

Article

Minimizing the Fluoride Load in Water Using the Electrocoagulation Method: An Experimental Approach

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Abstract: The abundant presence of fluoride (F^-) in surface water bodies is an environmental concern because of its effects on human health; medical reports confirmed that fluoride intake above 1.5 mg/L leads to many health complications, including but not limited to weak bones and enamel fluorosis. Thus, the World Health Organisation (WHO) defines 1.20 mg/L as the maximum permissible F^- concentration in drinking water. The electrocoagulation method (EC) is globally practised to remove many pollutants from water due to its cost-effectiveness, safety, and ease of use. However, EC has some drawbacks, such as the lack of reactors' design. In this study, a new EC reactor, which uses four drilled aluminium electrodes and a variant cross-section section container, was designed and used to remove F^- from water. The design of the new EC eliminated the need for water mixers. The ability of the new EC unit to remove F^- from synthetic water was evaluated at different current densities (CD) (1–3 mA/cm²), electrode distances (ELD) (5–15 mm), pH of the solution (pHoS) (4–10), and initial F^- concentrations (IFC) (5–20 mg/L). The outcomes of this study prove that the new reactor could remove as much as 98.3% of 20 mg/l of F^- at CD, ELD, pHoS, and IFC of 2 mA/cm², 5 mm, and 4 and 10 mg/L, respectively.

Keywords: electrocoagulation; fluoride removal; iron electrodes; optimization; RSM



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1. Introduction

The literature and technical reports rank fluorine as the 17th most abundant element in nature because it represents up to 0.6% of the crust of the Earth planet [1]. For example, the available literature has indicated that some natural formations, such as fluorspar, contain elevated fluoride concentrations (F^-). Therefore, weathering of such formations results in the enrichment of freshwater with high F^- concentrations, so this element is expected to be high in freshwater bodies [2]. In addition, F^- concentration in groundwater (aquifers), the primary source of drinking water in arid and semi-arid areas, is usually elevated due to water filtration through natural formations before reaching the aquifers [3,4]. The literature demonstrates that aquifers' average F^- concentration is 2–10 mg/L [5]. Besides the natural existence of F^- in freshwater, the industrial revolution led to a dramatic increase in F^- concentrations in the aquatic environment [6,7]. For instance, the effluents of semiconductors and aluminium industries are very rich in F^- , which is dumped into surface waters [8,9].

The recent shortage in freshwater due to climate change and population growth also contributed to intensifying the problem of water pollution [10,11]. F^- in freshwater at elevated concentrations is a reason for many diseases; it was reported that F^- intakes of 1.5 mg/L cause joint stiffness, weakness of bones, and enamel fluorosis [12]. Thus, the World Health Organisation (WHO) limited F^- concentration in drinking water to 1.20 mg/L [13].

A wide array of treatment methods are practised to minimize F^- concentration in drinking water to the permissible limit, 1.2 mg/L, including, but not limited to, membranes, precipitations, ion exchanges, and adsorption [14]. However, the majority of these methods suffer from drawbacks that limit their widespread use, especially in developing economies [15]. For instance, the technical reports demonstrated the high cost of membrane technologies and the need for pre-treatment arrangements to prevent fouling [16]. Adsorption methods also have some limitations, including the depletion of the adsorbents and the high production cost of some types of adsorbents [17]. Drawbacks of the other mentioned methods have been discussed in detail in some research works [18,19]. The need for efficient water treatment methods is becoming more urgent due to the severe shortage in freshwater availability and climate change.

In this study, the electrocoagulation method (EC) was adopted to difluoride water due to the unique merits of this method. EC is recognized as a cost-effective method that is safe, simple, and easy to perform [20,21]. More importantly, the EC is an eco-friendly method because it depends on the in situ generation of coagulants without the need for chemical additives; i.e., it does not produce secondary pollutants or toxic sludge [5]. These unique merits encouraged researchers and industry to adopt widespread use of the EC method to treat water and wastewater. For example, Ouaisa et al. [22] employed an EC cell shaving aluminium electrodes to remove chromium Cr(VI) from synthetic water. The best-obtained removal was 97% at a current density of $4.03 \text{ mA}\cdot\text{cm}^{-2}$ and initial pH of 3–6. Naje et al. [23] utilized a rotated bed EC unit (made from aluminium) to remove Imperon violet KB dye from textile wastewater. The results indicated that the best removal of dyes was about 98%, attained after 10 min at a current density of $4 \text{ mA}\cdot\text{cm}^{-2}$, and pH of about 5, and a water temperature of 25°C . Another study about removing *E. coli* from water using the EC method was conducted by Castro-Rios et al. [24]. This study used a 500 mL batch EC cell with aluminium electrodes to treat synthetic water samples containing 10^5 to 10^6 cfu/mL of *E. coli*. The outcomes of this investigation confirm that operating the EC cell at a current density of $2.27 \text{ mA}/\text{cm}^2$, an initial pH of 4, and 2.5 mg/L of Na_2SO_4 was enough to reduce the number of *E. coli* by 1.0- and 1.9-log after 40 min and 90 min, respectively.

