European Journal of Applied Sciences – Vol. 10, No. 4
Publication Date: August 25, 2022
DOI:10.14738/aivp.104.12671.
Sedou, M., Mande, S. A., Sanou, Y., & Arouna, K. (2022). Fluoride Removal in Synthetic Drinking Water by Electrocoagulation Using Aluminum Electrodes. European Journal of Applied Sciences, 10(4). 429-438.

Fluoride Removal in Synthetic Drinking Water by Electrocoagulation Using Aluminum Electrodes

Moudassirou Sedou
Laboratory of Water Resources and Environmental Engineering, University of Kara. Faculty of Sciences and Technics, B.P. 404, Kara-Togo

Seyf-Laye Alfa-Sika Mande
Laboratory of Water Resources and Environmental Engineering, University of Kara. Faculty of Sciences and Technics, B.P. 404, Kara-Togo
Laboratory of Applied Hydrology and Environment University of Lome, BP. 1515, Togo
Beijing Key Laboratory of Water Resources & Environmental Engineering, China University of Geosciences (Beijing), Beijing 100083, P.R. China

Yacouba Sanou
Laboratory of Water Resources and Environmental Engineering, University of Kara. Faculty of Sciences and Technics, B.P. 404, Kara-Togo
Laboratory of Analytical, Environmental and Bio-Organic Chemistry, University Joseph KI-ZERBO UFR/SEA, 03 BP 7021 Ouagadougou 03, Burkina Faso

Kadidja Arouna
Laboratory of Water Resources and Environmental Engineering, University of Kara. Faculty of Sciences and Technics, B.P. 404, Kara-Togo

ABSTRACT

Fluoride is recognized as an essential constituent in the human diet. Low fluoride concentration could prevent dental problem while higher fluoride concentration will cause dental and skeletal fluorosis. This study aimed to remove fluoride in synthetic drinking water by electrochemical system. It had been performed at laboratory scale using fluoride synthetic water. The process consisted to optimize the fluoride removal in drinking water using the Aluminum electrodes. The effects of operating conditions such as the initial ion fluoride concentration, current density or intensity, contact time, and NaCl amount were studied. Experimental results showed that with the initial concentration of 15 mg/L using the optimal intensity (1.5 A), an admitted residual concentration (1.5 mg/L) was observed during 45 min of electrolysis while maximum fluoride removal percentage of 100 % was achieved for 60 min. The variation of energy consumption from 1.18 to 22.35

Wh showed that the electrocoagulation can be applied using photovoltaic energy for fluoride treatment in drinking water.

Keywords: Aluminum, electrocoagulation, fluoride, removal efficiency, water treatment.

INTRODUCTION

Drinking water contamination by fluoride is a threat for human health. His presence in drinking water originate especially from anthropogenic sources in addition to the dissolution of minerals in rocks and sediments. Nearby the mining sites and industrialized areas, the concentration values measured in water bodies for fluoride (1 - 4ppm) (Tanouayi et al., 2016) are extremely higher than the values about 1.5 ppm (WHO, 2011). The mining areas of Hahotoé-Kpogamé (phosphorite ore) contain high levels of fluoride which has induced illness related to fluorosis among people living in the vicinity of mining and processing sites (Melila, 2013; Ouro-Sama et al., 2014; Tanouayi et al., 2016). However, recent studies revealed a high prevalence of fluorosis in Hahotoé-Kpogamé mining areas about 45 % (Melila, 2013). Moreover, there is no studies dealing with fluoride remediation in drinking water in Togo, a developing country with several priority areas. Thus, it appears essential to find a low-cost effective method for fluoride removal from drinking water sources in Togo.

