recommended by W.H.O. norms increased with the inter-electrode distance. An explanation of this may be that with a small inter-electrode distance, the gas which was blown in the cathode compartment induces a great floating phenomenon of the fluoroaluminium complexes formed on the anode electrode. This latter operation permitted further dissolution of the aluminium anode and increased the complex formation at the metal-solution interface (Sigg *et al.*, 1992). However, for short inter-electrode distances the current density becomes too high and can cause short circuit. Therefore, the experiment has been done with an inter-electrode distance of 2 cm, although the ohmic resistance will be higher. This distance is usually used by industrial electrochemical processes.

The Sahara waters of north Africa have variable fluoride concentrations. The fluoride concentration encountered was in the range of 0.1–10 mg/l. This interval was used to determine the influence of the fluoride concentration on the electrocoagulation process with aluminium electrodes. Therefore, the samples were prepared by using NaF salt dissolved in distilled water. The samples conductivity was

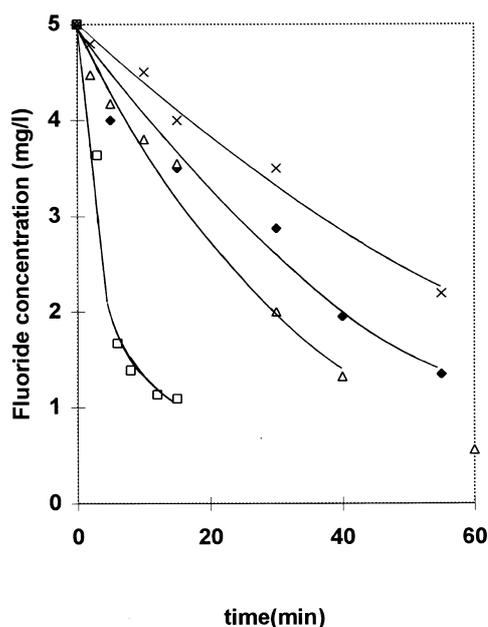


Fig. 2. Inter-electrode distance influence on the defluorination electrocoagulation process at $i = 3.12\ A\ m^{-2}$, $A/V = 6.4\ m^2\ m^{-3}$; (\square) $d = 0.5\ cm$, (\triangle) $d = 1\ cm$, (\bullet) $d = 2\ cm$, (\times) $d = 3\ cm$.

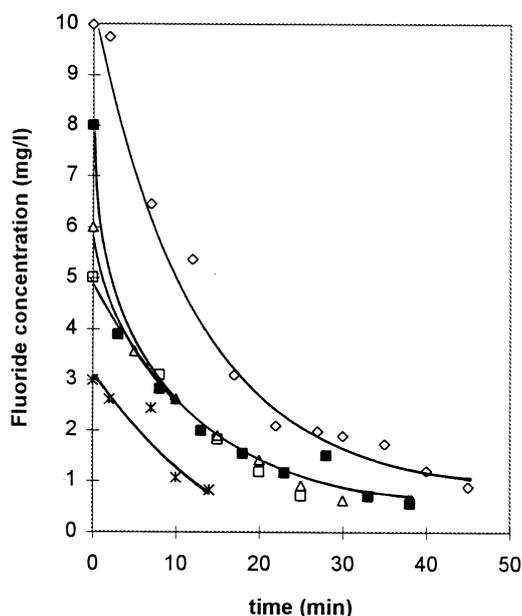


Fig. 3. Influence of the fluoride initial concentration on the electrocoagulation process at $i = 10.2\ A\ m^{-2}$, $d = 2\ cm$, $T = 20^\circ C$, (\diamond) $C = 10\ mg/l$, (\blacksquare) $C = 8\ mg/l$, (\triangle) $C = 6\ mg/l$, (\square) $C = 5\ mg/l$, (\times) $C = 3\ mg/l$.

between 40 and $80\ \mu S/cm$. The results presented in Fig. 3 show for a fixed pH and current density, that the concentration increase induces the time (t_N) increase. Also, an important decrease of the fluoride concentration appeared in the treated solution just after the first 10 min of operation, no matter the initial concentration. After a defluorination ratio of about 70%, the decrease continued slowly to reach a ratio of 90% at the end of the experiment.