There is a significant body of literature demonstrating the efficiency of the EC in removing a wide array of pollutants from water and wastewater within a short time, such as [25–29].

However, the EC is not free from drawbacks; some researchers highlighted a few weaknesses: (1) The high impact of solution pH and chemistry on the EC performance [30], and (2) no sufficient reactor designs, where the majority of the currently used EC reactors are simple rectangular, cylindrical or square in shape with parallelly installed electrodes [13]. Furthermore, the available models to simulate the performances of the EC reactors are still limited [31].

Thus, as a trial to enhance one of these drawbacks, this study presents a new design of the EC and uses it to remove F^- from synthetic water. The removal of F^- aims to achieve two goals: firstly, to prove the capability of the new reactor to perform as an EC unit, and secondly, to provide a new, safe, and cost-effective method to treat water. The details of the new EC reactor and the experimental procedures are explained in the next section.

2. Materials and Methods

2.1. Chemicals and Solution

The required chemicals to perform the experiments in this study, including NaF, NaOH, NaCl, and HCl, were imported from Merck, Darmstadt, Germany, and used as provided.

The synthetic solution was initially made by adding 442 mg of NaF to deionized water to have a concentrated stock solution with an F^- concentration of 200 mg/L. The weight of the NaF powder was accurately measured using a 4-digit scale (GRAM-FS, Gram Group, Barcelona, Spain). The solution was stirred using a magnetic stirrer (RS PRO Stirrer-SH4, Merck, Darmstadt, Germany) until the powder completely dissolved; then it was refrigerated and used later to prepare samples with lower initial F^- concentration (IFC) (from 5 to 20 mg/L). The initial pH of the solutions (pHoS) was changed to the required value (4 to 10) using either hydrochloric acid or sodium hydroxide, and the pHoS value was measured using a pocket meter (Model: HI 98130, Merck, Darmstadt, Germany). The initial electrical conductivity of the synthetic samples was maintained at 320 $\mu S/cm$ using sodium chloride. Both pHoS and electrical conductivity were measured using a pocket meter (Model: HI 98130).

2.2. New EC Reactor

A new benchtop EC reactor was designed and manufactured to reduce water mixers and minimize power consumption. The new EC has four drilled aluminium electrodes installed at the narrow reactor section. Each electrode is 6.5 cm in length and 4.2 cm in width and contains six vertical cuts (4.5 cm in length and 0.2 cm in width). The effective depth of water inside the reactor is 5.5 cm, which makes the submerged area of each electrode 33.2 cm^2 . The main body of the reactor, as shown in Figure 1, is made from Perspex. The shape of the reactor is 12 cm in width at the bigging of the reactor, narrowing in the middle to reach only 4 cm, then expanding again to 12 cm at the end of the reactor.

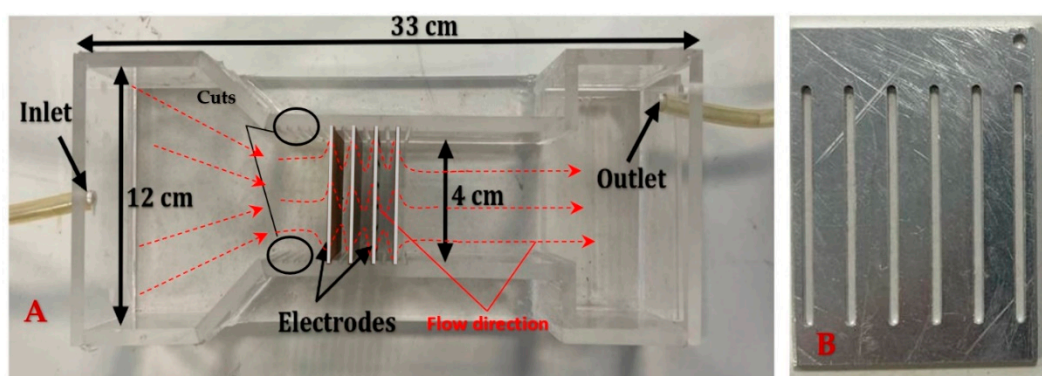


Figure 1. (A) The new EC reactor, (B) electrodes.

