

# Simultaneous Removal Fluoride and Nitrate from Water in a Batch Reactor Using Al and Fe anodes and Cu, Steel and Graphite Cathodes

Razieh Ashoori<sup>1</sup>, MSc;  
 Mohammad Reza Samaei<sup>1</sup>,  
 PhD; Abolfazl Azhdarpoor<sup>1</sup>,  
 PhD; Saeed Yousefinejad<sup>2</sup>,  
 PhD

## Abstract

**Background:** Simultaneous existence of excessive amounts of fluoride and nitrate in drinking water can cause health problems for humans. In this study, simultaneous removal of fluoride and nitrate from aqueous solutions was investigated using a combination of electroreduction and electrocoagulation processes in a batch reactor with different electrodes.

**Methods:** In this study, at first, an optimum electrode was selected. Afterward, the effects of different operating parameters such as the current density (12- 36 mA/cm<sup>2</sup>), initial pH (5.5-8.5), NaCl concentration (0.5-1.5gr/L), and electrolysis time (15-120 min), on the removal of fluoride (initial concentration: 6 mg/L) and nitrate (initial concentration: 150 mg/) were evaluated, respectively.

**Results:** The highest efficiency of the concurrent fluoride and nitrate removal with Al-Cu electrode and in optimal experimental conditions of the current density of 36 mA/cm<sup>2</sup>, pH of 7, NaCl concentration of 1gr/L, and electrolysis time of 90 minutes was obtained 87.04 and 89.70%, respectively.

**Conclusion:** High catalytic activity of the copper cathode resulted in better performance than other cathodes in the simultaneous removal of fluoride and nitrate. Generally, it can be concluded that the electrochemical process can reduce the levels of fluoride and nitrate to the amounts below the WHO standard limits, 1.5 mg/L and 50 mg/L, respectively.

Please cite this article as: Ashoori R, Samaei MR, Azhdarpoor A, Yousefinejad S. Simultaneous Removal Fluoride and Nitrate from Water in a Batch Reactor Using Al and Fe anodes and Cu, Steel and Graphite Cathodes. *J Health Sci Surveillance Sys*. 2018;6(2):72-79.

**Keywords:** Electrochemical, Water, Fluoride, Nitrate, Removal

<sup>1</sup>Department of Environmental Health Engineering, School of Health, Shiraz University of Medical Sciences, Shiraz, Iran

<sup>2</sup>Department of Occupational Health Engineering, School of Health, Shiraz University of Medical Sciences, Shiraz, Iran

## Correspondence:

Mohammad Reza Samaei,  
 Department of Environmental Health Engineering, School of Health, Shiraz University of Medical Sciences, Shiraz, Iran

**Email:** mrsamaei@sums.ac.ir

**Tel:** +98-917-7320737

**Received:** 25 January 2018

**Revised:** 18 February 2018

**Accepted:** 12 March 2018

## Introduction

Drinking water is the main source of fluoride for our body.<sup>1</sup> The presence of a small amount of fluoride in drinking water is essential because it strengthens the bones and teeth.<sup>2</sup> However, excessive intake of drinking water containing high levels of fluoride can damage the body and cause diseases such as osteoporosis, arthritis, cancer, infertility, Alzheimer's, brain damage, and skeletal and dental fluorosis.<sup>3</sup> Since various factors such as nutrition, gender, age, genetics, race, body weight, and physical activity affect the occurrence and severity of complications caused by high levels of fluoride,<sup>4,5</sup> available guidelines and standards are unique for each community. The World Health Organization (WHO) and the Iranian National Standard recommend a range of 0.5-1.5 mg/L in drinking water.<sup>6,7</sup>

High nitrate concentration in water aqueous environments is also an important issue due to undesirable effects on human, such as methemoglobinemia and diabetes in infants, abortion in pregnant women, gastrointestinal cancers, and cardiovascular effects.<sup>8</sup> WHO and Iran Industrial Research and Standard Institute have set the permissible limit of nitrate and nitrite ions in drinking water to be 50 mg NO<sub>3</sub>/L and 3 mg NO<sub>2</sub>/L, respectively.<sup>9</sup>

