Journal of
Water, Sanitation & Hygiene for Development

IWA PUBLISHING

© 2022 The Authors Journal of Water, Sanitation and Hygiene for Development Vol 12 No 4, 394 doi: 10.2166/washdev.2022.043

Research Paper

Electrochemical defluorination of water: an experimental and morphological study

Muhammad Abdulredha[a], Shatha Y. Al-Samarrai[b], Ameer H. Hussein[c], Isra'a Sadi Samaka[d], Nadhir Al-Ansari[e,*] and Omar A. Aldhaibani[f]

[a] Department of Civil Engineering, College of Engineering, University of Kerbala, Kerbala, Iraq
[b] Chemistry Department, College of Science, Tikrit University, Tikrit, Iraq
[c] Al-Mussaib Technical Institute/Al-Furat Al-Awsat Technical University, Babylon, Iraq
[d] Department of Environmental Engineering, College of Engineering, University of Babylon, Babylon, Iraq
[e] Department of Civil, Environmental and Natural Resources Engineering, Mining and Geotechnical Engineering, Luleå University of Technology, Luleå, Sweden
[f] Built Environment and Sustainable Technologies (BEST) Research Institute, Liverpool John Moores University, Liverpool, UK
*Corresponding author. E-mail: nadhir.alansari@ltu.se

ABSTRACT

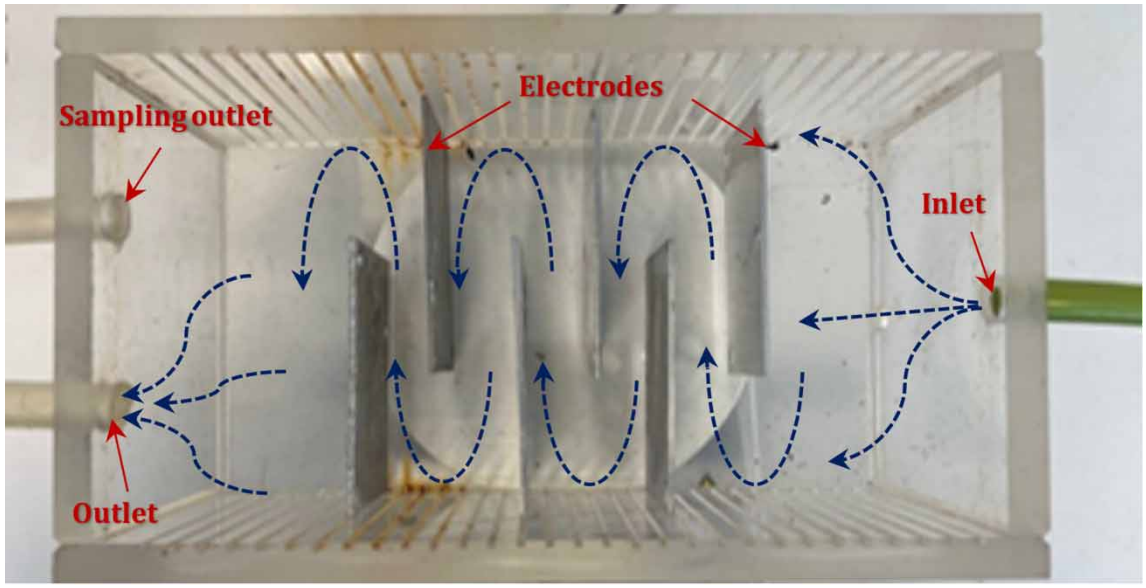
This experimental study concerns the elimination of fluoride from water using an electrocoagulation reactor having a variable flow direction in favour of increasing the electrolysing time, saving the reactor area, and water mixing. The detention time of the space-saver EC reactor (S-SECR) was measured and compared to the traditional reactors using an inert dye (red drain dye). Then, the influence of electrical current ($1.5 \leq \delta \leq 3.5$ mA cm^{-2}), pH of water ($4 \leq$ pH ≤ 10), and distance between electrodes ($5 \leq \phi \leq 15$) on the defluoridation of water was analysed. The effect of the electrolysing activity on the electrodes' morphology was studied using scanning electron microscopy (SEM). Additionally, the operational cost was calculated. The results confirmed the removal of fluoride using S-SECR met the guideline of the World Health Organization (WHO) for fluoride levels in drinking water of $\leq$1.5 mg/L. S-SECR abated fluoride concentration from 20 mg/L to the WHO's guideline at δ, ϕ, pH, operational cost, and power consumption of 2.5 mA cm^{-2}, 5 mm, 7, 0.346 USD m^{-3}, and 5.03 kWh m^{-3}, respectively. It was also found the S-SECR enhanced the detention time by 190% compared to the traditional reactors. The appearance of dents and irregularities on the surface of anodes in the SEM images proves the electrolysing process.

Key words: aluminium electrodes, defluoridation, engineered reactor, electrocoagulation

HIGHLIGHTS

- An engineered EC reactor (S-SECR) was used to remove fluoride from water.
- S-SERC achieved the WHO's guideline for fluoride in water in 30 min.
- S-SECR increased the detention time by about 190%.
- The operating cost was 0.346 USD m^{-3}.
- SEM images showed severe changes in the surface of anodes.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Surface water pollution with fluoride mainly results from natural and anthropogenic processes – naturally, fluoride results from weathering fluorine-rich geological formations, while the anthropogenic sources include industrial wastewater, such as the effluents of semiconductors and coal plants (Arif *et al.* 2013; Sandoval *et al.* 2019). Weathering of fluorine-riches formations increases the fluoride concentrations, especially in groundwater, up to 30 mg/L or more (Lee *et al.* 2021). However, the anthropogenic sources could elevate the fluoride concentration in water bodies up to 1,000 s mg/L (Lee *et al.* 2021). Fluoride has two opposite effects on human beings; 1.5 mg/L of fluoride is advantageous for bones and tooth health; however, higher concentrations of fluoride impose a negative influence on human health, including weak bones, enamel fluorosis, arthritis, cancers, infertilities, brain damage, and thyroid disorders (Aoudj *et al.* 2015; Alhassan *et al.* 2021; Halpegama *et al.* 2021). Hence, the World Health Organization (WHO) recommends that fluoride concentrations in drinking water are not exceeding 1.5 mg/L (Garg & Sharma 2016; Mousazadeh *et al.* 2021).