Additionally, 1 mm cuts were made in the sides of the reactor at the narrow section of the reactor (at each 5 mm of the length of the narrow section), which were used later to hold the electrodes at the required location (adjusting the electrode distances (ELD)). The purpose of narrowing the cross-sectional area in the middle of the reactor is to increase water velocity at this area. The presence of perforated electrodes at this critical area (high flow velocity) helps mix water effectively without the need for external water mixers because the water flows in convoluted paths, as shown in Figure 1. It must be mentioned that the effectiveness of perforated electrodes in mixing water was proved in the literature [32,33].

The electrodes were coupled to a DC source (HQ Power-30 V, Velleman Group, Gavere, Belgium), and a peristaltic water pump (Watson-504U, Gemini Equipment, Apeldoorn, Netherlands) was employed to circulate water through the new EC.

2.3. Experiments

Initially, the change in the flow velocity at the wide and narrow sections of the new EC was measured using the following Equation:

$$V = \frac{Q}{A} \quad (1)$$

where V , Q , and A are the flow velocity (m/sec), discharge (m³/sec), and the cross-sectional area of the reactor (at the required location), respectively.

Then, the F[−] removal experiments were started by flowing the polluted water samples through the new EC to be treated for 30 min at different pHoSs (4–10), current densities (CDs) (1–3 mA/cm²), ELDs (5–15 mm), IFCs (5–20 mg/L), and IFCs (from 5 to 20 mg/L).

The removal of F[−] was measured using a Hach–Lang spectrophotometer (DR-2800) and Hach–Lange F[−] cuvettes (LCK323) (after dilution in case the F[−] concentration is higher than the capacity of the cuvettes). Equation (2) was used to measure removal efficiency of F[−]:

$$R(\%) = \frac{(C_1 - C_2) \times 100\%}{C_1} \quad (2)$$

where C_1 and C_2 are the initial and residual F[−] concentrations (mg/L), respectively. After each experiment, the electrodes were taken out of the EC for cleaning with HCl (35%) and water before using them in the following experiments.

All experiments were initially conducted for 30 min following the average treatment time used in previous studies, such as the studies by Essadki et al. [34] and Hashim et al. [2]. Then, the optimum treatment time was experimentally determined.

Finally, for comparison purposes, a 5 litre water sample was collected from the Tigris River, Tikrit City, Iraq, on the 27 February 2022. The sample was collected using a plastic container and immediately transferred to the laboratory and enriched with fluoride to 10 mg/L. Then, it was treated using the new EC cell at optimum conditions obtained from the synthetic water samples.

All experiments were repeated three times to ensure the reliability of the results.

2.4. Operating Cost

The operating cost of the new EC was calculated using Equation (3) [35], which is suitable for laboratory scenarios because it does not include the labour, investment, and sludge handling costs. The power unit and metal prices were estimated according to the Iraq local markets.

$$\text{Operational cost} \left(\frac{\$}{m^3} \right) = \beta \times \text{Electrodes consumption} + \gamma \times \text{Energy consumption} \quad (3)$$

where β and γ are metal and power unit prices, respectively.

The power usage is calculated as follows:

$$\text{Power usage} \left(\frac{kWh}{m^3} \right) = \frac{\text{Voltage} \times \text{Current} \times \text{Time}}{\text{Volume of water}} \quad (4)$$

The electrodes consumption was measured as the difference in weight of electrodes before and after treatment. The weight difference was measured using a four-decimal weighing scale (FA2004B, Gram Group, Barcelona, Spain).

3. Results and Discussion

The experiments of F[−] removal from synthetic water were performed at several stages, which were:

3.1. Effects of CD on F[−] Removal

Synthetic water samples with 10 mg/L were flowed through the new EC for 30 min and subjected to three different CD values (1, 2, 3 mA·cm^{−2}). The ELD and pHoS were maintained at 10 and 4 mm, respectively.