Chemical coagulation-based treatment is the most common approach for treatment in drinking water production (WHO, 2011). A range of methods including chemical coagulation has been reported especially for the removal of arsenic and fluoride from aqueous solutions, these methods being mainly reduction, ion-exchange, electrodialysis, electrochemical precipitation, evaporation, solvent extraction, reverse osmosis, chemical precipitation and adsorption (Alfa-Sika Mande et al., 2009; Harisha et al., 2010; Jadhav et al., 2015; Ahoulé et al., 2015, WHO, 2011). Most of these methods are not cost-effective and suffers drawbacks such as disposal of the residual sludge which poses further problems for the environment (Jadhav et al., 2015; Shan et al., 2019). One of these techniques, electrocoagulation, appears to provide one of the most effective, low cost methods because of its simplicity of operation, easy handling and no risk of dangerous by-products (Guzmán et al., 2016; Shan et al., 2019). Electrocoagulation (EC) is the process using "sacrificial" anodes to form an active coagulant which is used to remove pollutants by precipitation and flotation in situ. EC is a complex electrochemical process, which includes chemical and physical processes involving many surface phenomena. The technology sits at the intersection of three fundamental technologies: electrochemistry, coagulation, and flotation (Holt et al. 2004).When a potential is applied to form an external power source, the anode material will undergo oxidation, while the cathode will experience reduction or reductive deposition of elemental metals. The aluminum electrodes generated Al^{3+}(aq) ions will immediately undergo further spontaneous reactions to produce the corresponding hydroxides and/or polyhydroxydes. These hydroxide compounds have a high affinity for dispersed particles as well as counter ions to cause coagulation (Zhu and al., 2007). It is considered to be a complex process with a multitude of synergistic mechanisms contributing to the treatment of pollution. (Holt et al., 2004) were able to identify three categories of mechanisms in electrocoagulation: electrochemical phenomena, coagulation and hydrodynamics.

The aim of this study is to decrease substantially the fluoride concentrations in synthetic drinking water by electrocoagulation using aluminum electrodes. The influence of key

parameters such as, current density, initial concentration, electrolysis time and the Sodium Chloride (NaCl) are investigated using this type of electrodes.

MATERIALS AND METHODS

Experimental device of electrocoagulation

The defluoridation of drinking water is studied using the electrocoagulation system with aluminium electrode. The figure 1 show the experimental setup of electrocoagulation (EC) study. The electrocoagulation unit consisted of a 0.4 L electrochemical reactor with Al/Al electrodes. The distance between electrodes set was 1 cm. The anode and cathode were both flat aluminium electrodes of rectangular shape (15 cm × 4.7 cm × 1 mm). They were vertically centred between the bottom of the reactor and the liquid level.

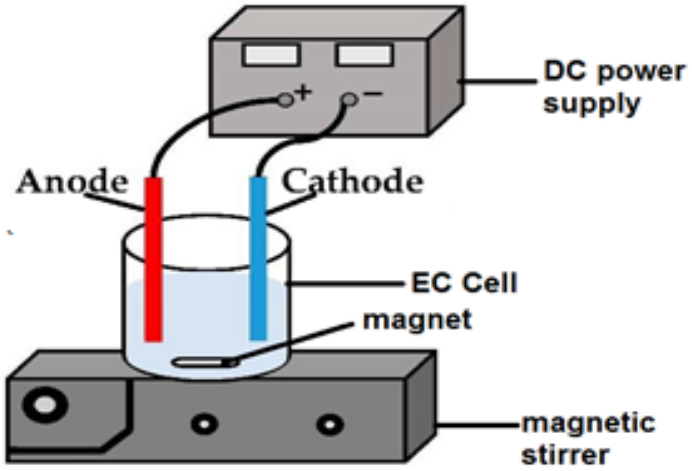


Figure 1: Experimental device of EC reactor at Laboratory Scale.

Fluoride removal experiments

Desired concentrations of fluoride solutions were obtained by dissolving a weight of sodium fluoride into distilled water. The conductivity of the sample water was varied by adding sodium chloride (NaCl). The electrochemical removal of fluoride was carried out in an electrolytic cell of 400 mL volume. Two electrodes (anode and cathode) were positioned vertically and parallel to each other. At the beginning of each run, 350 mL of synthetic fluoride wastewater (11.7 cm of height) was fed in the reactor and 0.5 g/L NaCl solution as electrolyte was added to increase the conductivity of solution. The intensity of current was adjusted from 0.75 A, 1 A to 1.5 A, corresponding to a current density between 13.64 and 27.28 mA/cm^2 using a digital DC power supply. Fluoride removal experiments were carried out with Aluminum electrodes electrode as anode and cathode. The electrolysis time was performed during 60 min and the treated solution was collected each 10 minutes. The electrocoagulated water samples were then filtered to remove the flocs generated during the electrocoagulation process to be analyzed using UV-Vis spectrophotometer (DIV-UV5600).