Water pollution with fluoride is currently remediated via various treatment technologies, ranging from single to complex approaches, including membrane, ion exchange, chemical precipitation, and adsorption (Garg & Sharma 2016; Al-Hashimi *et al.* 2021; Hashim *et al.* 2021a; Mousazadeh *et al.* 2021). However, many of the utilised methods do not meet the economic or environmental requirements (Das & Nandi 2020; Emamjomeh *et al.* 2020a; Abdulhadi *et al.* 2021; Karaghool *et al.* 2022). For example, the literature criticises the elevated operational cost of membrane-based methods and the sensitivity of membranes for organic matter, resulting in fouling problems and, accordingly, the need for pre-treatment units (Guo *et al.* 2012; Hashim *et al.* 2017). Similarly, the main disadvantages of the adsorption approach are adsorbent depletion and the high manufacturing costs of particular adsorbents (Teixeira & Nunes 2011; Hashim *et al.* 2021b). The disadvantages of the remaining strategies were explored in depth in several studies (Singh *et al.* 2013, 2016; Jadhav *et al.* 2015). The addition of aluminium and calcium salts is another method that is commonly adopted for fluoride removal from contaminated water in a process known as precipitation-flocculation (Singh *et al.* 2016). Although this method is very popular, especially in developing economies, scientists define the large-scale generation of sludge and the high consumption of coagulants as serious disadvantages of this method (Singh *et al.* 2013; Castañeda *et al.* 2020).

Electrochemical methods, particularly the electrocoagulation method (EC), are currently used to remove several contaminants, like fluoride, from contaminated water. The EC method depends on the destabilisation of the pollutants through liberating coagulation ions from sacrificial electrodes under the effects of electrical current, usually direct current (Fekete *et al.* 2016; Moussa *et al.* 2017; Mena *et al.* 2019). The minimal production of sludge, cost-effectiveness, compacted

installation, and automobility are the key advantages of the EC method that attracted both researchers and industry (Hakizimana *et al.* 2017; Lu *et al.* 2021; Arab *et al.* 2022). Additionally, external chemical additions are not required to perform the EC reactors, which is a huge advantage in favour of the environment (Das & Nandi 2020; Shahedi *et al.* 2020). Aluminium electrodes are commonly used in electrocoagulation cells because this metal has low potential and is available worldwide at low costs (Hashim *et al.* 2020; Tahreen *et al.* 2021). The chemical reactions at anodes in aluminium-based reactors are (Emamjomeh *et al.* 2011; Hashim *et al.* 2017):

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

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_{3(solid)} + 3H^{+} \quad (2)$$

while the cathodes will be emitting the hydrogen gas as follows:

$$3H_2O + 3e^{-} \rightarrow 1.5H^{+} + 3OH^{-} \quad (3)$$

The predominant path for fluoride removal by the aluminium-based is the precipitation of fluoro-aluminium complex and chemical substitution reactions between fluoride ions and aluminium hydroxides (Emamjomeh *et al.* 2011). Figure 1 summarises the EC reactions.

The effectiveness of the EC process was demonstrated in a significant body of literature (Tian *et al.* 2018; Bian *et al.* 2019; Islam 2019; Kumari & Kumar 2021; Xu *et al.* 2021). For example, a number of studies (Thakur & Chauhan 2018; Abdulhadi *et al.* 2019; Hashim *et al.* 2019; Abdulrazzaq *et al.* 2021) applied the EC method to remove dyes from solutions, and the results of the studies indicated the EC method could remove more than 90% of dyes in a relatively short time. Furthermore, the removal of chemical oxygen demand (COD) and dyes was investigated by Emamjomeh *et al.* (2020b) using an aluminium-based EC method, and the results showed 97 and 92% of the dye and COD were removed by the EC unit. Additionally, a significant body of literature (Franco *et al.* 2017; Abdel-Aziz *et al.* 2020; Amarine *et al.* 2020) demonstrated the successful application of the EC method in the removal of nutrients, such as phosphate and nitrate from solutions.

However, the literature indicated some weaknesses in the EC method, such as a lack of reactor design. The common configuration of the EC cells is a simple rectangular container with parallel electrodes (Un *et al.* 2013). This study, therefore, uses a new space-saver EC reactor to remove fluoride from water. The new EC cell depends on a variable direction of flow to increase the detention time instead of the commonly used configurations that need relatively long reactors.

2. MATERIALS AND METHODS

2.1. Space-saver EC reactor

The new space-saver EC cell, arrangement of electrodes, and flow direction are shown in Figure 2. The electrodes of the new reactor were fitted to the walls of the Perspex container opposite to each other, leaving a short opening between each

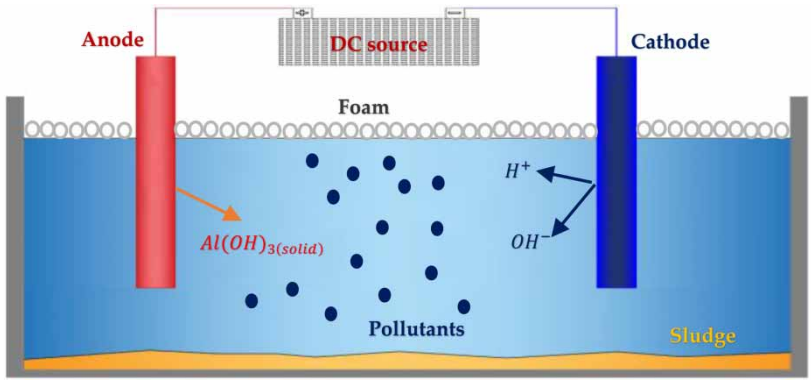


Figure 1 | The general configuration of EC reactors.

