The Use of Electrocoagulation to Remove Fluoride, Nitrates and Phosphorous from Water

Rusul K Abdulkhadher1*, Ali j Jaeel1

1 Department of Civil Engineering, College of Engineering, Wasit University, Iraq

* Corresponding author: rkareem@uowasit.edu.iq

Abstract. In recent times, the concentrations of fluoride, nitrates and phosphates in the water have increased as a result of a decrease in the quantities of water and an increase in industrial waste dumped into the rivers. In the current study, a method of electrocoagulation was depended on to treat water contaminated with fluoride, nitrates, and phosphates, using aluminum electrodes as anode and cathode electrodes. According to the obtained results, it can be noticed that the best fluoride, nitrates, and phosphates removal were 99%, 99%, 100%, respectively. It was obtained with a Hydraulic retention time HRT = 3 hours, an applied voltage = 40 volts, a gap between the used electrodes=2 cm, the anode's active area= 792 cm², and the flow angle= zero. By using an initial concentration (5 mg F/l, with pH= 4) for fluoride, (75 mg NO₃/ l, with pH= 6) for nitrates, and (0.5 mg PO₄ / l, with pH=9) for phosphates.

1. Introduction
Although water is an essential element for life on planet Earth. However, only 1% or less is drinkable [1-3]. Due to population growth, one of the main challenges currently facing human civilization is the provision of sufficient quantities of potable water [4,5]. Therefore, the ability of mankind to obtain sufficient sources of the required water should be considered as an essential global problem. Achieving sustainable solutions for water management is the possible solution for this challenge. The population of the world is over 7 billion and is estimated to reach 10 billion by 2040 [6]. Climate change also negatively affects fresh water resources in the middle of cities, which in turn affects the sustainable development of water availability and thus affects economic and social activities [7]. Additionally, several studies have shown that freshwater resources are commonly influenced by contamination [8,9]. Water scarcity situations are faced in different regions around the world, which means that the gap between water supply and demand is likely to grow in the future. In 2010, the European Environment Agency reported that municipal water consumption is driven by complex interactions between human and natural system factors on multiple spatial and time scales [10]. There is an inevitable need to maximise wastewater treatment. In order to avoid potential global crises. Due to depleting available resources of clean and potable water, making use of unlimited available saltwater in the oceans, and reducing discharges from water treatment plants. Thousands of years ago, people attempted and succeeded in improving the water quality through
processes of treatment such as the chemical alum addition, boiling, straining, and filtering through charcoal to separate suspended solids from water [11]. In recent decades the wastewater treatment industry has identified discharges of inorganic anions such as nitrates, phosphates, and fluorides in waterways pose a great danger to the natural environment, which has also been shown to be dangerous to humans. To remove inorganic pollutants from water supplies, many common treatment techniques are nowadays used. For instance, coagulation and precipitation, this treatment system has some disadvantages, such as dealing with large amounts of chemicals and the formation of great quantities of sludge that may be harmful to the environment and difficult to get rid of [12]. While in adsorption, ion exchange, and membrane separations, there are some problems such as the additional costs connected with elimination of utilised regenerants materials and regeneration [13,14]. Nutrients (nitrates and phosphates) and fluoride are inorganic anions, which are present in different concentrations in water where the excess of concentration leads to contamination of water and wastewater.

The presence of fluoride in drinking water in a small amount is essential due to it strengthens the teeth and bones [15]. Naturally present amounts of phosphates in ground water and surface water are not dangerous to the environment, animals, or human health [16]. For all life forms, phosphate (PO₄³⁻) is an essential nutrient element. For the living cells, it plays a fundamental role in the generation of deoxyribonucleic acid (DNA), ribonucleic acid (RNA), adenosine di- and triphosphate (ADP and ATP) [17]. The presence of phosphates and nitrates in a quantity that is greater than the permissible quantities leads to the occurrence of the eutrophication phenomenon that causes an imbalance in the environment [18]. Excessive doses of nitrates in drinking water also cause blue baby syndrome and stomach cancer. High phosphate concentrations also lead to digestive problems [19,20]. Among the diseases caused by excessive amounts of fluoride are brain damage, arthritis, and osteoporosis [21]. Due to the above problems the three anions need to be removed from water using the appropriate methods. The main purpose of the present research is the implementation of the electrocoagulation technique to remove fluoride, phosphorus, and nitrate from drinking water. Using the EC system to provide environmentally acceptable water. Test operating parameters through a continuous system running condition to enhance the removal efficiency performance of the EC.