The temperature of the Sahara groundwater varies between 20 and $55^\circ C$. In this study the temperature parameter was chosen in this range. The results, presented in the Fig. 4, indicate that the process efficiency decreases with increasing temperature. The reduction of the performance of the process may be explained, in part, by the fluoride desorption from aluminium hydroxide and, for another part, by the destruction of the fluoroaluminium complexes at high temperature (Mantell, 1945). To confirm the preliminary explanations, five samples were prepared with homogeneous solution initially treated by the process at $T = 20^\circ C$ and with a final fluoride concentration of $C_f = 0.19\ mg/l$. These samples were heated up to temperatures lying between 20 and $55^\circ C$. The samples were magnetically stirred during twenty minutes and filtered through filter paper. Then the fluoride concentrations of the filtered solution were determined. The reported results in Fig. 4b confirm that the fluoroaluminium complexes were not stable at high temperatures. It is therefore recommended to work at the ambient temperature for the defluorination

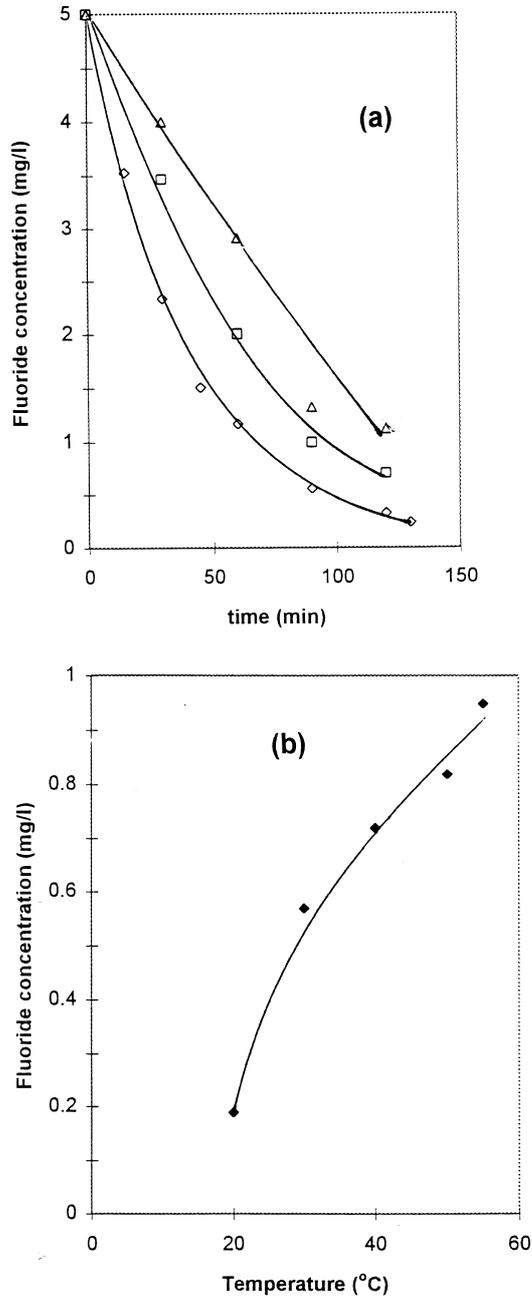


Fig. 4. Influence of the fluorinated water temperature on the electrocoagulation process and the stability of the fluoroaluminium complex. (a) Experimental conditions: $i = 3.12 \text{ A m}^{-2}$, $A/V = 6.4 \text{ m}^2 \text{ m}^{-3}$, $C = 5 \text{ mg/l}$, $d = 2 \text{ cm}$. (\diamond) $T = 17^\circ\text{C}$, (\square) $T = 40^\circ\text{C}$, (\triangle) $T = 50^\circ\text{C}$. (b) Effect of the temperature on the fluoroaluminium complex.

process by electrocoagulation with aluminium electrodes.

The influence of the pH on the defluorination was tested on the synthetic solution (distilled water + NaF salt) by simple addition of acid (HCl) or alkaline solution (NaOH). The studied range of the pH was 4 to 9. The results, reported in Fig. 5,