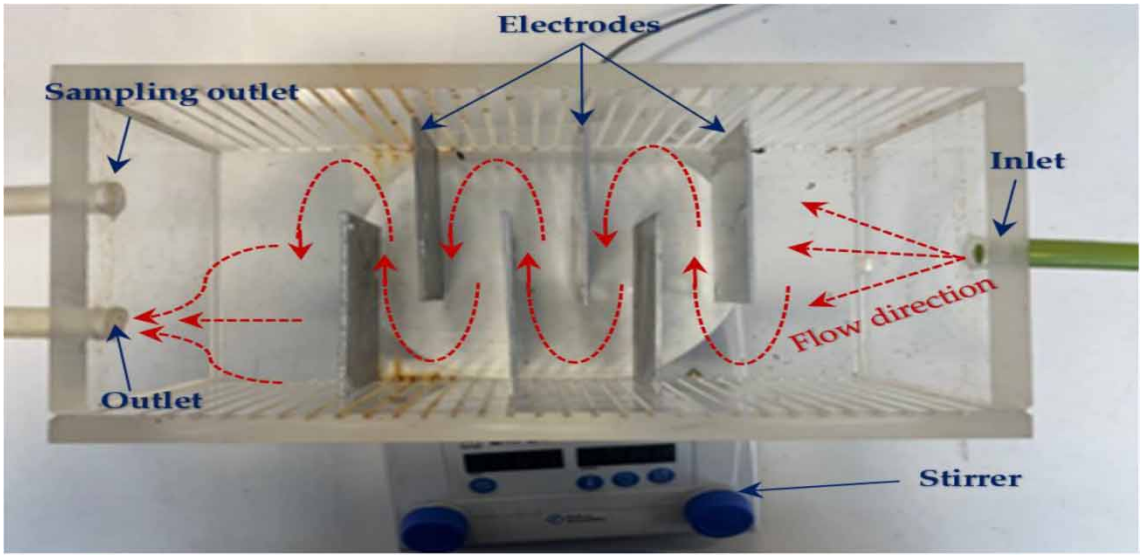


Figure 2 | The engineered EC reactor.

electrode and the opposite wall. This design enforces the water being treated to flow in a serpentine path, which increases the detention time by approximately 190%, as shown in the Results section. The new reactor has a total volume of 3,000 cm^3 and is supplied with six aluminium electrodes (8 cm × 5 cm, width and length, contacting water, respectively). The container has three 0.6 mm openings as a water inlet and outlet and a sampling outlet. The reactor is an open container to avoid the accumulation of hydrogen gas. A benchtop peristaltic water pump was connected to the reactor to flow water. A precision HQ DC source (30 volts) was used to provide the required current during the experimental work. A Fisher Scientific (Isotemp) stirrer was used to mix the water being treated to ensure a homogenous distribution of the dissolved aluminium electrodes.

2.2. Synthetic water and chemicals

All chemicals were with an analytical grade, provided by Merck, Germany, and used as supplied without further processing. Samples of fluoride contaminated water with an initial fluoride level of 20 mg/L were prepared a few minutes before the experiments to avoid any unwanted changes by adding 44.2 mg/L of NaF into deionised water and stirring the solution using a Fisher Scientific (Isotemp) stirrer. The conductivity of the solution was also measured and adjusted to 0.3 mS cm^{-1} before experiments using NaCl.

2.3. Experiments

2.3.1. Detention time

The first set of experiments was devoted to measuring the detention time in the new space-saver EC reactor (S-SECR) and comparing it with the traditional reactors. The experiments were carried out by filling the S-SECR with deionised water, then dyed water with red drain dye was flowed through the reactor at 100 mL/min until coloured water was received from the sampling outlet at the end reactor. The same procedures were repeated using the EC reactor but with a simple parallel arrangement of electrodes. By taking 5 mL water samples at 5-second intervals and measuring the colour concentration with a spectrophotometer, the change in the colour of effluent was carefully measured (Hach-Lang, DR-3900). The test was stopped when a colour change was detected, and that time was considered the detention time.

2.3.2. Fluoride removal

The electrolysing experiments were conducted using the S-SECR. In fluoride removal tests, three factors were considered: current density (δ) of 1.5, 2.5, and 3.5 mA cm^{-2}, pH of water of 4, 7, and 10, and the distance between electrodes (ϕ) of 5, 10, and 15 mm. The effect of these three factors on fluoride removability was examined separately in three phases. The first phases included the implementation of three levels of δ (1.5, 2.5, and 3.5 mA cm^{-2}) for 30 min and constant pH (4) and ϕ (10 mm). Once the solution left the S-SECR, it was immediately filtered using a 0.25 μm Whatman filter (purchased

from Merck, Germany) then tested for the fluoride concentration using DR-3900 spectrophotometer and fluoride cuvette test LCK-323. Dilution was used when the residual fluoride concentration was more than the capacity of the LCK-323 cuvettes (2.5 mg/L). To remove debris from the electrodes, electrodes have been cleaned with a strong acid (HCl acid) and rinsed with deionised water between each subsequent treatment.

The same procedures were used to examine the effect of pH and ϕ, keeping the previously tested factor at the optimum value. Equation (4) was adopted to calculate the removal efficiency of fluoride.

$$R(\%) = \frac{(\text{Initial fluoride concentration} - \text{Residual fluoride concentration})}{\text{Initial fluoride concentration}} \times 100 \quad (4)$$

A square metal sample (0.5 × 0.5 cm) was taken from both fresh and used aluminium anodes and examined by implementing scanning electron microscopy (SEM) to investigate the changes in the surfaces of these samples (morphological study).

Finally, the operating cost of the S-SECR was estimated in Iraqi dinars (IQD) per m^3 of water, using the equation below (Mena *et al.* 2019):

$$\text{Operating cost (IQD m}^{-3}) = \text{metal unit price} \times \text{electrodes consumption} + \text{power unit price} \times \text{power consumption} \quad (5)$$

The consumed power was measured by the voltage, current, and electrolysing time, as shown below:

$$\text{Power usage (kWh m}^{-3}) = \frac{\text{Average voltage} \times \text{applied current} \times \text{treatment time}}{\text{water volume}} \quad (6)$$

A precise analytical balance (4 digits accuracy – Ohaus) was used to calculate the consumed mass of the aluminium electrodes by weighting the anode before and after the electrolysing process.