Energy consumption (E) expressed in Wh was calculated as follow:
E = Voltage (V) x Current intensity (A) x Time (h) (6)

RESULTS AND DISCUSSION

Effect of current density

The Current density determines not only the coagulant dosage rate, but also the production rate and bubble size (Holt et al. 2004). Thus, this parameter should have a significant impact on the pollutant removal efficiencies. To study the effect of current density and charge on removal efficiency, a series of experiments were performed on solutions containing a constant charge of pollutants with current intensities of 0.75 A, 1 A and 1.5 A corresponding to a current density of 13.64, 18.18 and 27.28 mA/cm^2.

The figure 2 shows that all the intensities used in this study made it possible to obtain residual concentrations under the WHO's thresholds for all the series of concentrations of the study. Better results have been obtained using the intensity of 1.5 A which gives in 10 min a residual concentration lower than 1.5 mg/L with a rate of 70% removal using the initial concentration of 5 mg/L. By applying intensities of 1 A and 0.75 A, the normal residual concentration (<1.5 mg/L) was obtained respectively in 40 and 50 min of electrolysis with this rate of 70%. The increase in the current density stimulates the dissolution of the anode which in turn increases the amount of the coagulant **(**Baya Palahouane, 2014). This increase of current density provide better removal of pollutant. Removal efficiency depends on the quantity of Al ions. The amount of Al ion dissolved in the EC process is theoretically calculated according to the Faraday law, which shows the relation between current density and the quantity of dissolved material

$$m = \frac{I\,t\,M}{nF} \qquad (7)$$

With

m: mass of dissolved metal or gas formed (g).
I: intensity of the impressed current (A).
t: duration of electrolysis(s).
M: molecular weight of the element considered (g.mol-1).
F: Faraday's constant (96500 C.mol-1).
n: number of electrons involved in the reaction considered

In the study of (Emamjomeh and Sivakumar, 2006), the higher intensity of their study (2.5 A) resulted in faster removal producing more coagulant. But, they stipulate that not only at such as an intensity there is an excessive concentration of the coagulant (Al^{3+}). This excess is a source of pollution for the drinking water and also a high intensity is not economical in terms of energy. In our study with similar results, beyond 1.5A; a very great dissolution of the anode is observed in a relatively short electrolysis time. It was show that the fluoride removal efficiency increased quickly by increasing the applied current.

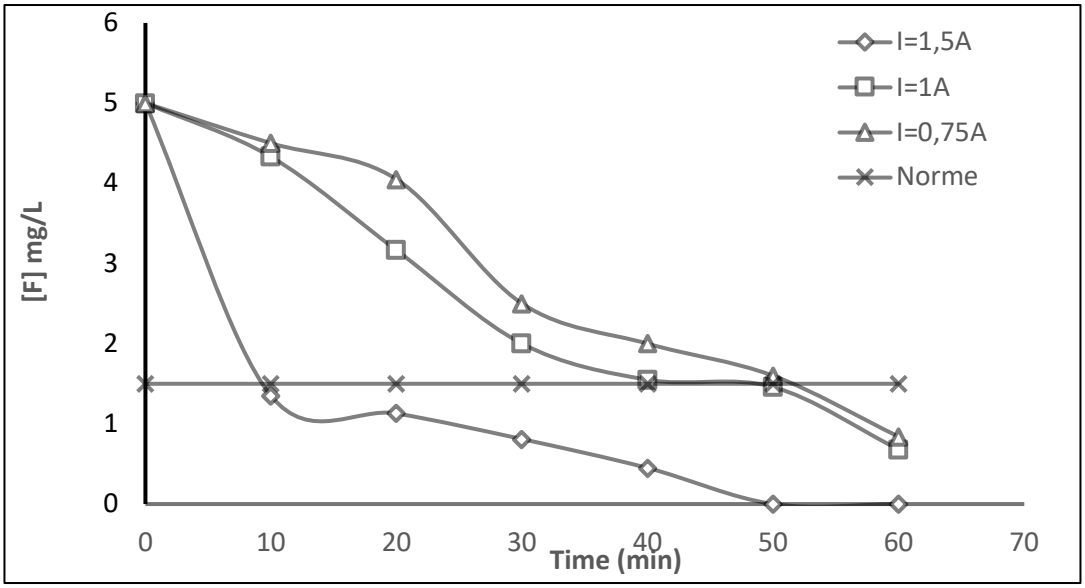


Figure 2: Effect of Current density on removal efficiency with C_0 = 5 mg/L; pH= 7.68; V=350 mL and [NaCl]= 0.5 g/L.