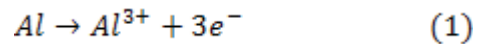
Our environment is contaminated with fluoride and nitrate through natural and anthropogenic sources. Fluoride enters groundwater via weathering and erosion of minerals and geochemical deposits.<sup>10</sup> Discharge of untreated wastewaters of industries such as semiconductor, aluminum, glass manufacturing, electroplating, and fertilizer industries into the environment leads to pollution of groundwater with fluoride.<sup>6, 10</sup> The amount of fluoride in groundwater depends on such factors as availability and solubility of fluoride-containing minerals, water flow rate, temperature, and concentration of calcium and bicarbonate ions in water.<sup>11</sup> Nitrate and fluoride are both high-soluble and electronegative anions in water, which have low ability for co-precipitation and adsorption.<sup>12, 13</sup> Thus, simultaneous elimination of them is a difficult task. So far, various methods such as adsorption and chemical precipitation, electrodialysis, reverse osmosis, ion exchange, biological treatment, and electrochemical methods have been used to remove fluoride and nitrate from the water.<sup>14-16</sup> Among the existing methods, the electrochemical method has attracted the attention of many researchers because of significant advantages such as maintaining the useful content of water during the removal of contaminants; simultaneous removal of the majority of all types of pollutions such as organic pollutants, turbidity, dyes, drugs, heavy metals and mineral anions including sulfide, fluoride and nitrate; removal of the smallest colloidal particles; no need to add chemicals; lower required investment and operating costs; and no need for care and maintenance.<sup>17-19</sup>

Some of the disadvantages of the electrochemical method include the need for frequent replacement of anode due to oxidation in the solution, degradation of the cathode activity due to the formation of impermeable oxide layer, the need for high electrical conductivity to suspend pollutants, dissolving metal hydroxides in some cases, anode passivation, and its dependency on expensive electric energy.<sup>18</sup>

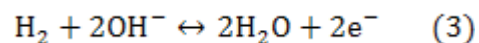
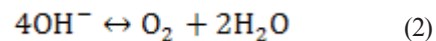
In the simplest form, the electrochemical unit is made up of an electrolyte cell with an anode and a cathode.<sup>20</sup> In the combined process of electrocoagulation and electroreduction, the anode material is corroded after applying the electrical current, which results in formation of coagulants

and charged metallic hydroxides, and eventually absorbs contaminants and suspended particles in the solution. The formation of hydrogen gas bubbles in the cathode and oxygen gas bubbles in the anode during the electrolysis process leads to floating the flocs on the surface of the solution, thereby facilitating their removal.<sup>21</sup>

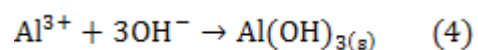
The electrochemical reactions in the reactor can be summarized as follows. For example, with Al metal:



Reaction in anode



Reaction in cathode



Reaction in aqueous solution

As a result of these reactions, charged complexes of aluminum hydroxide are formed which absorb fluoride and nitrate and neutralize the charged particles. Later, these complexes can be converted to amorphous  $Al(OH)_{3(s)}$ . This deposit, in turn, absorbs fluoride and nitrate.<sup>22</sup>

The removal of nitrate in water can be carried out by the process of electroreduction and electrocoagulation.<sup>23</sup> Electrolytic reduction of nitrate is a complex process.<sup>13</sup> In this process, nitrate is reduced to nitrite, nitrogen, and ammonia at the cathode. Nitrite and ammonia are undesirable products that may be oxidized to nitrate and nitrogen at the anode.<sup>24</sup> The products produced during electrolysis depend on operating conditions such as pH, coexisting ions, electrode type, and the cell shape.<sup>13</sup>

In Iran and many countries, groundwater resources are rapidly decreasing due to the population growth and climate change and this has increased the necessity of protecting water resources against pollutants.<sup>25</sup> Fluoride and nitrate are common pollutants of groundwater.<sup>26</sup> With regard to the side effects of high levels of fluoride and nitrate in drinking water, it is necessary to pay attention to removal of their excessive amounts from the drinking water. Therefore, appropriate methods should be applied to remove excessive amounts of these anions. In some regions of Iran, it has been observed that fluoride concentrations in drinking water sources are higher than the permissible level. Some of these areas are Bushehr, Bandar Abbas, Maku, Kerman, Yazd, Khorasan Razavi, West Azarbaijan, Fars, and Poldasht.<sup>27-29</sup> It has also been reported that in cities such as Tehran, Hamedan, Mashhad, Gorgan, Behbahan, Kerman, Fars, and Isfahan, the levels of nitrate in groundwater are higher than the permissible

level.<sup>25,30</sup> Concurrent presence of high concentrations of nitrate and fluoride in the water has been observed in some areas such as Lar.<sup>29</sup>

A method has been investigated for simultaneous removal of fluoride, nitrate and phosphate from synthetic wastewater by using stainless steel cathode and platinum anode and zerovalent iron; this method has achieved a removal efficiency of more than 90%.<sup>31</sup> Effective simultaneous removal of fluoride and nitrate by using the electrocoagulation process at small water treatment plants with monopolar and bipolar stainless steel and aluminum electrodes has been assessed (without taking into account the production of ammonia and nitrite).<sup>16</sup> However, so far no study has been conducted on the simultaneous removal of fluoride and nitrate electrochemically using this type of electrodes.

The general objective of this study was to evaluate the efficiency of the electrochemical process for the simultaneous removal of fluoride and nitrate from water with different electrodes, and to determine the effects of operating parameters such as the current density, pH, NaCl concentration, and electrolysis duration on the removal efficiency.