3. RESULTS

3.1. Fluoride removal experiments

The influence of δ on the removability of fluoride from the water was studied at 1.5, 2.5, and 3.5 mA cm^{-2} for half an hour. To maintain identical experimental conditions for all tests, both the level of pH and ϕ were kept at 4 and 10 mm, respectively. The outcomes of the δ tests are shown in Figure 3. Although all applied values of δ met the guideline of the WHO for fluoride in drinking water (not exceeding 1.5 mg/L) after 30 min, the higher the applied δ, the shorter electrolysing time. The needed time to achieve WHO's guideline at δ of 1.5, 2.5, and 3.5 mA cm^{-2} was 15, 20, and 25 min, respectively. The initial results initially indicate δ of 3.5 mA cm^{-2} could be the optimum value for fluoride removal by S-SECR. However, the results of Figure 4 highlight the negative effects of δ on the cost-effectiveness of the S-SECR, the increase of δ maximised the power

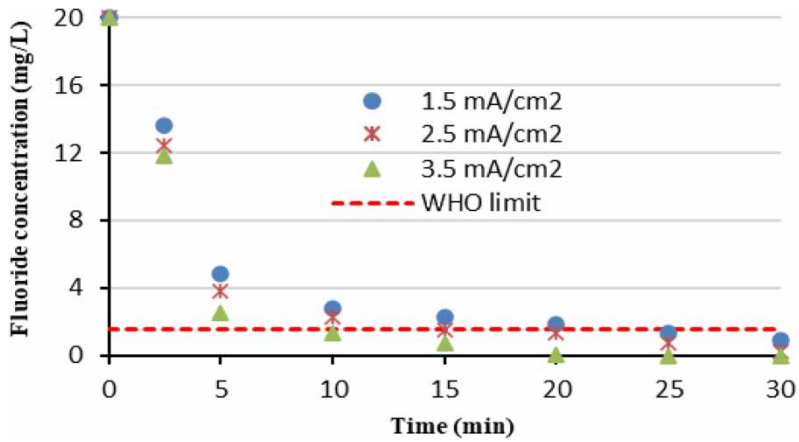


Figure 3 | The effects of δ on fluoride removal using S-SECR.

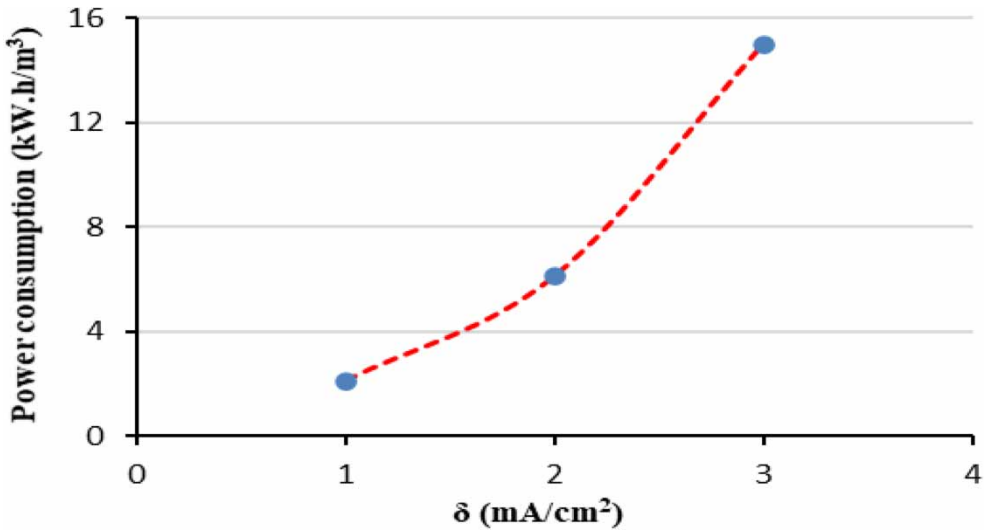


Figure 4 | The effects of δ on the power consumption of S-SECR.

consumption. Hence, a value of 2.5 mA cm^{-2} could be chosen as the optimal current for achieving reliable fluoride treatment while consuming a fair quantity of electricity.

The reason for the improvement in the removal with the rise in the δ might be related, as proved by previous studies (Palahouane *et al.* 2015; Alimohammadi *et al.* 2019; Sandoval *et al.* 2021), to the accelerated dissolution of the anodes, which accelerates the removal of contaminants.

Another set of trials was done to examine the effects of water pH on the removability of fluoride by the engineered reactor. In this set, the pH value was changed from 4 to 7 and 10, keeping δ and ϕ at 2.5 mA cm^{-2} and 10 mm, respectively. All experiments were run for 30 min. The outcomes of pH tests are presented in Figure 5, which revealed a minor difference between fluoride removal at pH of 4 and 7, but a significant difference was noticed between fluoride removal at pH of 4 and 10. The needed time to meet the WHO's guidelines at pH of 4 and 7 was 15 and 20 min, respectively. However, 30 min was not sufficient to meet the WHO's guidelines for fluoride in potable water at a pH of 10. The variation in fluoride removal with the initial water pH is related to the amphoteric properties of aluminium hydroxides, as proved in the previous studies