Effect of initial fluoride concentration

In order to examine the effect of the initial concentration on the fluoride removal, several solutions containing increasing concentrations (5-10-15 mg/L) of fluoride were treated and the residual concentrations were determined at intervals of electrolysis time. Figures 3 shows the evolution of the fluoride removal with the initial concentration by the aluminium electrodes. It appears that the elimination rate decreased with increasing of the initial concentration. Results show that the threshold for fluoride drinking in water is observed respectively at 10, 30 and 47 min for 5, 10 and 15 mg/L.

According figure 3, it observed that for higher initial fluoride concentration longer time is needed for removal of fluoride than compared to lower initial fluoride concentration. Higher concentration of fluoride was reduced significantly in relatively less time than lower concentration. This can be explained by theory of dilute solutions. In dilute solutions, formation of the diffusion layer has no effect on rate of diffusion or migration of metal ions to the electrode surface (Battula et al., 2014). Most of 63-95% of fluoride was removed in the first 20- 40 minutes as shown in Figure 4.

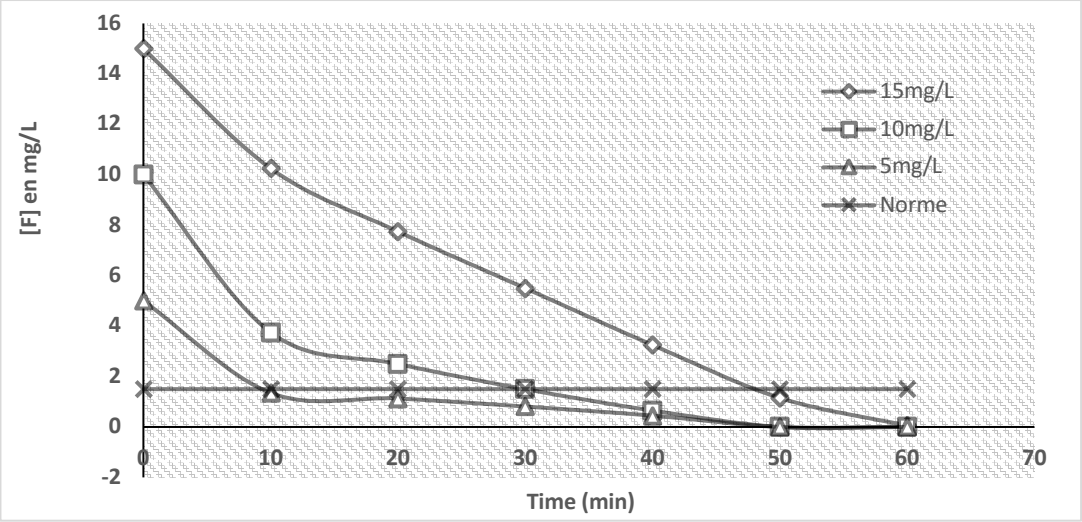


Figure 3: Effect of initial fluoride concentration on removal efficiency using I= 1.5 A; pH= 7.68; 0.5 g/L NaCl, and V=350 mL.

Effect of electrolysis time

The duration of electrolysis is an important functional parameter in the EC process. Longer operation time leads to more energy consumption and less treatment, and therefore, in order to achieve a cost-effective treatment these parameters should be optimized (Malakootian et al., 2010). Therefore, the effect of operation time on fluoride removal from drinking water was evaluated.