The results of these experiments are depicted in Figure 2A, which shows a clear variation in the removal of F[−] from water with the change in the applied CD (between 1 and 3 mA/cm²). Generally, the removal of F[−] improved with the applied CD, reaching complete removal after 20 min at CD of 3 mA/cm². For CD values less than 3 mA/cm², the removal

of F^- did not reach complete removal. A significant body of studies explains the effects of CD on the removal of pollutants by the EC method [36,37]. The increase in the removal of contaminants with the rise in CD is due to the effect of the electric current density on the melting of metal ions from the anodes, which increases the removal efficiency. The results of this study are in good agreement with the literature [38,39].

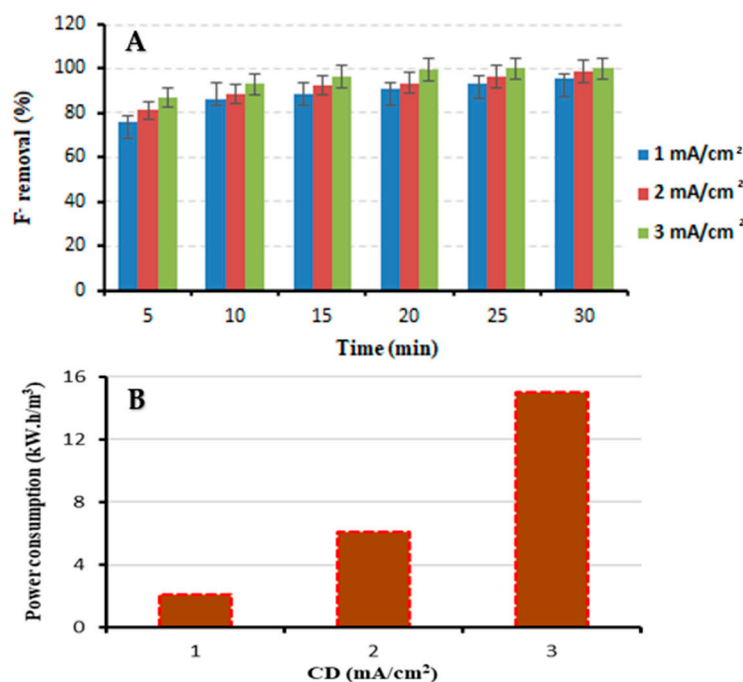


Figure 2. Effects of CD on (A) F^- removal and (B) power consumption.

Besides the positive effects of CD increase on F^- removal, there was a negative effect of CD on the performance of the EC, where it was noticed that increasing the CD amplified the power consumption, as shown in Figure 2B.

Therefore, a CD of 2 mA/cm² was adopted here as the best value because it achieves a good removal efficiency (92.4%) at a reasonable power consumption compared to a CD of 3 mA/cm².

3.2. Effects of pH_oS on F^- Removal

Based on the obtained results in Section 3.1, the pH_oS experiments were carried out at CD of 2 mA/cm² for 20 min, keeping the ELD constant at 10 mm. Three levels of pH_oS were considered in this investigation, namely 4, 7, and 11. Figure 3 illustrates the impact of pH_oS variations on F^- removal. It can be seen that the best removal of F^- (93.4%) was at acidic pH_oS (4), but beyond this value, the removal of F^- decreased slightly to 89.2% at pH_oS of 7 and to 80.1% at pH_oS of 11. Although pH_oS of 4 achieved the best removal of F^- , a pH_oS of 7 was adopted here to avoid the need for acidic addition, which has negative environmental impacts. This change in F^- removal with the pH_oS is because of the changes in the amphoteric properties of Al hydroxide. In the basic medium, the negatively charged aluminium species have low adsorption capacity for F^- , while in slightly acidic and neutral media, the predominant aluminium species is Al(OH)₃, which has a very good adsorption capacity for F^- [13]. The results of this part of the study agree with those in the literature [2,13].