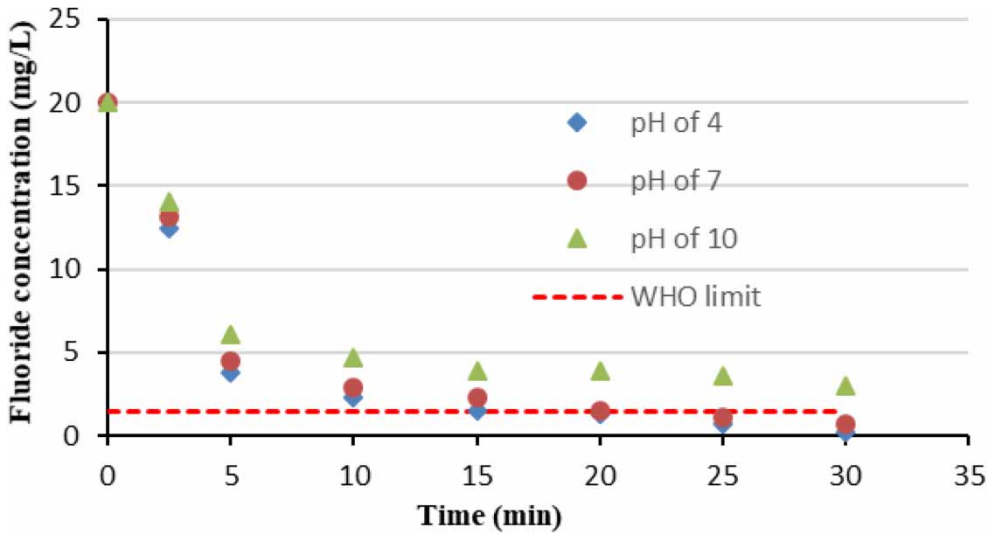


Figure 5 | The effect of pH on fluoride removal using S-SECR.

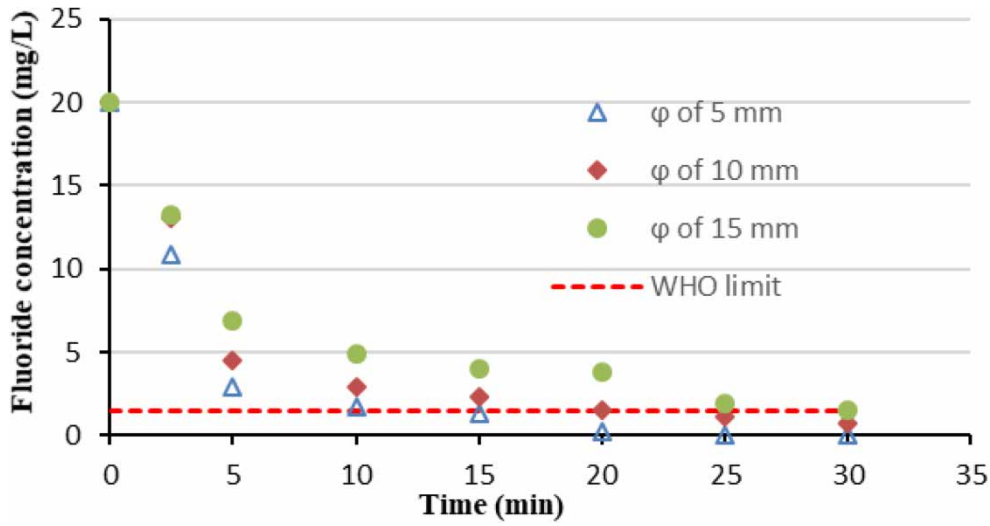


Figure 6 | Effects of ϕ on fluoride removal using S-SECR.

(Un *et al.* 2013). At high pH, the formed aluminium species have negative charges, which minimises fluoride adsorption by the fresh coagulants. Oppositely, the formed aluminium species in slightly acidic and neutral pH values have positive charges, such as $Al(OH)_3$, which improve fluoride removal. Although pH 4 achieved better removal efficiency than pH 7, the latter could be the best value in favour of the environment due to eliminating the need for acids.

The third tested factor was the ϕ that was conducted at three distances, 5, 10, and 15 mm, using the best values of pH (7) and δ (2.5 mA cm^{-2}) from the previous two experiments. Widening the space between electrodes maximises resistance for electricity flow inside the reactor, which leads to a drop in pollutants removal (Palahouane *et al.* 2015; Sandoval *et al.* 2021). The obtained results in this study, see Figure 6, approved this fact. According to Figure 5, the remaining fluoride in water decreased from 20 to the WHO's guideline after 15 and 20 min at ϕ of 5 and 10 mm, but 30 min were not enough to meet the WHO's limitations at ϕ of 15 mm (fluoride concentration was 1.54 mg/L). Thus, ϕ of 5 mm could be the most suitable value for this study in favour of both treatment time and fluoride removal.

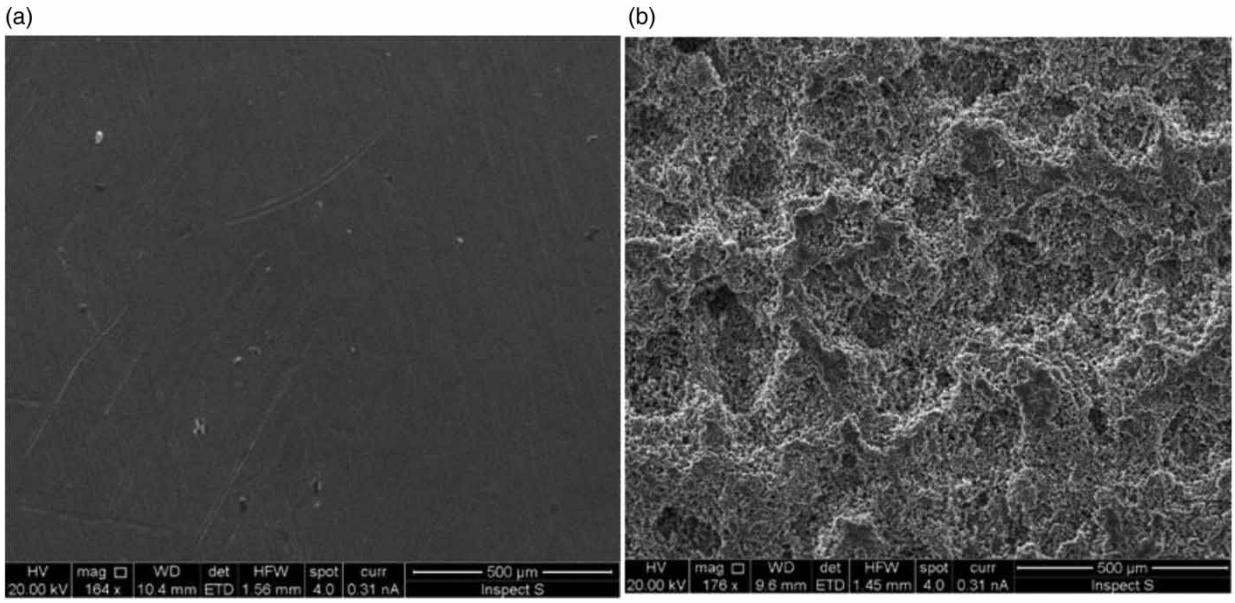


Figure 7 | SEM images of (a) fresh anode and (b) electrolysed anode.

