

## Article

# Continuous Electrocoagulation for a Sustainable Water Treatment: Effects of Electrode Configuration, Electrical Connection Mode, and Polarity Reversal on Fluoride Removal

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**Abstract:** Water pollution in southern Tunisia, particularly in the mining basin of Gafsa, is primarily due to elevated levels of fluoride ions. This study focuses on removing fluoride from Metlaoui's tap water through a continuous electrocoagulation (EC) treatment. With a fluoride concentration of 3.5 mg·L<sup>-1</sup>, this water exhibits the highest fluoride levels in Gafsa's mining basin. The study investigates the impact of electrode configuration on fluoride removal from tap water through continuous electrocoagulation treatment. Configuring the electrodes perpendicular to the water flow improves the aluminum dissolution by electrocoagulation and the fluoride removal efficiency. Additionally, the study explores the effect of electrical connection modes on electrode performance, showing consistent fluoride removal yield under identical current densities and electrochemical cell numbers. Furthermore, the study examines cathodic deposit removal through polarity reversal, demonstrating its effectiveness in eliminating deposits and achieving high fluoride removal yields, especially with polarity reversal every minute. This method proves to be an efficient approach for a more sustainable fluorinated water treatment, eliminating cathodic deposits without the need for chemical or mechanical interventions, and without producing additional effluents or waste. The optimization of these parameters not only enhances fluoride removal efficiency, but also reduces energy consumption and operational costs, thereby promoting the sustainable management of energy and water resources.

**Keywords:** fluoride removal; electrode configuration; energy consumption; human health; polarity reversal; sustainable water treatment



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## 1. Introduction

The fluoride anion, commonly found in drinking water, plays a crucial role in human health. It can have many effects depending on its concentration. When it is present in concentrations ranging from 0.5 to 1.5 mg·L<sup>-1</sup>, fluoride contributes positively to the strengthening of bones and teeth.

When fluoride concentrations exceed 1.5 mg·L<sup>-1</sup>, individuals may face a variety of health issues. These encompass the following [1–3]:

- Dental fluorosis, marked by tooth discoloration and mottling, potentially affecting both aesthetics and functionality;

- Respiratory complications, such as irritation of the respiratory tract and breathing difficulties;
- Cardiovascular concerns, including hypertension and irregular heartbeats;
- Neurological disorders ranging from mild symptoms like dizziness to severe conditions such as nerve damage and cognitive decline, including Alzheimer's disease;
- Endocrine disturbances, which may result in imbalances in hormone levels, such as hyperglycemia and hyperthyroidism;
- Dermatological ailments such as rashes and skin irritation.

These health risks highlight the importance of monitoring and regulating fluoride levels in drinking water to mitigate potential adverse effects on public health [1–3]. According to the World Health Organization (WHO), the highest acceptable concentration in drinking water is  $1.5 \text{ mg}\cdot\text{L}^{-1}$  [4]. However, the concentration of fluoride ions in groundwaters can exceed this level [3,5–9]. Studies indicate that over 200 million people worldwide consume water with high fluoride concentrations, exceeding the limit permitted by the WHO [3,10,11]. Fluoride contamination worldwide is correlated with several factors, such as geology, hydrology, and anthropogenic activities in regions, highlighting the involvement of various natural and human factors. Africa records the highest number of affected countries with 38, followed by Asia with 28 affected countries, and Europe with 25 affected countries [3]. Tunisia is among the countries facing the issue of water fluoridation. In fact, waters in the south of Tunisia, especially in the Gafsa region, exhibit high concentrations of fluoride ions in the range of 2 to  $4 \text{ mg}\cdot\text{L}^{-1}$ , exceeding the standard set by the WHO [5,7,12]. Water contamination by fluoride has become a pressing global issue. In response, researchers are actively exploring and developing several techniques to address the challenge of fluoride removal. In recent years, many processes have been cited and used in fluoride removal, such as adsorption [13], ion-exchange resins [14], co-precipitation [15], membrane processes [16,17], and electrochemical methods such as electrodialysis and electrocoagulation (EC) [7,18–20]. Each of these methods offers distinct advantages and presents certain limitations. Adsorption techniques, employing a wide range of adsorbents and resins, have demonstrated high performance in fluoride removal. However, these methods often face competition from carbonates, phosphates, and sulfates for adsorption sites. Moreover, the regeneration of adsorbents and resins is costly and results in decreased performance over time with repeated regenerations [10,13,21,22].

Co-precipitation is a low-cost method utilizing calcium salts, but it necessitates high chemical usage, increases water hardness, and alters the taste of treated water [15,23,24].

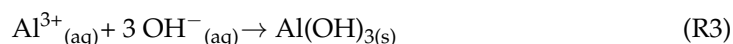
Membrane processes, particularly reverse osmosis and nanofiltration, are highly effective in fluoride removal and enable the simultaneous removal of various organic and inorganic pollutants, as well as suspended matter, without the use of chemicals. However, they come with high installation costs, significant water wastage in the reject stream, and substantial ongoing maintenance expenses for the membranes [25,26].