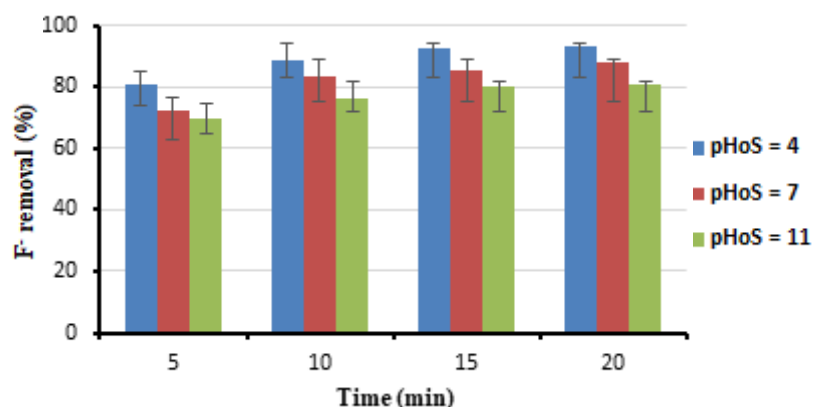


Figure 3. Effects of pHoS on F^- removal by the new EC.

3.3. Effects of ELD on F^- Removal

In this section, the effects of the ELD on the removal of F^- by the new EC was investigated using the best values of the DC and pHoS obtained from the above results. The contaminated water samples were treated in the EC at three levels of ELD, namely 5, 10, and 15 mm, for 20 min. The results of this investigation are shown in Figure 4, which indicates the negative impact of the long ELD on F^- removal. The best removal efficiency (98.3%) was achieved at the shortest ELD (5 mm); beyond this distance, the removal efficiency decreased to 81.3% at an ELD of 15 mm. The literature attributes this decrease to the drop in the electric current flow between the electrodes (decrease in electric field intensity), which decreases the melting of metal ions from the anodes and consequently decreases the removal efficiency [40,41]. The outcomes of this investigation encourage the authors to select the ELD of 5 mm to achieve the best removal efficiency of F^- .

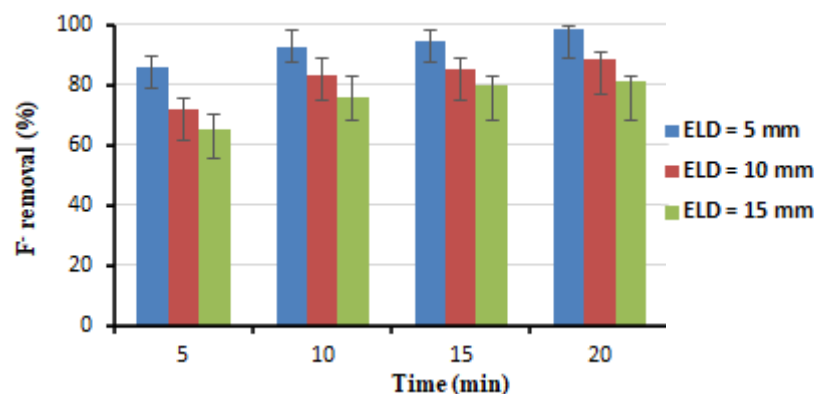


Figure 4. Effects of ELD on F^- removal by the new EC.

3.4. Effects of IFC on F^- Removal

The effect of pollutant concentration on the efficiency of the EC was studied for three concentrations of F^- , namely 5, 10, and 20 mg/L. The results of the investigation can be observed in Figure 5, which confirms that the removal efficiency of the high F^- concentration was less than the removal of low F^- concentrations. The relevant studies in the literature [42,43] relate the drop in the removal efficiency with the increase in F^- concentration to the fact that the high concentrations of F^- require more aluminium ions, which increase the required time to accomplish the complete removal.

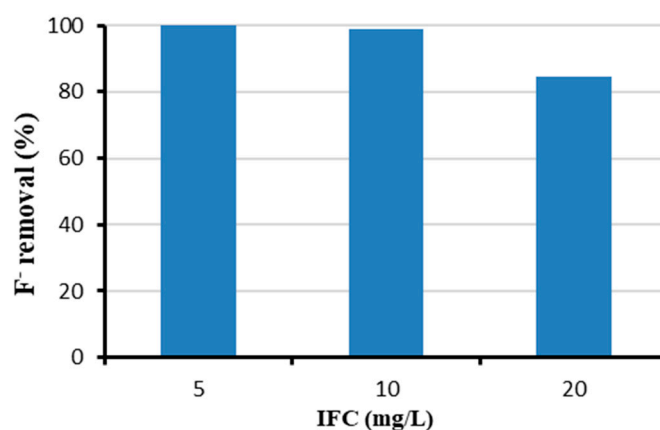


Figure 5. Effects of IFC on F⁻ removal by the new EC.