2. Experimental work

2.1. Material and methods

Synthetic water samples, containing fluoride, nitrates and phosphates, were treated using electrocoagulation cell at different the pollutant's initial concentration (Cᵢ), the acidic function (pH), the applied voltage(V), the gap between the used electrodes (S), the anode's active area(Aₑ) and the flow angle(θ) in addition to the hydraulic retention time. The desire pH value (4 to 9) was provided using HCl and NaCl. Voltage value was provided using a DC power supply named LW Longwel (LW-K3010D), (0-10 A, 0–40 V). While the electrode distance and the anode's active area were manually adjusted to the required value (2 to 8 cm), (198 to 792 cm²), respectively. In the EC reactor, at a flow rate of = 25 ml/min, the electrolysis process was carried out by continuously pumping the synthetically prepared water samples. The HRT time changed from (1-3) hours. 10 mL of water samples were taken from the EC outlet reactor on a regular basis to track the removal process. Pollutant concentrations are measured spectrophotometrically by using single beam UV spectrophotometer (SpectroDirect). In each step, one variable was changed and then their optimum value was proposed for the next part of the experiment. All experiments were carried out at room temperature (25±5°C). The removal efficiency of Fluoride, Nitrates and Phosphor in the artificial polluted water treated by electrocoagulation is calculated as follows:

$$\eta\% = \left( \frac{C_o - C_p}{C_o} \right) \times 100$$ (1)
Where \( \eta \% \) is removal efficiency of fluoride, nitrates and phosphor, \( C_i \) is the initial concentration of pollutants (mg/l), and \( C_e \) is final pollutant concentration (mg/l).

2.2. Electrocoagulation cell
The removal of fluoride, nitrate and phosphorous was carried out using an EC reactor. The electrocoagulation cell is rectangular in dimensions 20 cm ×16cm×12cm with 6mm thickness made of transparent plastic served as the EC reactor. Eight aluminum electrodes size 20 cm ×6cm×0.09cm. As shown in Figure 1.

![Image of EC cell](image)

**Figure 1.** A photograph of the EC cell 1. Influent tank (polluted water). 2. Pump. 3. Power supply. 4. EC cell. 5. Anode and cathode. 6. Final storage.

3. Results and discussion

3.1. Influence of HRT on F, NO\(_3\), PO\(_4\) elimination relative to the initial concentration of F, NO\(_3\), PO\(_4\) (\(C_i\))

The influence of HRT on removal of F, NO\(_3\), PO\(_4\) which has been examined for solutions with initial concentrations (\(C_i\)) of 5, 10, 15, and 20 mg/L for F, 75, 100, 150, and 200 mg/L for NO\(_3\), 0.5. 1, 1.5, and 2 mg/L for PO\(_4\). While HRT was changed to 1, 1.5, 2, 2.5 and 3 hours. When pH environment, the applied voltage (V), the distance between electrodes (S), the anode effective area (A), and the angle of flow (\(\theta\)) were kept constant at 7, 20 volts, 4 cm, 198 cm\(^2\), and 90\(^\circ\), respectively. Figure 6 showed that higher initial concentration extended the treatment duration. The explanation for this decrease in removal with an increase in the initial concentration is the fact that this may be due to the formation of an insufficient amount of coagulation complexes [22,23].
3.2. The influence of HRT on F, NO₃, PO₄ removal relative to pH

To examine the influence of (HRT), the pH was fixed at a certain value (4, 5, 6, 7, 8 and 9) while HRT was changed to 1, 1.5, 2, 2.5 and 3 hours. When the Cᵢ, V, S, Aₑffect, and the angle of flow (θ) were maintained at a steady level of (5 F mg/l, 75 NO₃ mg/l, 0.5 PO₄ mg/l) 20V, 4cm, 198 cm² and 90º respectively. A decrease of fluoride removal can be observed when the primary pH value raised from 4 to 8, and it reached the smallest value at primary pH 8. While by increasing the pH from 4 to 6, the removal efficiency of nitrates would increase. After pH 6, the removal efficiency gradually decreased. While, the highest removal efficiency of phosphate can be achieved near pH 9, this agrees with [24,25].
3.3. Influence of HRT on F, NO₃, PO₄ elimination relative to the voltage applied (V)