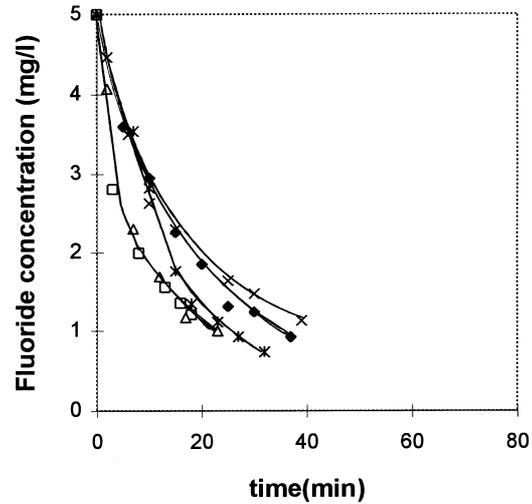


Fig. 5. Influence of the initial pH on the electrocoagulation process at $i = 9.1 \text{ A m}^{-2}$, $A/V = 6.4 \text{ m}^2 \text{ m}^{-3}$, $d = 2 \text{ cm}$, $T = 20^\circ\text{C}$. (\diamond) $\text{pH}_i = 4.4$, (\square) $\text{pH}_i = 5.2$, (\triangle) $\text{pH}_i = 5.7$, ($*$) $\text{pH}_i = 6.7$, (\times) $\text{pH}_i = 9.0$.

show that the defluorination process is more efficient for a pH ranging from 5 to 7.6. These results may be explained mainly by the strong presence of the hydroxyaluminium in this pH range which maximized the fluorohydroxide aluminium complex formation (Michard, 1981; Ming *et al.*, 1983; Sigg *et al.*, 1992). For all experiments whatever initial pH, the final pH of the solution converged to the value of 7.6. This value represents the pH where the fluoroaluminium complexes AlF_3 , AlOHF_3^- and $\text{Al}(\text{OH})_2\text{F}_2^-$ are strongly present inducing an efficient aluminium complexation by fluoride. Working at a pH above 8 led to a negligible complexation by fluoride rather than the hydroxide complexation of the aluminium (Michard, 1981). This result means that it was possible to use directly Sahara waters which have an initial pH between 5 and 7.6 and a strong mineralization. According to the chemical characteristics of Sahara waters there is no a need to increase the price of the treatment by the addition of chemical products (acid or base) in the electrocoagulation process, contrary to the processes based on the precipitation or the adsorption (Rabosky and Miller, 1974; Mazounie and Mouchet, 1984).

The defluorination process by electrocoagulation with aluminium was applied to the treatment of the Sahara water (Ain Boukhal region). The main characteristics of this water, presented in Table 3, shows a strong mineralization (2 g/l) with a hardness of 78 French degrees and high concentrations of Na^+ , SO_4^{2-} and Cl^- . The concentration of fluoride ions in this region was about 2.5 mg/l. In a preliminary study of the defluorination of the natural water, the intensity potential curve, where the applied potential was fixed between 1 and 20 V, was achieved. The current intensity measured for each potential indicate that the defluorination operation

Table 3. Characteristics of the Ain Boukhial water

Ions	Ca ²⁺	Mg ²⁺	K ⁺	Na ⁺	Cl ⁻	HCO ₃ ⁻	F ⁻	SO ₄ ²⁻	pH
Concentration (mg/l)	242	230	17.4	280	710	104	2.5	942	7.6

follows the Faraday law until a current density of 350 A m⁻²; the value usually applied in the industrial electrolysis processes.

The influence of the current density on the defluorination experiment was studied with two monopolar electrodes where fluoride concentration of the drinking water of Ain Boukhial was determined during the electrocoagulation experiments. The results presented in Fig. 6, show a steep decrease of the fluorine concentration just after the beginning of the experiments for all current densities. The experiments led to a fluorine elimination ratio of about 80%, *i.e.* final fluoride concentration in the range of 0.5 mg/l. With this drinking water, a fluoride elimination ratio of 68% could be attained; the concentration 0.8 mg/l recommended by the W.H.O. norms for tropical countries. The evolution of fluorine concentration with time can be expressed by the relation:

$$\frac{dC}{dt} = -K_i C \tag{14}$$

where *t*, *C* and *K_i* are the time, fluoride concentration and kinetic constant, respectively. The constant *K_i* depends on the current intensity and the temperature of the experiment. By simple integration, equation 14 can be rearranged as follows:

$$C(t) = C_o \cdot e^{-K_i t} \tag{15}$$

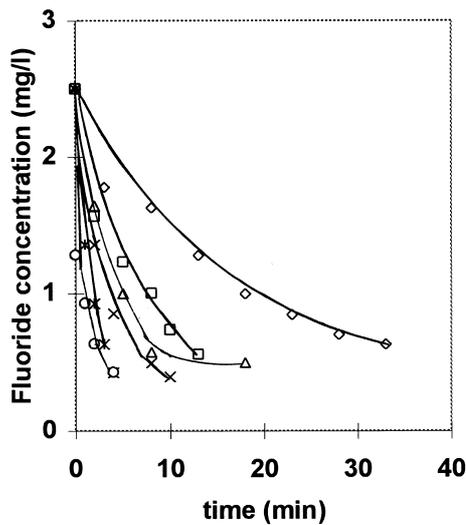


Fig. 6. Influence of the current density on the defluorination of drinking water of Ain Boukhail. *T* = 20°C, *A/V* = 6.9 m² m⁻³, *d* = 2 cm. (◇) *i* = 28.9 A m⁻², (□) *i* = 72.2 A m⁻², (△) *i* = 101.1 A m⁻², (×) *i* = 145 A m⁻², (*) *i* = 217 A m⁻², (○) *i* = 289 A m⁻².