Electrocoagulation is another effective electrochemical method for fluoride removal that uses electrical current to dissolve metallic electrodes and generate coagulants in situ to remove fluoride ions. This process is relatively simple and environmentally friendly since it does not require the addition of chemicals. However, it needs regular electrode maintenance and replacement [9,27].

In this study, fluoride removal by electrocoagulation is addressed using aluminum electrodes. This process involves the electro-dissolution of aluminum anodes to produce metal cations ( $\text{Al}^{3+}$ ) (Reaction (1)) and the reduction of water at the cathode to produce hydroxide ions ( $\text{OH}^-$ ) and dihydrogen gas ( $\text{H}_2$ ) (Reaction (2)).

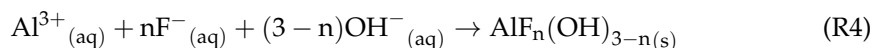


Aluminum cations and hydroxide ions react in solution to form solid flocs  $\text{Al}(\text{OH})_3$  (Reaction (3)).



Researchers have shown that fluoride removal by electrocoagulation is governed by two mechanisms [7,8,28]:

- The co-precipitation of fluoride with aluminum species (Reaction (4)).



- The adsorption of fluoride ions on the  $\text{Al}(\text{OH})_3$  flocs.

Previous studies on the fluoride removal mechanism by EC have suggested chemical adsorption through the substitution of hydroxide ions in  $\text{Al}(\text{OH})_3$  flocs with fluoride ions [8,28]. However, our research on the mechanism of fluoride removal via electrocoagulation and adsorption on flocs has shown the absence of such ion substitution reactions. Instead, the adsorption process is purely physical [7].

Electrocoagulation treatment has been widely studied in recent years to treat different pollutants using various types and configurations of reactors. Among these, batch and continuous reactors are prominent. In batch reactors, sacrificial electrodes are submerged in a specific volume of water, and an electric current is applied to induce coagulation and remove pollutants. This type of reactor is used for small-scale applications. It is widely used in laboratory studies and pilot-scale experiments [7,8,18,20].

Continuous flow reactors, as their name suggests, treat water continuously. Various reactor configurations can be achieved by adjusting reactor characteristics (volume, water inlet and outlet, residence time. . .) and electrode forms and arrangements (parallel, perpendicular, inclined. . .) [29,30].

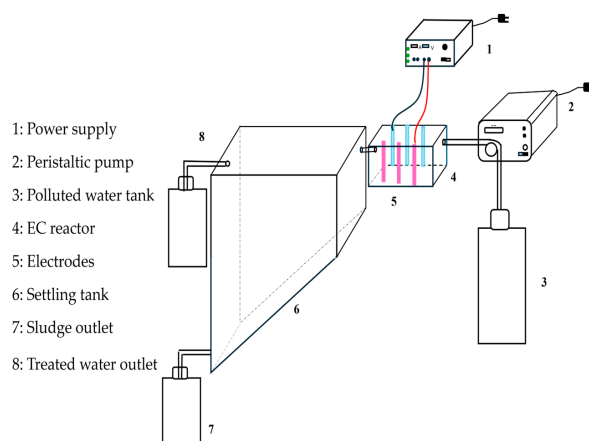
Research has often been carried out on batch electrocoagulation reactors [12,18,20,31]. Studies carried out with continuous reactors are not very developed. This study will deal with a continuous electrocoagulation reactor to treat tap water from Metlaoui, where the tap water is harmful for human health because of the high fluoride concentration higher than the WHO standard ( $1.5 \text{ mg} \cdot \text{L}^{-1}$ ). The investigation encompasses three main aspects. Firstly, it explores the impact of electrode positioning on metal dissolution and fluoride removal efficiency. Secondly, it delves into comparing different electrical connection modes—parallel monopolar (P-MP), bipolar (BP), and serial monopolar (S-MP)—using different parameters such as current, current density, number of electrodes, and number of electrochemical cells. Finally, to extend the treatment duration without interruptions for electrode cleaning and achieve a more sustainable process, the study investigates the effectiveness of electrode polarity reversal in eliminating cathodic deposits using various reversal durations.