As indicated in figure 4, the fluoride removal significantly increases with increased electrolysis time until 20 min (fluoride removal > 63, 75 and 77% respectively for 15; 10 and 5 mg/L) and after this time a significant increase is not seen in efficiency in 50 min and in 60 min. As time of electrolysis progresses, because of anode oxidation, metal decomposition increases as well and a higher quantity of coagulants accumulate inside the EC cell, which corresponds to more fluoride removal and is attributed to the high absorbing capacity of aluminium coagulants produced in situ for fluoride ions. According to theory and based on Faraday's law (Eq. 7), electrolysis time is effective on the quantity of Al ions released inside the EC cell by using Al anode electrodes and an increase in electrolysis time leads to an increase Al ion release in the cell and, on the other hand, this leads to increased operation cost and more energy consumption according to formula (6) (Emamjomeh and Sivakumar, 2009) It show that the fluoride removal increased significantly upon the time. In the same direction as the current density, when the time of electrolysis increases the elimination becomes better. But in this, the cost of the process must be taken into account (increase in energy and dissolution of the electrode) why it necessary to optimize this parameter. In this study, according to the performance in different electrolysis times, a time of 40 min was reported as the optimum time but to reach the WHO concentration threshold, the 50 min time is adapted with the residual concentrations 0.67 mg/L using initial fluoride concentration of 15 mg/L.

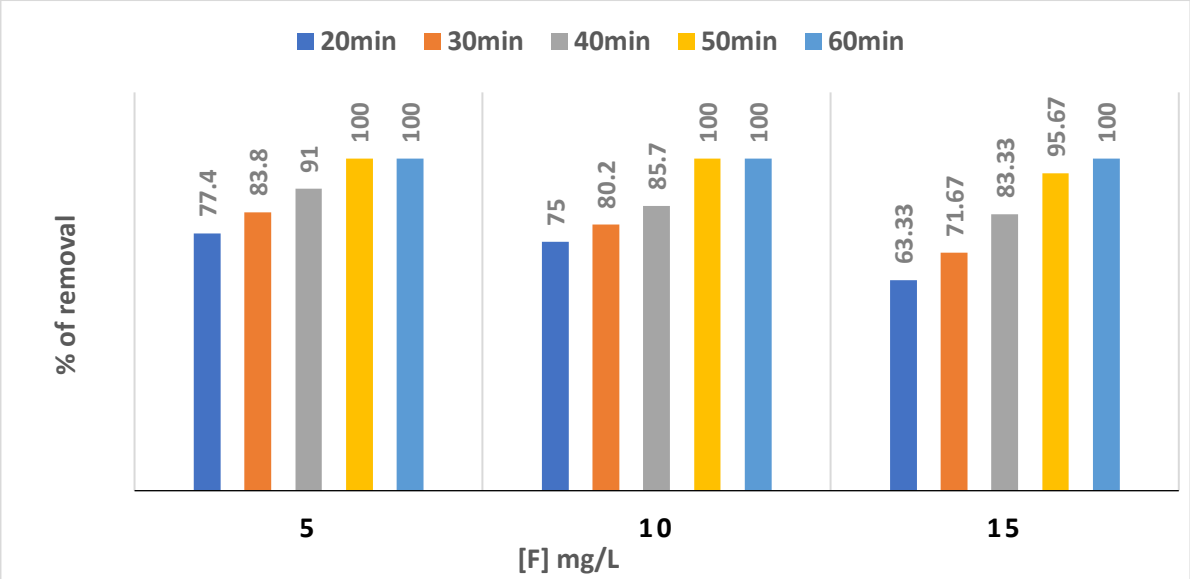


Figure 4: Effect of electrolysis time on removal efficiency using I= 1.5 A; pH= 7.68; 0.5 g/L NaCl, and V=350 mL.

Effect of NaCl amount

The addition of an electrolyte increases the electrical conductivity of the medium and reduces its resistance to ensure better dissolution of the anodes. It observed a higher current density when using NaCl electrolyte. This is in the same way according to the study of (Merzouk et al., 2009) in which the addition of NaCl increases the flow of current in the EC cell, which leads to an overconsumption of the electrodes under the effect of undesirable corrosion. As shown in figure 5, the NaCl electrolyte increases the fluoride removal. At the end of the 60 min, 100% of removal was obtained using NaCl electrolyte when it was 54.66% for without NaCl.