Plotting $-\log C/C_o$ vs. *t* (Fig. 7) permitted to obtain a linear relation between these parameters for each current density used. This result confirms that the kinetic of the defluorination of the drinking water follows the exponential law with time. This equation has also been obtained by Ming *et al.* (1983). Therefore, the kinetic laws of the fluoride elimination for Ain Boukhial were of first order for the electrocoagulation process. Sigg *et al.* (1992) have suggested that the kinetics of fluoroaluminium complex formation in natural water are generally of second order. This is not the case for the electrocoagulation process where two phenomena contribute to the formation of fluoroaluminium complexes. The first phenomenon is the formation of these complexes with Al³⁺ ions on the electrodes, whilst the second phenomenon is the formation of the fluoroaluminium complexes by fluoride adsorption on aluminium hydroxide (Ming *et al.*, 1983; Cheng, 1985).

The principal purpose of this work was to reduce the fluorine concentration to the W.H.O.'s standards. Then, the necessary time *t_N* was determined for each current density used. The results presented in Fig. 8, show an important drop in the time *t_N* with an increase of the current density ranging from 9.1 to 200 A m⁻². For values greater than 200 A m⁻², the time *t_N* was practically independent of the current density. It appeared that the formation of the fluoroaluminium complexes was the lowest step of the process compared with the formation of Al³⁺ ions in the anodic compartment. This implies that the defluorination process by electrocoagulation was controlled by the formation of fluoroaluminium complexes and it is not necessary to work with a current density greater than an optimum value determined for a chosen area/volume (*A/V*) ratio.

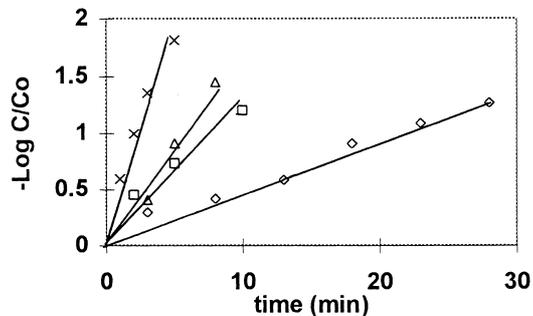


Fig. 7. Determination of the kinetic law of the defluorination of drinking water. *T* = 20°C, *A/V* = 6.9 m² m⁻³, *d* = 2 cm. (◇) *i* = 28.9 A m⁻², (□) *i* = 72.2 A m⁻², (△) *i* = 101.1 A m⁻², (×) *i* = 217 A m⁻².

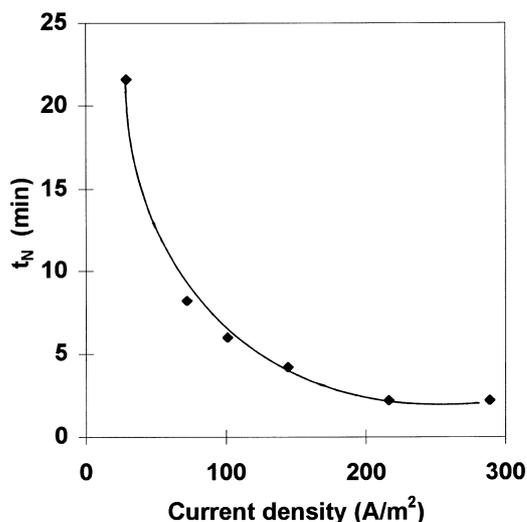


Fig. 8. Evolution of the necessary time (t_N) with the current density at $A/V = 6.9 \text{ m}^2 \text{ m}^{-3}$, $d = 2 \text{ cm}$, $T = 20^\circ \text{C}$.