The HRT was changed to 1, 1.5, 2, 2.5, and 3 hours while the voltage applied (V) was set at a specific value (10, 20, 30, and 40 V). When the Ci, pH, S, Aₚₑ, and the angle of flow (θ) were kept constant at (5 F mg/l, pH 4), (75 NO₃ mg/l, pH 6), (0.5 PO₄ mg/l, pH 9), 4, 4 cm, 198 cm², and 90º, respectively. Removal efficiency increases with increasing voltage. Actually, the supplied voltage governs both the rates of bubble generation and dosage of coagulant, it also affects the mixing of solution and transfer of mass at the electrodes [26].

3.4. Influence of HRT on F, NO₃, PO₄ elimination relative to electrode distance (S)
To examine the influence of (HRT), HRT was changed to 1, 1.5, 2, 2.5 and 3 hours while the electrode distance (S) was fixed at a specific value (2, 4, 6, and 8 cm). When the \( C_i \), pH, V, \( A_{\text{eff}} \), and the angle of flow (\( \theta \)) were kept constant at (5 mg/l, pH 4), (75 NO\(_3\) mg/l, pH 6), (0.5 PO\(_4\) mg/l, pH 9), 40 V, 198 cm\(^2\), and 90\(^\circ\) respectively. The distance = 2 cm represents the optimal distance in this study. This can be attributed to the face that as the value of S raises, resistance of the cell and the development of anodic passive film increase significantly. Thus, the current reduces, and the quantity of the generated floc reduces as well, so the efficiency changes [27].

Figure 11. Influences of the S on the removal of F.  

Figure 12. Influences of the S on the removal of NO\(_3\).  

Figure 13. Influences of the S on the Removal of PO\(_4\).

3.5. HRT's impact on F, NO\(_3\), PO\(_4\) elimination in relative to an anode's effective area (\( A_{\text{eff}} \))

To examine the influence of (HRT), HRT was changed to 1, 1.5, 2, 2.5 and 3 hours while an anode's effective area (\( A_{\text{eff}} \)) was fixed at a specific value (198, 396, 594 and 792 cm\(^2\)). When the \( C_i \), pH, V, S, and the angle of flow (\( \theta \)) were kept constant at (5 mg/l, pH 4), (75 NO\(_3\) mg/l, pH 6), (0.5 PO\(_4\) mg/l, pH 9), 40 volt, 2 cm, and 90\(^\circ\) respectively. Increasing the active area of the anode from (198 to 792 cm\(^2\)) enhanced removal efficiency and reduced processing time that was required. This can be explained by that the use of a larger active area generates large amounts of aluminum which traps more pollutants and improves the removal efficiency [28,29].
3.6. Influence of HRT on fluoride elimination relative to flow direction

To examine the influence of (HRT), the angle of flow was set to a specific value (the original direction where the flow is perpendicular to the surface area of the anode electrode, i.e., $\theta=90^\circ$ and the other direction where the flow is parallel to the surface area of the anode electrode, i.e., $\theta=0^\circ$). While HRT was changed to 1, 1.5, 2, 2.5 and 3 hours. When the $C_i$, pH, V, S, and $A_{eff}$ have been kept constant at (5 F mg/l, pH 4), (75 NO$_3$ mg/l, pH 6), (0.5 PO$_4$ mg/l, pH 9), 40V, 2cm, 792 cm$^2$, respectively. The removal efficiency increased when the direction of flow was changed from perpendicular ($\theta=90^\circ$) to parallel ($\theta=0^\circ$) relative to anode electrode surface area plane. This can be attributed to the fact that the horizontal flow, i.e. with angle zero, enhances the homogeneity of the bubbles over the cross-section and leads to an increase in the diffusion rate of Al$^{3+}$ dissolved from the anode to the emulsion, this agrees with [30,31].
4. Conclusions