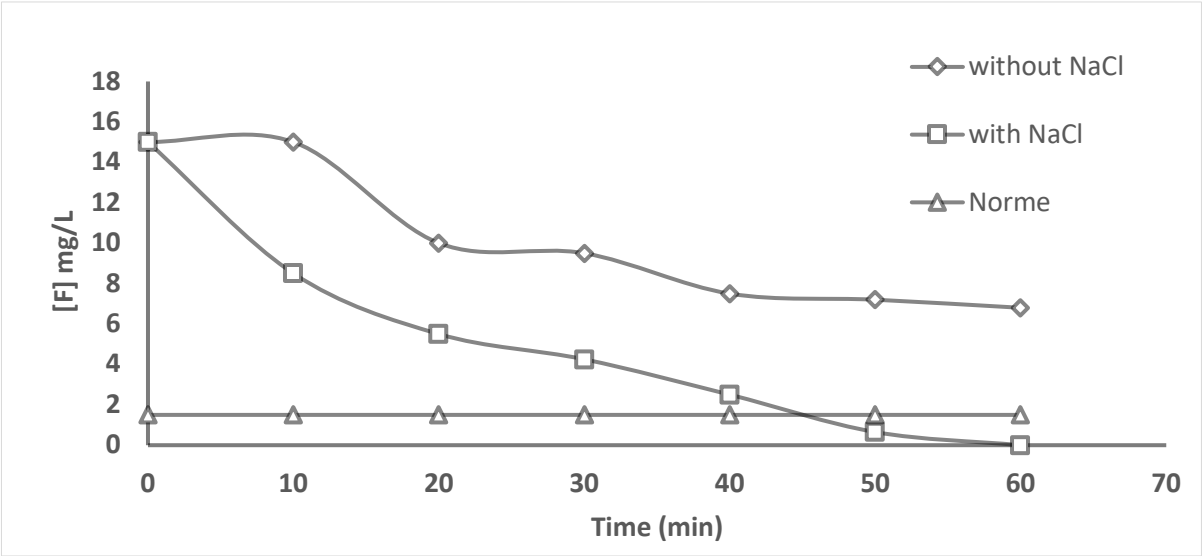


Figure 5: Effect of NaCl on removal efficiency using I = 1.5 A; pH= 7.68; 0.5 g/L NaCl, C_0=15 mg/L and V=350 mL.

Energy consumption

One of the most important parameters that must be determined to evaluate a method of the treatment is the cost. The operating cost includes materials cost (mainly electrodes), operating cost (mainly electrical energy) and other cost items such as labour, maintenance, sludge dewatering, and disposal (Baya Palahouane, 2014). In this study, energy and electrode material costs have been taken into account as major cost items. Energy consumption is a very important parameter in industry and can be calculated as shown the Equation 5. The energy consumption depends on electrolysis time: the increase in electrolysis time leads to energy consumption increase. As shown in Figure 6 when the current density was increased from 13.64 to 27.28 mA/cm^2, the fluoride removal was raised consequently from 86.67%, to 100% for 15 mg/L, whereas the Energy consumption varied from 1.18 to 22.35 Wh. The observed results involve that EC process could be an appropriate treatment for fluoride since it provides high removal efficiency of pollutant with moderate energy consumption.