The achieved removal efficiency by the EC is comparable to those in the literature that used water stirrers [5,44], which confirms one of the most important advantages of the new EC. The new EC utilizes its container's design and electrodes, which increase the flow velocity at the centre of the reactor and force the water to flow in convoluted paths. According to Equation (1), the flow velocity at the narrow section of the reactor is 3 times the velocity at the wide section (the entrance of the reactor), and the velocity inside the vertical cuts in the electrodes is 12 times the flow velocity at the entrance to the reactor. Therefore, water being treated will be efficiently mixed without the need for mixers.

Finally, for the purposes of comparison, a natural water sample, collected from Tigris River, containing 10 mg/L, was treated at the optimum operating conditions (CD of 2 mA/cm², pHoS of 7, and ELD of 5 mm). The treatment process was performed until the complete removal of fluoride from the natural water sample was achieved. The results obtained from these experiments are presented in Figure 6, which shows that the progress of fluoride removal was much slower than that in the synthetic water.

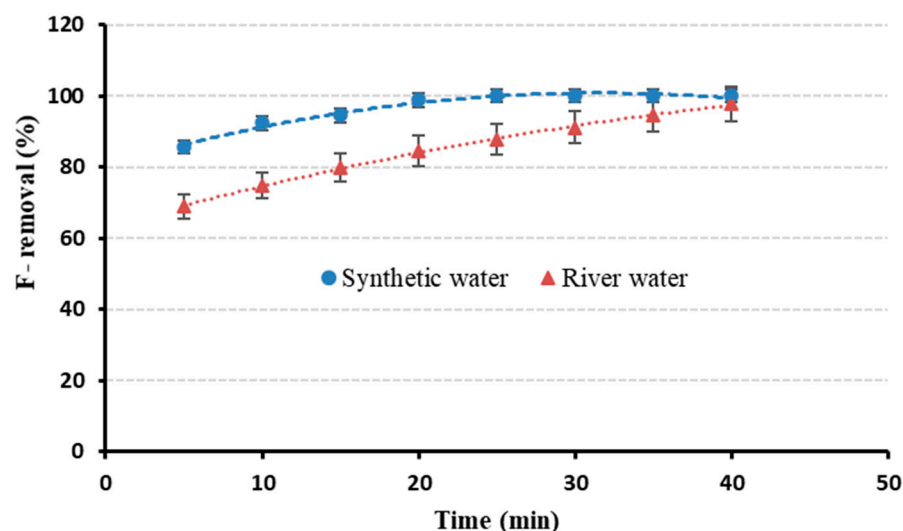


Figure 6. Comparison between fluoride removal from synthetic and river water samples.

3.5. Power Consumption and Operating Cost

The power consumption of the EC reactor during the removal of F⁻ was calculated using Equation (4) at the optimum operating conditions that achieved the best removal efficiency (98.3%), namely ELD of 5 mm, IFC of 10 mg/L, CD of 2 mA/cm², pHoS of 4, and treatment time of 20 min. The calculated power consumption was 4.06 kW·h/m³.

The calculated power consumption was then used, along with the consumed amount of the aluminium metal, to calculate the operating cost according to Equation (3). The prices

of electricity and aluminium were estimated according to the Iraqi markets in January 2022, which were 2.4 cents/kWh for electricity and 3.00 USD per 1.0 kg of aluminium. The calculated operating cost was 0.292 USD per m³ of treated water, which is cheaper than reported costs in the literature, such as 0.358 USD/m³ [45] and 0.354 USD/m³ [46], which are mainly attributed to the elimination of the need for stirrers. Bearing in mind that this cost is for laboratory units, which is much smaller than the field cost because the latter covers other expensive parameters, such as the labour and sludge-handling costs.

4. Conclusions

The presented study investigates the design and use of a new EC reactor to remove F⁻ from water. The obtained results demonstrate the following:

1. The new EC reactor can remediate water from F⁻, with an efficiency of 98.3%.
2. The new design of the reactor and the electrodes reduce the need for external water mixers, which in turn minimizes the power consumption.
3. The removal of F⁻ by the EC increases with the applied CD but decreases with the increase of pH_oS, ELD, and IFC.
4. The calculated operating cost of the new EC reactor was slightly cheaper in comparison with the traditional electrocoagulation reactor.

There is room for future applications of the new EC, such as using it to remove heavy metals, nitrate, and phosphate from water or wastewater.

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