The fluoride removal experiments indicated that the best performance of the S-SECR is achievable at pH, ϕ, and δ of 5 mm, 4 and 2.5 mA cm^{-2}, respectively. Therefore, these values were adopted in operating cost calculations.

3.2. Operating cost and morphological studies

The cost of operating the S-SECR was determined by implementing the experimental conditions that achieved the best fluoride removal. The corresponding voltage to the used δ of 2.5 mA cm^{-2} was recorded directly from the screen of the rectifier, and the treatment time was 15 min. The average power consumption was 5.03 kWh m^{-3}. The prices of electricity and metal were calculated according to the local market of Iraq in 2022, which were 348 IQD kWh^{-1} for electricity and 4,348 IQD/1.0 kg of aluminium.

The cost of operating the S-SECR was calculated using Equation (5). The latter showed the minimum operating cost was 501.44 IQD m^{-3}, equivalent to 0.346 USD m^{-3}. This cost is comparable to the stated charges in scientific research, like 0.358 USD m^{-3} (Thakur *et al.* 2019) and 0.354 USD m^{-3} (Thakur & Mondal 2016).

Finally, samples of fresh and used anodes (in the electrocoagulation process) were examined using scanning electron microscopy (SEM) to identify morphological changes. The surface of the fresh anode was smooth, with a few tiny dents and scratches that might result from the manufacturing of electrodes, see Figure 7(a). Oppositely, the surface of the used anode in the electrocoagulation process showed a significant number of dents and biting effects that prove the production of aluminium ions; thereby, it demonstrates the occurrence of electrolysing process (Figure 7(b)).

Sensors could be used to make the suggested EC unit a smart one by monitoring the influent and effluent concentrations of fluoride. Microwave sensors could be an effective option due to their proven efficiency in monitoring pollutants in solutions (Ryecroft *et al.* 2019).

4. CONCLUSION

The results showed the remaining fluoride concentration in treated water using the S-SECR was within the WHO's guideline for fluoride concentration in potable water of 1.5 mg/L. The best removal for fluoride was obtained at pH of 7, ϕ of 5 mm, and δ of 2.5 mA cm^{-2}, with an operating cost of 501.44 IQD m^{-3} (0.346 USD m^{-3}) and power consumption of 5.03 kWh m^{-3}. The SEM images showed significant changes in the surfaces of the anodes after the electrolysing process.

Generally, the best work of the S-SECR concerning fluoride removal could be attained by reasonably maximising the value of the electric current, minimising the gap between the electrodes, and using a neutral level of water pH. The results obtained here could be used as preliminary evidence of the ability of S-SECR to remediate water from elevated fluoride concentrations (similar to those concentrations in groundwater and surface water).

There is still scope for more experiments to examine the work of the S-SECR concerning other pollutants removals, such as heavy metals, from water or wastewater. Additionally, more experiments could be conducted to examine the effects of other factors, such as water temperature, on the performance of the S-SECR.

FUNDING

This research received no specific grant from any funding agency, commercial or not-for-profit sectors.

CONFLICTS OF INTEREST

The authors declared that they have no conflict of interest.

DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

REFERENCES

Abdel-Aziz, M., El-Ashtoukhy, E. Z., Zoromba, M. S., Bassyouni, M. & Sedahmed, G. 2020 Removal of nitrates from water by electrocoagulation using a cell with horizontally oriented Al serpentine tube anode. *Journal of Industrial and Engineering Chemistry* **82**, 105–112.

Abdulhadi, B. A., Kot, P., Hashim, K. S., Shaw, A. & Khaddar, R. A. 2019 Influence of current density and electrodes spacing on reactive red 120 dye removal from dyed water using electrocoagulation/electroflotation (EC/EF) process. In: *Proceedings of the First International Conference on Civil and Environmental Engineering Technologies (ICCEET)*, University of Kufa, Iraq, pp. 12–22.

Abdulhadi, B., Kot, P., Hashim, K., Shaw, A., Muradov, M. & Al-Khaddar, R. 2021 Continuous-flow electrocoagulation (EC) process for iron removal from water: experimental, statistical and economic study. *Science of The Total Environment* **760**, 1–16. https://doi.org/10.1016/j.scitotenv.2020.143417.

Abdulrazzaq, N. N., Al-Sabbagh, B. H. & Shanshool, H. A. 2021 Coupling of electrocoagulation and microflotation for the removal of textile dyes from aqueous solutions. *Journal of Water Process Engineering* **40**, 101906.

Al-Hashimi, O., Hashim, K., Loffill, E., Marolt Čebašek, T., Nakouti, I., Faisal, A. A. & Al-Ansari, N. 2021 A comprehensive review for groundwater contamination and remediation: occurrence, migration and adsorption modelling. *Molecules* **26**, 5913. https://doi.org/10.3390/molecules26195913.

Alhassan, S. I., Huang, L., He, Y., Yan, L., Wu, B. & Wang, H. 2021 Fluoride removal from water using alumina and aluminum-based composites: a comprehensive review of progress. *Critical Reviews in Environmental Science and Technology* **51**, 2051–2085.

Alimohammadi, M., Mesdaghinia, A., Shayesteh, M., Mansoorian, H. & Khanjani, N. 2019 The efficiency of the electrocoagulation process in reducing fluoride: application of inductive alternating current and polarity inverter. *International Journal of Environmental Science and Technology* **16**, 8239–8254.