The influence of the area/volume (A/V) ratio parameter on the evolution of the time t_N with the current density was achieved by using bipolar cells. This allowed an increase of the aluminium anode area. The results obtained (Fig. 9) show that the increase of the anode area, *i.e.* increase of the (A/V) parameter, induces a decrease of the necessary time t_N for a given current density. These results confirm the previous results obtained with the ratio $A/V = 6.9 \text{ m}^2 \text{ m}^{-3}$, *i.e.* the existence of an optimum current density for a fixed A/V ratio. Beyond the value of $28.7 \text{ m}^2 \text{ m}^{-3}$, the influence of this parameter on the necessary time t_N was practically insignificant. The optimum current density values deduced for different applied A/V ratios reported in Fig. 10, show a decrease of the optimum current density as the parameter A/V increases.

The necessary time t_N may be expressed by the equation based on the Faraday relation:

$$t_N = \frac{m \cdot n F}{M_i A_0 N} \quad (16)$$

This relation was established with a current efficiency ratio C_E equal to 100% as working hypothesis. equation 16 was transformed to:

$$t_N = \frac{B}{i} \quad (17)$$

where the B constant is dependent on the cell number used in the experiment.

Plotting the evolution of the necessary time (t_N) obtained experimentally against the inverse of the current density ($1/i$) for different A/V ratios, confirmed linear evolution of the time t_N against $1/i$ foreseen by equation 17 (results not shown).

As the excess of the fluoride ions mass in the solution was constant for all experiments and equal to 1.7 mg/l , the theoretical aluminium mass needed for

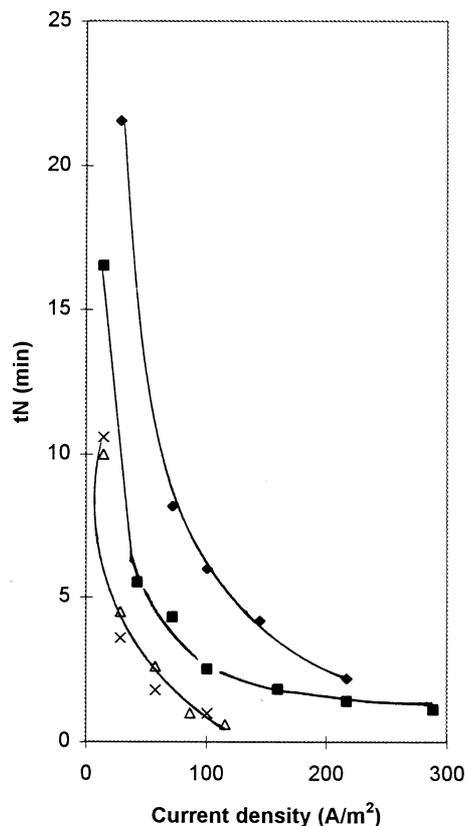


Fig. 9. Effect of the area/volume (A/V) ratio on the necessary time vs. current density. $d = 2 \text{ cm}$, $T = 20^\circ \text{C}$, (\blacklozenge) $A/V = 6.9 \text{ m}^2 \text{ m}^{-3}$, (\blacksquare) $A/V = 13.8 \text{ m}^2 \text{ m}^{-3}$, (\triangle) $A/V = 27.7 \text{ m}^2 \text{ m}^{-3}$, (\times) $A/V = 34.6 \text{ m}^2 \text{ m}^{-3}$.

the elimination of the fluoride ions was between $20\text{--}22 \text{ mg/l}$. The theoretical mass ratio (Al/F^-) is between $11.5\text{--}12$. This ratio was evaluated with a current efficiency ratio of 100% as a working hypothesis. The experiments achieved with $A/V = 13.8 \text{ m}^2 \text{ m}^{-3}$ permitted to deduce the experimental aluminium mass dissolved during the necessary time t_N at different current densities. The results show that the aluminum mass dissolved by the electrochemical process was practically constant (about $30 \text{ mg}/\text{A}_0$) and the experiment mass ratio Al/F^- was between $16\text{--}17.5$. This result is in agreement with