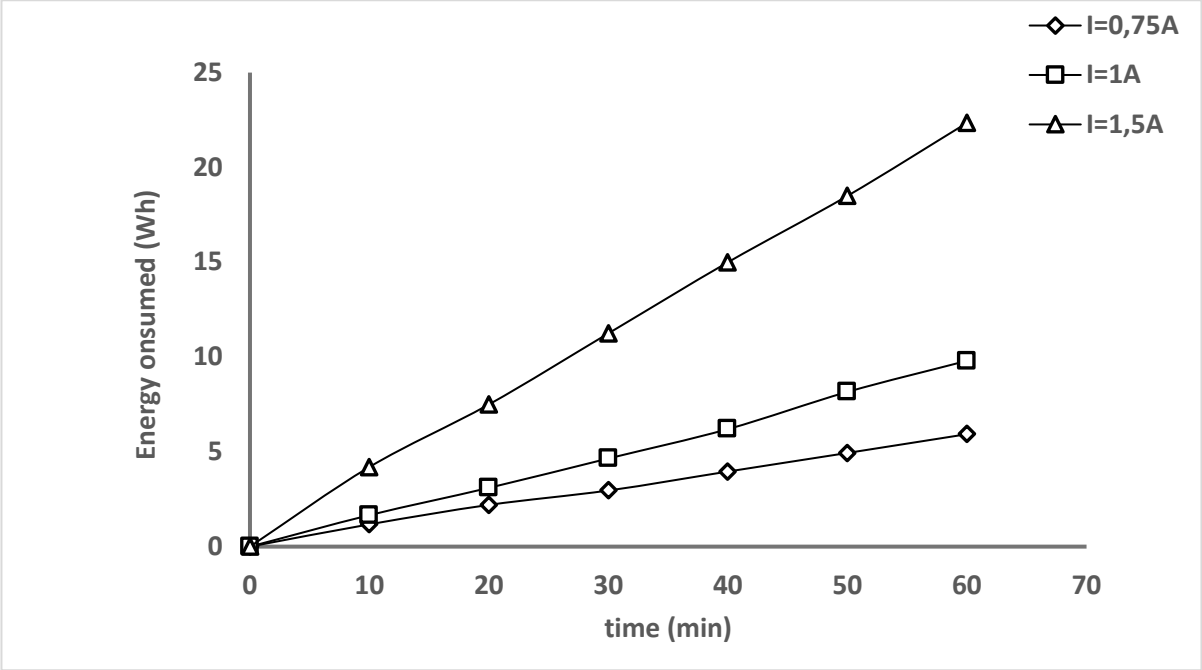


Figure 6: Energy consumption using 0.5 g/L NaCl, Co=15mg/L and V=350 mL.

Mechanisms of fluoride removal by EC

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. The electrochemical reactions at the electrodes can be summarized as follows while the electrolytic dissolution of Al anodes by oxidation in water produces aqueous Al^{3+}species (Eq. (1)) (Yousuf A. Mollah et al., 2000):

Anode: $Al_{(s)} \rightarrow Al^{3+} + 3e^{-}$ (1)

Cathode: $H_2O + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}$ (2)

The H_2 bubbles float and hence drive the flotation process. The Al^{3+} ions further react as shown in Eq. (3) to form a solid $\mathrm{Al}(OH)_{3(s)}$precipitate: $Al^{3+} + 3H_2O \rightarrow \mathrm{Al}(OH)_{3(s)} + 3H^+$ (3)

It had also distinguished coprecipitation and adsorption on the electrode surface, denoted "attachment", from mechanisms involving the bulk. In summary, three mechanisms have been reported to compete (Zhu and al., 2007):

- Adsorption step on $\mathrm{Al(OH)_3}$ particles:

$Al_n(\mathrm{OH})_{3n} + m\ F^- \rightarrow Al_nF_m(\mathrm{OH})_{3n-m} + m\ OH^-$ (4)

- Precipitation step:

$nAl^{3+} + (3\mathrm{n} - \mathrm{m})\mathrm{OH}^- + m\ F^- \rightarrow Al_nF_m(\mathrm{OH})_{3n-m}$ (5)

- Fluoride attachment to the electrodes.

CONCLUSIONS

The current study demonstrated the efficiency of electrocoagulation method on the fluoride removal. The influence of different parameters including the current intensity, the initial fluoride concentration and the electrolyse time was found to be very significant on the removal of fluoride. Additionally, it was seen from the experiments that 10 min EC was enough for the reaching the standard limits to the destined drinking water for fluoride when using 5 mg/L as initial concentration with 27.28 mA/cm^2. According to this time consequently, electrical energy consumption of 4.47 Wh was necessary to remove fluoride under the threshold fixed by WHO Guidelines. Experimental results show that the electrocoagulation process can be applied using photovoltaic energy for fluoride treatment in drinking water. Furthermore coupling this process with photovoltaic energy will help for this cost effective implementation within rural areas of sunny and developing country such as Togo.

References

Ahoulé, D.G., Lalanne, F., Mendret, J., Brosillon, S., Maïga, A.H., 2015. Arsenic in African Waters: A Review. Water. Air. Soil Pollut. 226, 302. *https://doi.org/10.1007/s11270-015-2558-4*

Alfa-Sika Mande, S.-L., Liu, M., Liu, F., Chen, H., 2009. Kinetics of the Adsorption of Chromium(VI) Ions from Water by Vegetable Precursors Treated with Sulphuric Acid. Adsorpt. Sci. Technol. 27, 965–974. *https://doi.org/10.1260/0263-6174.27.10.965*

Guzmán, A., Nava, J.L., Coreño, O., Rodríguez, I., Gutiérrez, S., 2016. Arsenic and fluoride removal from groundwater by electrocoagulation using a continuous filter-press reactor. Chemosphere 144, 2113–2120. https://doi.org/10.1016/j.chemosphere.2015.10.108