Amarine, M., Lekhlif, B. & Echaabi, J. 2020 Nitrate removal from groundwater in Casablanca region (Morocco) by electrocoagulation. *Groundwater for Sustainable Development* **11**, 100452.

Aoudj, S., Khelifa, A., Drouiche, N., Belkada, R. & Miroud, D. 2015 Simultaneous removal of chromium (VI) and fluoride by electrocoagulation–electroflotation: application of a hybrid Fe-Al anode. *Chemical Engineering Journal* **267**, 153–162.

Arab, M., Faramarz, M. G. & Hashim, K. 2022 Applications of computational and statistical models for optimising the electrochemical removal of cephalexin antibiotic from water. *Water* **14**, 344–359. https://doi.org/10.3390/w14030344.

Arif, M., Hussain, J., Hussain, I. & Kumar, S. 2013 An assessment of fluoride concentration in groundwater and risk on health of north part of Nagaur District, Rajasthan, India. *World Applied Sciences Journal* **24**, 146–153. doi:10.5829/idosi.wasj.2013.24.02.993.

Bian, Y., Ge, Z., Albano, C., Lobo, F. L. & Ren, Z. J. 2019 Oily bilge water treatment using DC/AC powered electrocoagulation. *Environmental Science: Water Research & Technology* **5**, 1654–1660.

Castañeda, L. F., Coreño, O., Nava, J. L. & Carreño, G. 2020 Removal of fluoride and hydrated silica from underground water by electrocoagulation in a flow channel reactor. *Chemosphere* **244**, 125417.

Das, D. & Nandi, B. K. 2020 Simultaneous removal of fluoride and Fe (II) ions from drinking water by electrocoagulation. *Journal of Environmental Chemical Engineering* **8**, 103643.

Emamjomeh, M. M., Sivakumar, M. & Varyani, A. S. 2011 Analysis and the understanding of fluoride removal mechanisms by an electrocoagulation/flotation (ECF) process. *Desalination* **275**, 102–106.

Emamjomeh, M. M., Mousazadeh, M., Mokhtari, N., Jamali, H. A., Makkiabadi, M., Naghdali, Z., Hashim, K. S. & Ghanbari, R. 2020a Simultaneous removal of phenol and linear alkylbenzene sulfonate from automotive service station wastewater: optimisation of coupled electrochemical and physical processes. *Separation Science and Technology* **55**, 3184–3194. doi:10.1080/01496395.2019.1675703.

Emamjomeh, M. M., Kakavand, S., Jamali, H. A., Alizadeh, S. M., Safdari, M., Mousavi, S. E. S., Hashim, K. S. & Mousazade, M. 2020b The treatment of printing and packaging wastewater by electrocoagulation–flotation: the simultaneous efficacy of critical parameters and economics. *Desalination and Water Treatment* **205**, 161–174. https://doi.org/10.5004/dwt.2020.26339.

Fekete, É., Lengyel, B. & Cserfalvi, T. 2016 Electrocoagulation: an electrochemical process for water clarification. *Journal of Electrochemical Science and Engineering* **6**, 57–65.

Franco, D., Lee, J., Arbelaez, S., Cohen, N. & Kim, J.-Y. 2017 Removal of phosphate from surface and wastewater via electrocoagulation. *Ecological Engineering* **108**, 589–596.

Garg, U. K. & Sharma, C. 2016 Electrocoagulation: promising technology for removal of fluoride from drinking water–a review. In: *Proceedings of the Biological Forum-An International Journal*, pp. 248–254.

Guo, W., Ngo, H.-H. & Li, J. 2012 A mini-review on membrane fouling. *Bioresource Technology* **122**, 27–34.

Hakizimana, J. N., Gourich, B., Chafi, M., Stiriba, Y., Vial, C., Drogui, P. & Naja, J. 2017 Electrocoagulation process in water treatment: a review of electrocoagulation modeling approaches. *Desalination* **404**, 1–21.

Halpegama, J., Heenkenda, K., Wu, Z., Nanayakkara, K., Rajapakse, R., Bandara, A., Herath, A. C., Chen, X. & Weerasooriya, R. 2021 Concurrent removal of hardness and fluoride in water by monopolar electrocoagulation. *Journal of Environmental Chemical Engineering* **9**, 106105.

Hashim, K. S., Shaw, A., Al Khaddar, R., Pedrola, M. O. & Phipps, D. 2017 Energy efficient electrocoagulation using a new flow column reactor to remove nitrate from drinking water – experimental, statistical, and economic approach. *Journal of Environmental Management* **196**, 224–233. doi:10.1016/j.jenvman.2017.03.017.

Hashim, K. S., Hussein, A. H., Zubaidi, S. L., Kot, P., Kraidi, L., Alkhaddar, R., Shaw, A. & Alwash, R. 2019 Effect of initial pH value on the removal of reactive black dye from water by electrocoagulation (EC) method. In: *Proceedings of the 2nd International Scientific Conference*, Al-Qadisiyah University, Iraq, pp. 12–22.

Hashim, K., Kot, P., Zubaid, S., Alwash, R., Al Khaddar, R., Shaw, A., Al-Jumeily, D. & Aljefery, M. 2020 Energy efficient electrocoagulation using baffle-plates electrodes for efficient *Escherichia coli* removal from wastewater. *Journal of Water Process Engineering* **33**, 101079–101086. https://doi.org/10.1016/j.jwpe.2019.101079.

Hashim, K. S., Shaw, A., AlKhaddar, R., Kot, P. & Al-Shamma'a, A. 2021a Water purification from metal ions in the presence of organic matter using electromagnetic radiation-assisted treatment. *Journal of Cleaner Production* **280**, 1–17. https://doi.org/10.1016/j.jclepro.2020.124427.

Hashim, K. S., Ewadh, H. M., Muhsin, A. A., Zubaidi, S. L., Kot, P., Muradov, M., Aljefery, M. & Al-Khaddar, R. 2021b Phosphate removal from water using bottom ash: adsorption performance, coexisting anions and modelling studies. *Water Science and Technology* **83**, 77–89. https://doi.org/10.2166/wst.2020.561.