In this study, aluminum electrodes were used in the EC process, based on previous studies. As these electrodes proved their cost and efficiency in removing fluoride, nitrates, and phosphorus from water to obtain environmentally acceptable water. It was clearly indicated that the fluoride, nitrate, and phosphate removal efficiency of 99%, 99%, and 100%, respectively, were achieved from the generation of aluminum hydroxide in the EC cell. Within a HRT = 3 hours, an applied voltage =40 volts, a gap between the used electrodes=2 cm, the anode's active area= 792 cm$^2$, and the flow angle= zero. The aluminum hydroxide removes the fluoride, nitrate, and phosphate found in the water and reduces the concentration of fluoride, nitrate, and phosphate to less than 1.5, 50, and 0.1 mg/l, respectively and makes it within the acceptable range for drinking water as mentioned by WHO guidelines.

5. References

[1] Al-Saati N H, Hussein T K, Abbas M H, Hashim K, Al-Saati Z N, Kot P, Sadique M, Aljefery M H and Carnacina I 2019 Statistical modelling of turbidity removal applied to non-toxic natural coagulants in water treatment: a case study Desalination and Water Treatment 150 406-12.

[2] Omran I I, Al-Saati N H, Hashim K S, Al-Saati Z N, Patry K, Khaddar R A, Al-Jumeily D, Shaw A, Ruddock F and Aljefery M 2019 Assessment of heavy metal pollution in the Great Al-Mussaib irrigation channel Desalination and Water Treatment 168 165-74.

[3] Zubaidi S L, Kot P, Hashim K, Alkhaddar R, Abdellatif M and Muhsin Y R 2019 Using LARS–WG model for prediction of temperature in Columbia City, USA IOP Conference Series: Materials Science and Engineering 584.

[4] Emamjomeh M M, Mousazadeh M, Mokhtari N, Jamali H A, Makiabadi M, Naghdali Z, Hashim K S and Ghanbari R 2020 Simultaneous removal of phenol and linear alkylbenzene sulfonate from automotive service station wastewater: Optimization of coupled electrochemical and physical processes Separation Science and Technology 55 3184-94.

[5] Zubaidi S L, Al-Bugharbee H, Muhsin Y R, Hashim K and Alkhaddar R 2020 Forecasting of monthly stochastic signal of urban water demand: Baghdad as a case study IOP Conference Series: Materials Science and Engineering 888.

[6] Coleman D 2004. V. world population in 2300: A century too far?. World Population to 2300.
[7] Zubaidi S L, Al-Bugharbee H, Muhsen Y R, Hashim K, Alkhaddar R M, Al-Jumeily D and Aljaaf A J 2019 The Prediction of Municipal Water Demand in Iraq: A Case Study of Baghdad Governorate 12th International Conference on Developments in eSystems Engineering (DeSE)

[8] Hashim K S, Al-Saati N H, Alquzweeni S S, Zubaidi S L, Kot P, Kraidi L, Hussein A H, Alkhaddar R, Shaw A and Alwash R 2019 Decolourization of dye solutions by electrocoagulation: an investigation of the effect of operational parameters First International Conference on Civil and Environmental Engineering Technologies (ICCEET) 584.

[9] Hashim K S, Hussein A H, Zubaidi S L, Kot P, Kraidi L, Alkhaddar R, Shaw A and Alwash R 2019 Effect of initial pH value on the removal of reactive black dye from water by electrocoagulation (EC) method 2nd International Scientific Conference.

[10] Hashim K, Kot P, Zubaid S, Alwash R, Al Khaddar R, Shaw A, Al-Jumeily D and Aljefery M 2020 Energy efficient electrocoagulation using baffle-plates electrodes for efficient Escherichia Coli removal from Wastewater Journal of Water Process Engineering 33 101079-86.

[11] Nguyen D D, Yoon Y S, Bui X T, Kim S S, Chang S W, Guo W, and Ngo H H 2017 Influences of operational parameters on phosphorus removal in batch and continuous electrocoagulation process performance Environmental Science and Pollution Research 24(32), 25441-25451

[12] Duan, J., & Gregory, J. 2003 Coagulation by hydrolysing metal salts. Advances in colloid and interface science, 100 475-502.

[13] Li Y, Liu C, Luana Z, Peng X, Zhu C, Chena Z, Zhang Z, Fan J, Jia Z  2006 Phosphate removal from aqueous solutions using raw and activated red mud and fly ash, Journal of hazardous materials 137 374-383.