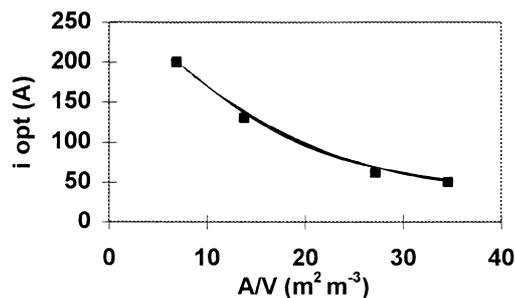


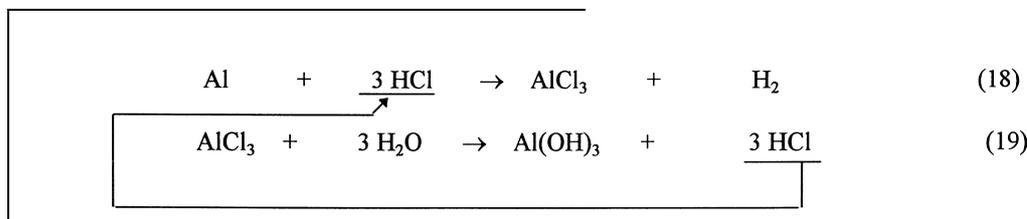
Fig. 10. Optimum current density vs. the area/volume ratio.

those of Ming *et al.* (1983). The experiment's current efficiency ratios lie between 130–140%, as obtained previously with synthetic water.

This difference in mass may be explained by the "corrosion Pitting" phenomenon which caused holes

decrease of the chloride ions. This confirms the corrosion of the aluminium anode by the chlorides.

The mechanism suggested for the aluminium metallic hydrolysis is as below:



and led practically to a metallic aluminum loss on the electrode surface. This mass was erroneously calculated as the aluminum dissolved by the electrochemical process.

The evolution of the Boukhial water characteristics after electrocoagulation treatment was determined. The chemical composition of the Boukhial water before and after treatment was reported in Table 4. The solution conductivity did not vary during the experiments. As far as the mineralization is conserved, a slight decrease (5%) was observed. This value is not in agreement with results of Prisyazhnyuk (1992), who has obtained a decrease in mineralization of about 50%. This high ratio was obtained for water with 200 mg in mineralization. Boukhial's water has a mineralization 10 times more important than the water used by Prisyazhnyuk (1992). So the mineralization loss in both cases was practically similar (about 100 mg/l). It is important to note that the remaining mineralization (1.9 g/l) of the treated water is too high compared to the W.H.O. requirements for drinking water.

The mineral ions eliminated during electrocoagulation experiments were combined with fluoro-complex and insoluble compounds. These were later added with the aluminium hydroxide precipitate and produced a sludge during the electrocoagulation treatment. The analysis of this sludge using X-rays, showed the presence of an amorphous aluminium compound (results not shown). The chloride ion analysis before and after the electrocoagulation experiment shows a slight

For all experiments with synthetic and Boukhial's waters the pH decreases from the initial pH to 7.6. This later value corresponds to the minimum pH which gave aluminium hydroxide and cryolite (Ming *et al.*, 1983; Cheng, 1985). These results confirm those obtained previously by other research teams (Ming *et al.*, 1983; Cheng, 1985; Prisyazhnyuk, 1992).

The efficiency and the feasibility of the electrocoagulation process is dependent on the aluminium concentration after the electrochemical treatment. The analysis of this heavy metal before and after electrocoagulation (Table 4), showed that the dissolved aluminium concentration decreased after the treatment with the electrochemical process. This process did not appear dangerous for the contamination of the drinking water. Nevertheless, an increase in the turbidity parameter of the treated water was observed. Values of the turbidity of the order of 122 NTU were attained after defluorination of Boukhial's water with this process.

CONCLUSION

The electrocoagulation process with bipolar aluminium electrodes allowed the defluorination of drinking water from southern Algeria. The kinetic of the defluorination of the drinking water follows an exponential law with the time. The use of bipolar electrodes induces a reduction of the necessary time of the treatment and an increase of the anode area. The studies of the influence of different experimental parameters permitted to obtain the optimum conditions in batch mode. The mass ratio Al/F⁻ obtained with the bipolar electrodes system is of the same order as the ratio previously determined with a monopolar electrodes system. The determination of the characteristics of the water before and after the treatment has confirmed the metallic hydrolysis phenomenon for the couple: aluminium-chloride. No contamination of the natural water by aluminium during electrocoagulation treatment was observed.

The results obtained show that the aluminium bipolar electrodes system is an interesting process

Table 4. Evolution of the characteristics of the Ain Boukhial water during the electrocoagulation treatment

Ions	Concentration before treatment (mg/l)	Concentration after treatment (mg/l)
Ca ²⁺	242	239
Mg ²⁺	230	228
Cl ⁻	710	686
Na ⁺	280	264
K ⁺	17.4	17.1
SO ₄ ²⁻	632	586
Al ³⁺	0.043	0.001
F ⁻	2.5	0.5

for the defluorination of the Sahara waters and could also be utilized to the defluorination of industrial waste waters.

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