## Materials and Methods

### Preparation of Synthetic Solution

The chemicals used in this study were purchased from Merck and Sigma companies. These materials were made in Germany and the United States. First, fluoride and nitrate stock solutions were prepared by dissolving certain amounts of sodium fluoride and sodium nitrate in distilled water. Then, the solutions with the required concentrations were prepared using stock solutions and double-distilled water. Sodium chloride increases the electrical conductivity of the solution. It was used at various concentrations (0.5, 1 and 1.5 g/l) in the process of nitrate and fluoride removal. The pH of the initial solution was adjusted using 1 N hydrochloric acid and sodium hydroxide.

### Preparation of Electrochemical Reactor

The reactor used in this study was a batch (1 liter cylindrical plastic container) with a useful capacity of 500 ml of electrolyte (Figures 1 and 2). Rectangular anode and cathode electrodes with the size of 14×5×0.1 cm and effective area of 32.5 cm<sup>2</sup> were connected to a direct current power supply (model MEGATEK, MP3003 D) in a monopolar and parallel manner. A magnetic stirrer (model JENWAY1000) was used to homogenize the solution. Before using the electrodes, their surfaces were cleaned using hydrochloric acid 0.1 M. Then, the electrodes were washed with distilled water, dried, and eventually placed in the reactor. At

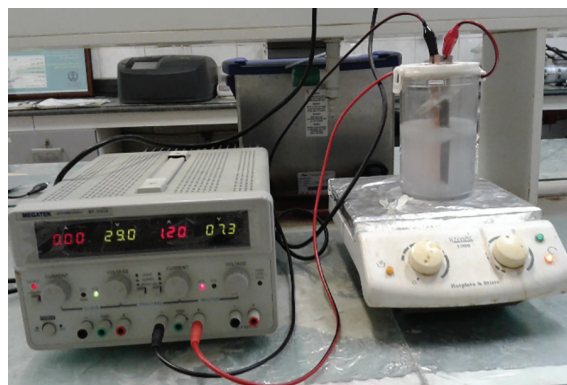


Figure 1: Electrochemical reactor and DC power supply

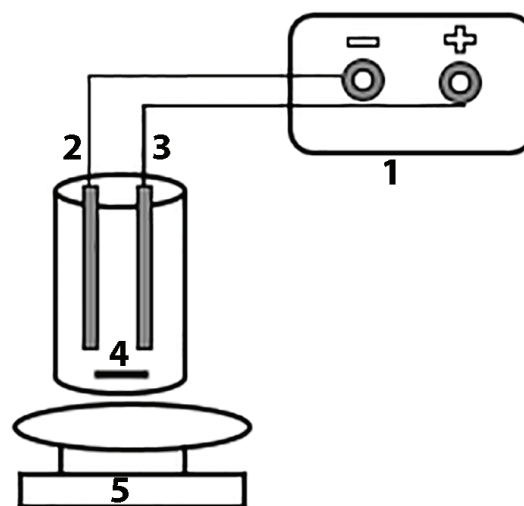


Figure 2: Schematic diagram of the electrochemical reactor used in this work: 1. DC power supply, 2. Cathode, 3. Anode, 4. Magnet, 5. Magnetic stirrer

the end of each test step, hydrochloric acid 0.1 M was used to wash the reactor and remove the remaining solids on its surfaces; then, the reactor was dried.

### Variables and Experiments Conditions

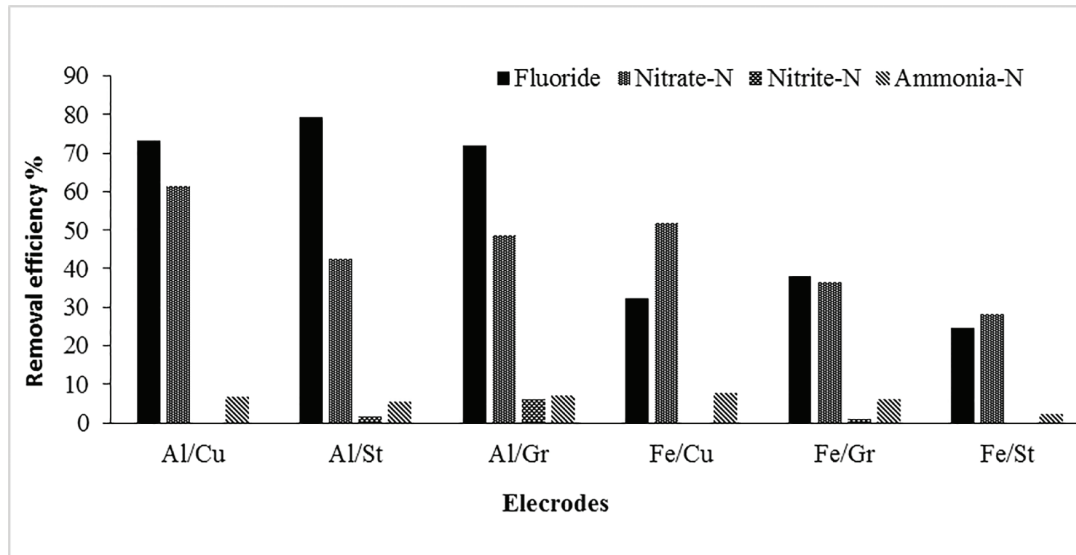
The variables and their levels are shown in Table 1. One factor at a time was used to design runs for this research. First, fluoride and nitrate concentrations were fixed at 6 mg/L and 150 mg/L, respectively. Then, the parameters of anode and cathode type, current density, pH, NaCl concentration, and electrolysis time were optimized. The total number of experiments was 52, and each experiment was repeated at least twice.