Islam, S. 2019 Electrocoagulation (EC) technology for wastewater treatment and pollutants removal. *Sustainable Water Resources Management* **5**, 359–380.

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. *Journal of Environmental Management* **162**, 306–325. https://doi.org/10.1016/j.jenvman.2015.07.020.

Karaghool, H. A. K., Hashim, K., Kot, P. & Muradov, M. 2022 Preliminary studies of methylene blue remotion from aqueous solutions by *Ocimum basilicum*. *Environments* **9**, 1–17. https://doi.org/10.3390/environments9020017.

Kumari, S. & Kumar, R. N. 2021 River water treatment using electrocoagulation for removal of acetaminophen and natural organic matter. *Chemosphere* **273**, 128571.

Lee, J.-I., Kang, J.-K., Hong, S.-H., Lee, C.-G., Jeong, S. & Park, S.-J. 2021 Thermally treated *Mytilus coruscus* shells for fluoride removal and their adsorption mechanism. *Chemosphere* **263**, 128328.

Lu, J., Zhang, P. & Li, J. 2021 Electrocoagulation technology for water purification: an update review on reactor design and some newly concerned pollutants removal. *Journal of Environmental Management* **296**, 113259.

Mena, V., Betancor-Abreu, A., González, S., Delgado, S., Souto, R. & Santana, J. 2019 Fluoride removal from natural volcanic underground water by an electrocoagulation process: parametric and cost evaluations. *Journal of Environmental Management* **246**, 472–483.

Mousazadeh, M., Alizadeh, S., Frontistis, Z., Kabdaşlı, I., Karamati Niaragh, E., Al Qodah, Z., Naghdali, Z., Mahmoud, A. E. D., Sandoval, M. A. & Butler, E. 2021 Electrocoagulation as a promising defluoridation technology from water: a review of state of the art of removal mechanisms and performance trends. *Water* **13**, 656.

Moussa, D. T., El-Naas, M. H., Nasser, M. & Al-Marri, M. J. 2017 A comprehensive review of electrocoagulation for water treatment: potentials and challenges. *Journal of Environmental Management* **186**, 24–41.

Palahouane, B., Drouiche, N., Aoudj, S. & Bensadok, K. 2015 Cost-effective electrocoagulation process for the remediation of fluoride from pretreated photovoltaic wastewater. *Journal of Industrial and Engineering Chemistry* **22**, 127–131.

Ryecroft, S. P., Shaw, A., Fergus, P., Kot, P., Hashim, K. & Conways, L. 2019 A novel gesomin detection method based on microwave spectroscopy. In: *Proceedings of the 12th International Conference on Developments in ESystems Engineering (DeSE)*, Kazan, Russia, pp. 429–433.

Sandoval, M. A., Fuentes, R., Nava, J. L., Coreño, O., Li, Y. & Hernández, J. H. 2019 Simultaneous removal of fluoride and arsenic from groundwater by electrocoagulation using a filter-press flow reactor with a three-cell stack. *Separation and Purification Technology* **208**, 208–216.

Sandoval, M. A., Fuentes, R., Thiam, A. & Salazar, R. 2021 Arsenic and fluoride removal by electrocoagulation process: a general review. *Science of The Total Environment* **753**, 142108.

Shahedi, A., Darban, A., Taghipour, F. & Jamshidi-Zanjani, A. 2020 A review on industrial wastewater treatment via electrocoagulation processes. *Current Opinion in Electrochemistry* **22**, 154–169.

Singh, K., Lataye, D. H., Wasewar, K. L. & Yoo, C. K. 2013 Removal of fluoride from aqueous solution: status and techniques. *Desalination and Water Treatment* **51**, 3233–3247. doi:10.1080/19443994.2012.749036.

Singh, J., Singh, P. & Singh, A. 2016 Fluoride ions vs removal technologies: a study. *Arabian Journal of Chemistry* **9**, 815–824. https://doi.org/10.1016/j.arabjc.2014.06.005.

Tahreen, A., Jami, M. S., Ali, F., Yasin, N. M. F. M. & Ngabura, M. 2021 Promising potential of electro-coagulation process for effective treatment of biotreated palm oil mill effluents. *Pollution* **7**, 617–632.

Teixeira, M. R. & Nunes, L. M. 2011 The impact of natural organic matter seasonal variations in drinking water quality. *Desalination and Water Treatment* **36**, 344–353. doi:10.5004/dwt.2011.2524.

Thakur, S. & Chauhan, M. 2018 Treatment of dye wastewater from textile industry by electrocoagulation and Fenton oxidation: a review. *Water Quality Management* **1**, 117–129.

Thakur, L. S. & Mondal, P. 2016 Techno-economic evaluation of simultaneous arsenic and fluoride removal from synthetic groundwater by electrocoagulation process: optimisation through response surface methodology. *Desalination and Water Treatment* **57**, 28847–28863.

Thakur, L. S., Goyal, H. & Mondal, P. 2019 Simultaneous removal of arsenic and fluoride from synthetic solution through continuous electrocoagulation: operating cost and sludge utilisation. *Journal of Environmental Chemical Engineering* **7**, 102829.

Tian, Y., He, W., Liang, D., Yang, W., Logan, B. E. & Ren, N. 2018 Effective phosphate removal for advanced water treatment using low energy, migration electric-field assisted electrocoagulation. *Water Research* **138**, 129–136.

Un, U. T., Koparal, A. S. & Bakir Ogutveren, U. 2013 Fluoride removal from water and wastewater with a bach cylindrical electrode using electrocoagulation. *Chemical Engineering Journal* **223**, 110–115. doi:10.1016/j.cej.2013.02.126.

Xu, J., Du, X., Zhao, W., Wang, Z., Lu, X., Zhu, L., Wang, Z. & Liang, H. 2021 Roofing rainwater cleaner production using pilot-scale electrocoagulation coupled with a gravity-driven membrane bioreactor (EC-GDMBR): water treatment and energy efficiency. *Journal of Cleaner Production* **314**, 128055.

First received 6 March 2022; accepted in revised form 8 April 2022. Available online 21 April 2022