[14] Lin L, Wan C, Lee D J, Lei Z, and Liu X 2014 Ammonium assists orthophosphate removal from high-strength wastewaters by natural zeolite Separation and Purification Technology 133 351-356.

[15] Li Y, Jiang Y, Wang T J, Zhang C, and Wang H 2017 Performance of fluoride electrosorption using micropore-dominant activated carbon as an electrode Separation and Purification Technology 172 415-421.

[16] Jiang C, Jia L, He Y, Zhang B, Kirumba G, and Xie J 2013 Adsorptive removal of phosphorus from aqueous solution using sponge iron and zeolite Journal of colloid and interface science 402 246-252.

[17] Ohtake H, Kuroda A, Chandrasekaran M, Wu H, Tanaka S, Morohoshi T, and Takiguchi N 2001 Molecular Genetics of Bacterial Polyphosphate Accumulation to Better Understand the Mechanism Underlying Biological Phosphorus Removal Environmental Monitoring and Biodiagnostics of Hazardous Contaminants Springer 181–196.

[18] Jorgensen S E, & Williams W D 2001 Water quality: The impact of eutrophication. UN Environment Programme.

[19] Chiu H F, Tsai S S, and Yang C Y 2007 Nitrate in drinking water and risk of death from bladder cancer: an ecological case-control study in Taiwan, Journal of toxicology and environmental Health, Part A, 70(12), 1000-1004.

[20] Kumar E, Bhatnagar A, Ji M, Jung W, Lee S H, Kim S J, Lee G, Song H, Choi J Y, Yang J S, Jeon B H  2009 Defluoridation from aqueous solutions by granular ferric hydroxide (GFH), Water Research, 43(2), 490-498.

[21] Ün Ü T, Koparal A S, Öğütveren Ü B, and Durucan A 2010 Electrochemical process for the treatment of drinking water Fresenius Environmental Bulletin, 19(9), 1906-1910.
[22] Vasudevan S, Lakshmi J, and Sozhan G 2009 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, 37(4-5), 372-378.

[23] Hendaoui K, Ayadi M T, and Ayari F 2021 Optimization and mechanisms analysis of indigo dye removal using continuous electrocoagulation, Chinese Journal of Chemical Engineering 29 242-252.

[24] İrdemez Ş, Demircioğlu N, and Yildiz Y Ş 2006 The effects of pH on phosphate removal from wastewater by electrocoagulation with iron plate electrodes Journal of hazardous materials 137(2), 1231-1235.

[25] Shalaby A, Nassef E, Mubark A, and Hussein M 2014 Phosphate removal from wastewater by electrocoagulation using aluminium electrodes American Journal of Environmental Engineering and Science 1(5) 90-98.

[26] Maitlo H A, Lee J, Park J Y, Kim J C, Kim K H, and Kim J H 2019 An energy-efficient air-breathing cathode electrocoagulation approach for the treatment of arsenite in aquatic systems Journal of Industrial and Engineering Chemistry 73 205-213.

[27] Hashim K S, Shaw A, Al Khaddar R, Pedrola M O, and Phipps D 2017 Defluoridation of drinking water using a new flow column-electrocoagulation reactor (FCER)-Experimental, statistical, and economic approach Journal of environmental management 197 80-88.

[28] Sharma A K and Chopra A K 2017 Removal of nitrate and sulphate from biologically treated municipal wastewater by electrocoagulation Applied Water Science 7(3) 1239-1246.

[29] Amarine M, Lekhlif B, Sinan M, El Rharras A, and Echaabi J 2020 Treatment of nitrate-rich groundwater using electrocoagulation with aluminum anodes Groundwater for Sustainable Development 11 100371.

[30] Fouad Y O A, Konsowa A H, Farag H A, and Sedahmed G H 2009 Performance of an electrocoagulation cell with horizontally oriented electrodes in oil separation compared to a cell with vertical electrodes Chemical Engineering Journal, 145(3) 436-440.

[31] Liu Y, Jiang W, Yang J, Li Y, Chen M, and Li J 2017 Experimental study on evaluation and optimization of tilt angle of parallel-plate electrodes using electrocoagulation device for oily water demulsification Chemosphere 181 142-149.