### Analytical Methods

At the end of each experiment, the reactor was left stationary for 20 minutes to allow the suspended solids to be deposited. The samples were pulled out of the upper part of the reactor and filtered, using 0.2 μm cellulose acetate syringe tip filter. The pH of the final solution was then measured. The pH of the samples was determined using a calibrated pH meter (model Metrohm 827 pH lab). To measure nitrite, nitrate, and fluoride concentrations, we injected the samples

**Table 1:** Variables and their levels

Level parameter	1	2	3	4	5	6
pH	5.5	7	8.5	-	-	-
Current density (mA/cm <sup>2</sup> )	12	24	36	-	-	-
NaCl dosage (gr/L)	0.5	1	1.5	-	-	-
Anode	Fe	Al				
Cathode	Cu	Steel	Graphite	-	-	-
Reaction time (min)	15	30	45	60	90	120

**Figure 3:** Removal efficiency of fluoride and nitrate with different electrodes at the density current 12 mA/cm<sup>2</sup>; pH, 7.0 ; inter-electrode distance, 1cm; NaCl dosage, 0.5g/l; electrolysis time, 90 min

into an IC device (model Metrohm, 850 Professional IC). The amount of ammonia was measured by the standard method,<sup>32</sup> using a spectrophotometer (model DR5000 HACH) and a Nessler's reagent. Given the initial values of nitrate and fluoride and amounts of the residuals, the removal efficiency was calculated, using Excel software and the corresponding graphs were plotted. The tests were repeated at least twice and their mean values were recorded.

## Results and Discussion

### Effect of Electrode Materials

The effects of various types of electrodes on nitrate and fluoride removal and the production of nitrite and ammonia byproducts were examined and the results are shown in Figure 3. As shown in the Figure, capabilities of the steel and copper cathodes for removal of fluoride and nitrate were better than other cathodes. The aluminum anode had a higher removal efficiency than the iron anode due to the higher affinity of aluminum coagulants to absorb fluoride and nitrate. These results were consistent with the findings of Takdastan et al.<sup>33</sup> In general, the efficiency of Al-Cu electrode in the simultaneous removal of fluoride and nitrate was 73.16% and 61.15%, respectively. It had the highest efficiency compared to other electrodes. The aluminum-graphite (Al-Gr) electrode had the highest ratio of nitrite produced (5.84

mg/L) to the nitrate removed, and the Al-Cu electrode had the lowest ratio of nitrite produced (0.33 mg/L) to the nitrate removed. High electrocatalytic activity of copper cathode in the reduction of nitrate has also been reported in the study of Bouzek et al. (2001).<sup>34</sup>

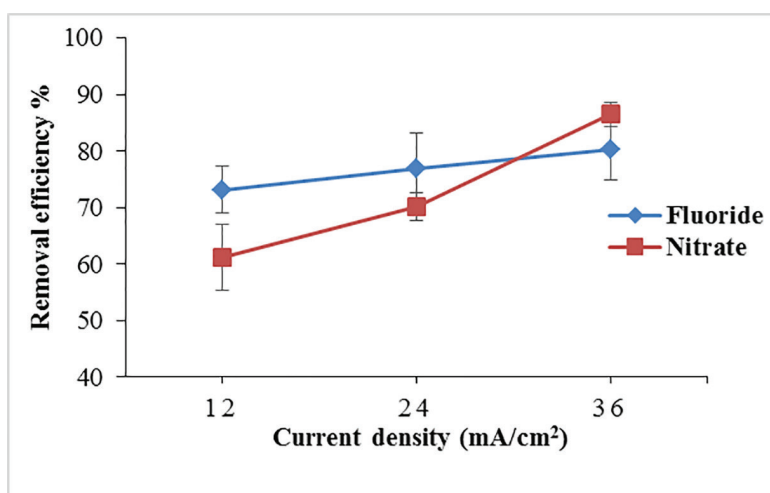
### Effect of Current Density

The current density has a significant effect on the coagulant and bubble production and also on the size and growth of the flocs.<sup>22</sup> Figure 4 shows the effect of the current density on the efficiency of the removal of fluoride and nitrate in constant conditions with the Al-Cu electrode. At the current density (36 mA/cm<sup>2</sup>), the highest removal efficiency of fluoride and nitrate was obtained 80.17% and 86.52%, respectively. Increasing the current intensity increases the release rate of OH<sup>-</sup> and Al<sup>3+</sup> ions from the cathode and anode, which leads to flocs formation and, therefore, increase of the removal rate of fluoride and nitrate.<sup>22</sup> These results were consistent with the findings of Emamjomeh and Sivakumar.<sup>35</sup> Increasing the current density also resulted in an increase in ammonia production and the maximum nitrite production (2.90 mg/L) occurred at the current density of 36 mA/cm<sup>2</sup>.