Harisha, R.S., Hosamani, K.M., Keri, R.S., Nataraj, S.K., Aminabhavi, T.M., 2010. Arsenic removal from drinking water using thin film composite nanofiltration membrane. Desalination 252, 75–80. https://doi.org/10.1016/j.desal.2009.10.022

Jadhav, S.V., Bringas, E., Yadav, G.D., Rathod, V.K., Ortiz, I., Marathe, K.V., 2015. Arsenic and fluoride contaminated groundwaters: A review of current technologies for contaminants removal. J. Environ. Manage. 162, 306–325. https://doi.org/10.1016/j.jenvman.2015.07.020

J. Zhu, H. Zhao, et J. Ni, « Fluoride distribution in electrocoagulation defluoridation process », Sep. Purif. Technol., vol. 56, no 2, p. 184-191, août 2007, doi: 10.1016/j.seppur.2007.01.030.

Malakootian M, Mansoorian HJ, Moosazadeh M (2010) Performance evaluation of electrocoagulation process using iron-rod electrodes for removing hardness from drinking water. Desalination 255:67–71. https ://doi.org/10.1016/j.desal .2010.01.01.

Melila, M., 2013. Bioaccumulation du cadmium, du plomb et du fluor et conséquences biologiques sur l'Homme et Sphyraena barracuida (poisson marin) dans la zone de traitement des phosphates au Togo. Thèse de Doctorat, Université de Lomé, Togo.

Merzouk B., Gourich B., Sekki A., Madani K., Chibane M. Removal turbidity and separation of heavy metals using electrocoagulation–electroflotation technique: a case study. J. Hazard. Mater. 2009; 164: 215–222.

M. Emamjomeh, M. Sivakumar, Fluorideremoval by a continuous flow electrocoagulation reactor, Journal of Environmental Management 90 (2009) 1204–1212.

M. M. Emamjomeh et M. Sivakumar, « An empirical model for defluoridation by batch monopolar electrocoagulation/flotation (ECF) process », J. Hazard. Mater., vol. 131, no 1-3, p. 118-125, 2006.

M.Y.A. Mollah, R. Schennach, J.R. Parga, D.L. Cocke, Electrocoagulation (EC) — science and applications, J. Hazard. Mater. 84 (2001) 29–41

Ouro-Sama, K., Solitoke, H.D., Gnandi, K., Afiademanyo, K.M., Bowessidjaou, E.J., 2014. Évaluation et risques sanitaires de la bioaccumulation de métaux lourds chez des espèces halieutiques du système lagunaire togolais. VertigO - Rev. Électronique En Sci. Environ. https://doi.org/10.4000/vertigo.15093

Palahouane B., Drouiche N., Aoudj S., Bensadok K. Cost-effective electrocoagulation process for the remediation of fluoride from pretreated photovoltaic wastewater. J. Ind. Eng. Chem. 2015; 22:127–131.

Holt, P.K., Barton, G.W. and Mitchell, C.A. (2005) The Future for Electrocoagulation as a Localised Water Treatment Technology. Chemosphere, 59, 355-367. https://doi.org/10.1016/j.chemosphere.2004.10.023

Shan, Y., Mehta, P., Perera, D., Varela, Y., 2019. Cost and Efficiency of Arsenic Removal from Groundwater: A Review (No. UNU-INWEH Report Series, Issue 05). United Nations University-Institute for Water, Environment and Health, Hamilton, Canada.

Tanouayi, G., Gnandi, K., Ouro-Sama, K., Aduayi-Akue, A.A., Ahoudi, H., Nyametso, Y., Solitoke, H.D., 2016. Distribution of Fluoride in the Phosphorite Mining Area of Hahotoe–Kpogame (Togo). J. Health Pollut. 6, 84–94. https://doi.org/10.5696/2156-9614-6.10.84

WHO, 2011. Guidelines for Drinking-water Quality, 4th ed. ed.

Sailaja Kumari Battula, Jyostna Cheukuri, Raman N. V. V. S. S, Himabindu .V, Bhagawan. D. Effective removal of fluoride from ground water using electro-coagulation. Int. Journal of Engineering Research and Applications. 2014, pp.439-445.