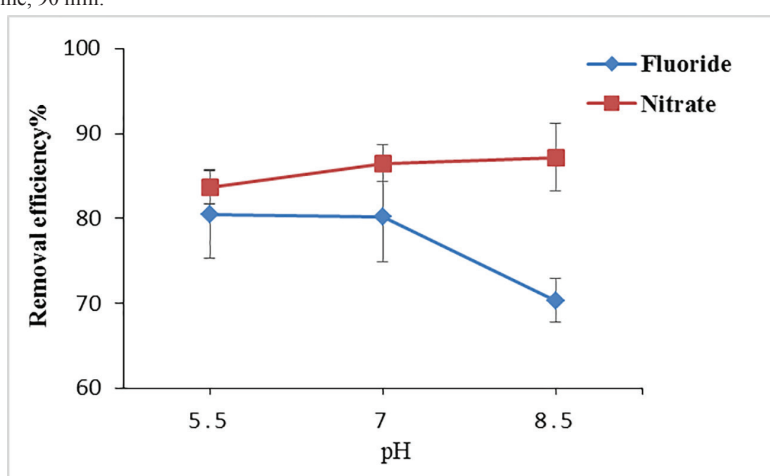
### Effect of Initial pH

As shown in Figure 5, amounts of fluoride removal with Al-Cu electrode for pHs of 5.5 and 7 were almost





**Figure 4:** The effect of the current density on the removal of nitrate and fluoride at pH, 7.0; inter-electrode distance, 1cm; NaCl dosage, 0.5g/l and electrolysis time, 90 min.



**Figure 5:** The effect of initial pH on the removal of nitrate and fluoride at the density current of 36 mA/cm<sup>2</sup>; inter-electrode distance of 1cm; NaCl dosage, of 0.5 g/L and electrolysis time of 90 min.

constant (80.51% and 80.17%, respectively), but amounts of nitrate removal increased from 83.68 to 86.52%. At pH of 8.5, the amount of fluoride removal decreased to 70.33%, and the level of nitrate removal slightly increased (87.21%). Aluminum speciation is influenced by pH. At pH of 5-7, positively charged aluminum hydroxide complexes such as  $Al(OH)_2^+$  and  $Al(OH)^{2+}$  can be formed.<sup>36</sup> Therefore, in this pH range, the removal efficiency of fluoride and nitrate increased due to the adsorption and charge neutralization. Decrease in fluoride removal at alkaline pHs is probably due to the formation of negatively charged hydroxyl aluminum complexes.<sup>19</sup> This is in accordance with the results obtained by Vasudevan et al.<sup>37</sup> At acidic pHs, oxidation of ammonia and nitrite increases as a result of the formation of hypochlorite acid. This leads to a decrease in the nitrate reduction relative to the neutral and alkaline states. The formation of ammonia in alkaline pHs was higher than neutral and acidic pHs.

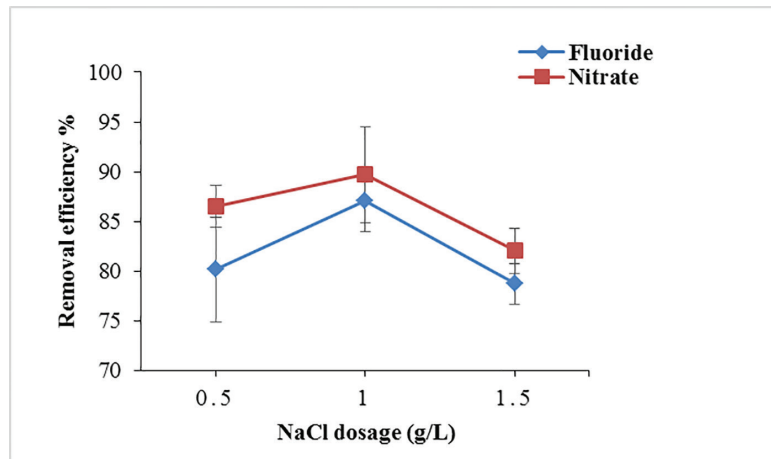
#### Effect of NaCl Dosage

Sodium chloride, in addition to its role as

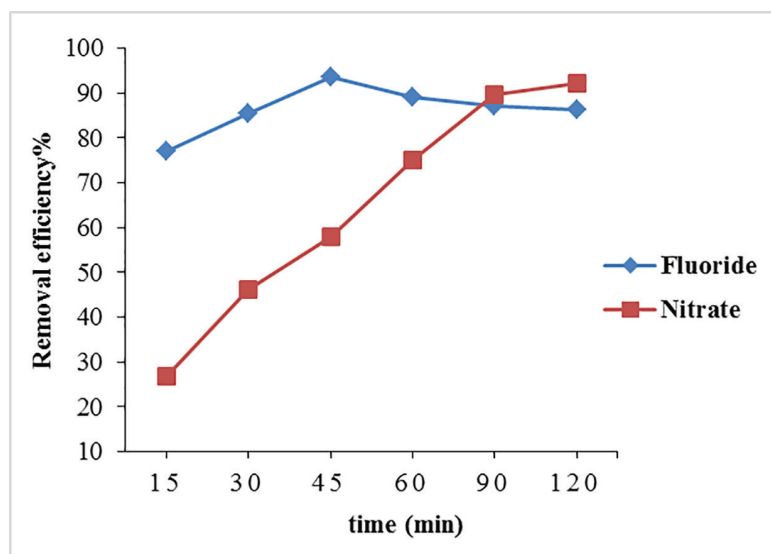
supporting electrolyte and increasing the solution conductivity, leads to the destruction of the anode passive membrane and produces more coagulants.<sup>38</sup> Figure 6 displays the effect of NaCl concentration on the percentage of the removal of fluoride and nitrate in optimal conditions. As shown in this Figure, by increasing the concentration of NaCl from 0.5 to 1 g/L, fluoride and nitrate removal increased to 87.04 and 89.7%, respectively; then, at NaCl concentration of 1.5 g/L, both percentages were decreased, which may be due to the competitive effect of anions and reduction of the anodic oxidation capacity in high concentrations of NaCl.<sup>22</sup> This is in line with the results of Fan et al.<sup>39</sup> In addition, at high concentrations of NaCl, more hypochlorite acid and hypochlorite were formed, which resulted in increasing oxidation of ammonia and nitrite and the reproduction of nitrate.<sup>40</sup>

#### Effect of Electrolysis Time

Fluoride and nitrate removal amounts are a function of the reaction duration. Figure 7 presents the effect of different electrolysis durations on the



**Figure 6:** The effect of NaCl dosage on the removal of nitrate and fluoride at the density current of 36 mA/cm<sup>2</sup>; pH of 7.0; inter-electrode distance of 1cm, and electrolysis time of 90 min.



**Figure 7:** The effect of electrolysis time on the removal of nitrate and fluoride at a density current of 36 mA/cm<sup>2</sup>; pH of 7.0; inter-electrode distance of 1cm, and NaCl dosage of 1g/L.

removal of fluoride and nitrate in optimal conditions. As the Figure shows, fluoride removal level was 77.05% at electrolysis time of 15 minutes and it reached 93.61% at electrolysis time of 45 minutes. The results indicated that at electrolysis times greater than 45 minutes, the percentage of fluoride removal decreased. By increasing the electrolysis time, the amount of nitrate significantly increased. The amount of nitrate removal was 26.88% at electrolysis time of 15 minutes and it reached 92.06% (highest removal level) at electrolysis time of 120 minutes. Reducing the fluoride removal efficiency for electrolysis times greater than 45 minutes may be due to several factors. By increasing the electrolysis time, the corrosion of the anode generates more coagulants in the solution and, with the accumulation of flocs on the anode surface, the functioning of the anode electrode is decreased, which decreases the coagulant production rate and subsequently reduces the removal of fluoride. Abrupt increase in mixing can also lead to breakdown of the flocs and release of fluoride trapped in the flocs.

Since two mechanisms of electrical reduction and coagulation are involved in the removal of nitrate, by increasing the electrolysis time, its removal level was higher than fluoride. According to Figure 6, at electrolysis times of 90 and 120 minutes, there was no significant increase in the nitrate removal, which can be due to the decrease of cathode activity.<sup>41</sup> Therefore, in general, the electrolysis time of 90 minutes is suitable for simultaneous removal of fluoride and nitrate.

## Conclusion

In the current study, the electrochemical process for simultaneous removal of fluoride and nitrate from synthetic water with different electrodes was evaluated. Various parameters such as anode and cathode material, the current density (12-36 mA/cm<sup>2</sup>), initial pH (5.5-8.5), NaCl concentration (0.5-1.5gr/L) and electrolysis time (15-120 min) were studied and optimized. The results showed that the highest percentage of simultaneous removal of fluoride and nitrate with Al-Cu electrode (87.04 and 89.7)

was obtained in optimum conditions, and copper cathode due to its high catalytic activity than other cathodes had better performance in the simultaneous removal of fluoride and nitrate. Therefore, electrochemical method is an effective process for treatment of nitrate and fluoride contaminated waters. However, some limitations can be noticed for the suggested approach such as decreasing the cathode and anode activity due to formation of coagulum on the electrode surface, and complexity of the mechanism in elimination of nitrate. On the other hand, the experimental conditions should be controlled precisely to prevent formation of ammonia and more concentration of electrolyte might be required in the case of using graphite electrodes to retain electrical conductance. Also, suggestion of appropriate conditions for simultaneous removal of fluoride and nitrate using low cost electrodes is the significant advantage of this work.

### Acknowledgement

The authors would like to thank Shiraz University of Medical Sciences, Shiraz, Iran and also Center for Development of Clinical Research of Nemazee Hospital and Dr. Nasrin Shokrpour for editorial assistance.

**Conflict of Interest:** None declared.

### References

- Dehghani M, Omrani R, Zamanian Z, Hashemi H. Determination of DMFT index among 7-11 year-old students and its relation with fluoride in Shiraz drinking water in Iran. 2013
- Li Y, Jiang Y, Wang T-J, Zhang C, Wang H. Performance of fluoride electrosorption using micropore-dominant activated carbon as an electrode. *Separation and Purification Technology*. 2017;172:415-21.
- Umran Tezcan U, Koparal AS, Ogutveren UB, Durucan A. Electrochemical process for the treatment of drinking water. *Fresenius Environmental Bulletin*. 2010;19(9):1906-10.
- Mousny M, Banse X, Wise L, Everett ET, Hancock R, Vieth R, et al. The genetic influence on bone susceptibility to fluoride. *Bone*. 2006;39(6):1283-9.
- Alvarez JA, Rezende KMP, Marocho SMS, Alves FBT, Celiberti P, Ciamponi AL. Dental fluorosis: exposure, prevention and management. *Journal of Clinical and Experimental Dentistry*. 2009;1(1):14-8.
- Ghanim A. Electrocoagulation of fluoride potable water utilizing bipolar electrodes: Statistical analysis and optimization through response surface methodology. *International Journal of Environmental Sciences*. 2014;4(5):660-75
- Azhdarpoor A, Radfard M, Rahmatinia M, Hashemi H, Hashemzadeh B, Nabavi S, et al. Data on health risk assessment of fluoride in drinking water in the Khash city of Sistan and Baluchistan province, Iran. *Data in brief*. 2018;21:1508-13.
- Tabash TM. Nitrate removal from groundwater using continuous flow electrocoagulation reactor: M. Sc. thesis, The Islamic University-Gaza; 2013.
- Ansari MH, Parsa JB. Removal of nitrate from water by conducting polyaniline via electrically switching ion exchange method in a dual cell reactor: Optimizing and modeling. *Separation and Purification Technology*. 2016;169:158-70.
- Zhu J, Zhao H, Ni J. Fluoride distribution in electrocoagulation defluoridation process. *Separation and Purification Technology*. 2007;56(2):184-91.
- Malay DK, Salim AJ. Comparative study of batch adsorption of fluoride using commercial and natural adsorbent. *Research Journal of Chemical Sciences*. 2011;1(7):68-75.
- Ghosh D. Removal of Fluoride, Iron and Arsenic from Drinking water using a combination of electrocoagulation and Microfiltration 2009.
- Dash BP, Chaudhari S. Electrochemical denitrification of simulated ground water. *Water Research*. 2005;39(17):4065-72.
- Han S-H, Chang I-S. Comparison of Nitrate and Fluoride Removals between Reverse-Osmosis, Nano-Filtration, Electro-Adsorption, Electro-Coagulation in Small Water Treatment Plants. *Journal of the Korea Academia-Industrial cooperation Society*. 2013;14(4):2027-36.
- Mekonen A, Kumar P, Kumar A. Integrated biological and physiochemical treatment process for nitrate and fluoride removal. *Water research*. 2001;35(13):3127-36.
- Han S-H, Chang I-S. Fluoride and nitrate removal in small water treatment plants using electro-coagulation. *Journal of Korean Society of Water and Wastewater*. 2011;25(5):767-75.
- Yehya T, Chafi M, Balla W, Vial C, Essadki A, Gourich B. Experimental analysis and modeling of denitrification using electrocoagulation process. *Separation and Purification Technology*. 2014;132:644-54.
- Mollah MYA, Schennach R, Parga JR, Cocke DL. Electrocoagulation (EC)—science and applications. *Journal of hazardous materials*. 2001;84(1):29-41.
- Hashim KS, Shaw A, Al Khaddar R, Pedrola MO, Phipps D. Defluoridation of drinking water using a new flow column-electrocoagulation reactor (FCER)-Experimental, statistical, and economic approach. *Journal of environmental management*. 2017;197:80-8.
- Vasudevan S. Electrochemical Processes for Water Quality Upgradation. *International Journal of Waste Resources*. 2013;3(2).
- Garg UK, Sharma C, editors. *Electrocoagulation: Promising technology for removal of fluoride from drinking water—a review*. *Biological Forum-An International Journal*; 2016.
- Thakur LS, Mondal P. Simultaneous arsenic and fluoride removal from synthetic and real groundwater by electrocoagulation process: Parametric and cost evaluation. *Journal of environmental management*.

- 2017;190:102-12.
- 23 Koparal AS, Ögütveren ÜB. Removal of nitrate from water by electroreduction and electrocoagulation. *Journal of hazardous materials*. 2002;89(1):83-94.
- 24 Seetharam BN, Brahmaiah T, Basha UI, Murthy H, Kalkur JN. Effect of Operational Parameters on Nitrate Removal from the Simulated Groundwater Using Electrochemical Method *International Journal of Trend in Research and Development*, 2016; 3(1).
- 25 Pirsahab M, Khamutian R, Molok P, Shekoohizade MJ. Survey of nitrate in ground waters of different regions of Iran: a systematic review. *The 1th Conference and Exhibition on Environmental Energy and Clean industry*.
- 26 Waikar M, Dhole AA. Reduction of Fluoride from Groundwater by Electrocoagulation using Iron Electrode. *Journal of Civil Engineering and Environmental Technology*. 2015;2(9):54-7.
- 27 Ejlali A, Taghipour H, Khashabi E. The study of fluoride level in drinking water in villages of makoo, in 2014. *URMIA MEDICAL JOURNAL*. 2015;26(9):754-63.
- 28 Ahmadimarzaleh M, Balideh H, Hosseindoost GR. the concentration of fluoride in drinking water in different cities of iran and Its health effects. *The second national conference in environmental pollution and the sustainable development2015*.
- 29 Rezaei M, Nikbakht M, Shakeri A. Geochemistry and sources of fluoride and nitrate contamination of groundwater in Lar area, south Iran. *Environmental Science and Pollution Research*. 2017;24(18):15471-87.
- 30 Mahvi A, Naseri S, Mohamadi A, Shekarriz M, Alimohamadi M. Study of nitrate reduction from water using nanosized iron. *Iranian Journal of Health and Environment*. 2011;4(3):313-20.
- 31 Jeong J-Y, Song Y-H, Kim J-H, Park J-Y. Simultaneous removal of nitrate, phosphate, and fluoride using a ZVI-packed bed electrolytic cell. *Desalination and Water Treatment*. 2014;52(4-6):737-43.
- 32 American Public Health Association A. Standard methods for the examination of water and wastewater. American Public Health Association (APHA): Washington, DC, USA2005.
- 33 Takdastan A, Tabar SE, Neisi A, Eslami A. Fluoride removal from drinking water by electrocoagulation using iron and aluminum electrodes. *Jundishapur Journal of Health Sciences*. 2014;6(3).
- 34 Bouzek K, Paidar M, Sadilkova A, Bergmann H. Electrochemical reduction of nitrate in weakly alkaline solutions. *Journal of Applied Electrochemistry*. 2001;31(11):1185-93.
- 35 Emamjomeh MM, Sivakumar M. Denitrification using a monopolar electrocoagulation/flotation (ECF) process. *Journal of environmental management*. 2009;91(2):516-22.
- 36 Kim K-J, Baek K, Ji S, Cheong Y, Yim G, Jang A. Study on electrocoagulation parameters (current density, pH, and electrode distance) for removal of fluoride from groundwater. *Environmental Earth Sciences*. 2016;75(1):45.
- 37 Vasudevan S, Lakshmi J, Sozhan G. Studies on a Mg-Al-Zn Alloy as an Anode for the Removal of Fluoride from Drinking Water in an Electrocoagulation Process. *Clean-Soil, Air, Water*. 2009;37(4-5):372-8.
- 38 Behbahani M, Moghaddam MA, Arami M. Techno-economical evaluation of fluoride removal by electrocoagulation process: Optimization through response surface methodology. *Desalination*. 2011;271(1-3):209-18.
- 39 Fan N, Li Z, Zhao L, Wu N, Zhou T. Electrochemical denitrification and kinetics study using Ti/IrO<sub>2</sub>-TiO<sub>2</sub>-RuO<sub>2</sub> as the anode and Cu/Zn as the cathode. *Chemical Engineering Journal*. 2013;214:83-90.
- 40 Pérez G, Ibáñez R, Urtiaga A, Ortiz I. Kinetic study of the simultaneous electrochemical removal of aqueous nitrogen compounds using BDD electrodes. *Chemical Engineering Journal*. 2012;197:475-82.
- 41 Kalaruban M, Loganathan P, Kandasamy J, Naidu R, Vigneswaran S. Enhanced removal of nitrate in an integrated electrochemical-adsorption system. *Separation and Purification Technology*. 2017;189:260-6.