## 2. Materials and Methods

### 2.1. Experimental Set-Up

The experimental set-up of continuous electrocoagulation used in this study is shown in Figure 1. It is composed of an electrocoagulation reactor with a volume (V) of 2.5 L that can hold up to 10 electrodes separated by an inter-electrode distance of 1.5 cm, and an active surface of  $105 \text{ cm}^2$ . The aluminum electrodes used in this study are positioned in two configurations: parallel or perpendicular to the water flow direction. In the case of perpendicular positioning, the electrodes were fixed alternately on the right (anode side) and left (cathode side) sides of the reactor, and vertically to the direction of flow. In the other case, electrodes were fixed parallel to the direction of water flow within the reactor, with a constant interelectrode distance of 1.5 cm. This interelectrode distance value was selected to facilitate water circulation while avoiding an excessive rise in ohmic drop. The two electrodes at the extremities were also positioned 1.5 cm away from the reactor's sides. The settling tank, with a capacity of 38 L, was characterized by a bottom inclined at a  $45^\circ$  angle to enhance the settling surface area. It was connected to two recovery tanks

for both sludge and treated water. Direct current was generated using a power supply (VOLTcraft HPS-13015, Conrad Electronic, Hirschau, Germany). Polluted tap water, which had a temperature ( $T$ ) of 20 °C, was supplied by a peristaltic pump (Fisher Scientific GP1100, Thermo Fisher Scientific, Waltham, MA, USA). A conductivity meter (METTLER TOLEDO, Columbus, OH, USA) was used to measure conductivity in this work.



**Figure 1.** Experimental set-up of the electrocoagulation unit.

## 2.2. Experimental Procedures

Various experiments were carried out in this study by varying the operating conditions of the continuous electrocoagulation process. We treated simulated Metlaoui tap water, prepared by dissolving various salts ( $\text{NaF}$ ,  $\text{CaCl}_2$ ,  $2\text{H}_2\text{O}$ ,  $\text{MgSO}_4$ ,  $\text{NaHCO}_3$ ,  $\text{KH}_2\text{PO}_4$ , and  $\text{CaSO}_4$ ) in deionized water. Table 1 shows the chemical composition of the natural and the synthetic water used in all experiments.

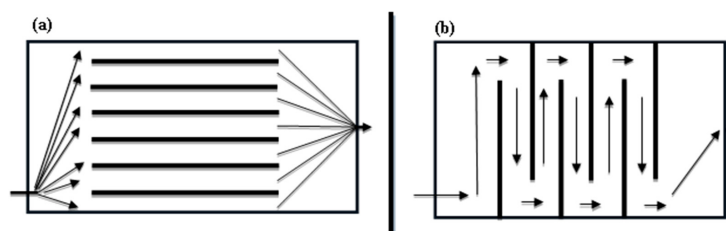
**Table 1.** Real and synthetic water composition.

|                                                      | Real Water Composition | Synthetic Water Composition |
|------------------------------------------------------|------------------------|-----------------------------|
| $\text{F}^-$ ( $\text{mg}\cdot\text{L}^{-1}$ )       | $3.50 \pm 0.02$        | $3.50 \pm 0.02$             |
| $\text{Ca}^{2+}$ ( $\text{mg}\cdot\text{L}^{-1}$ )   | $320 \pm 1$            | $320 \pm 1$                 |
| $\text{Cl}^-$ ( $\text{mg}\cdot\text{L}^{-1}$ )      | $490 \pm 1$            | $490 \pm 1$                 |
| $\text{K}^+$ ( $\text{mg}\cdot\text{L}^{-1}$ )       | $10.2 \pm 0.2$         | $1.4 \pm 0.2$               |
| $\text{Na}^+$ ( $\text{mg}\cdot\text{L}^{-1}$ )      | $308.9 \pm 0.2$        | $220.0 \pm 0.2$             |
| $\text{SO}_4^{2-}$ ( $\text{mg}\cdot\text{L}^{-1}$ ) | $1169 \pm 1$           | $1169 \pm 1$                |
| $\text{Mg}^{2+}$ ( $\text{mg}\cdot\text{L}^{-1}$ )   | $192 \pm 1$            | $192 \pm 1$                 |
| $\text{HCO}_3^-$ ( $\text{mg}\cdot\text{L}^{-1}$ )   | $183 \pm 1$            | $183 \pm 1$                 |
| $\text{PO}_4^{3-}$ ( $\text{mg}\cdot\text{L}^{-1}$ ) | $3.4 \pm 0.1$          | $3.4 \pm 0.1$               |

Calcium and magnesium concentration were determined through complexometric analysis using a 0.1 M ethylenediaminetetraacetic (EDTA) solution. Bicarbonate and chloride ions were analyzed by titration, using 0.01 M hydrochloric acid (HCl) and 0.1 M silver nitrate ( $\text{AgNO}_3$ ) solutions, respectively. Sulfate ions were analyzed by a gravimetric method using barium chloride ( $\text{BaCl}_2$  10%). Aluminum and phosphate concentrations were assessed using a HACH colorimeter (DR/890) with the reagent kits HACH 22420-00 and HACH 27425-45 (Hach Company, Loveland, CO, USA), respectively. Potassium and sodium concentrations were analyzed using flame spectrometry (Elico CL 378, Elico, Hong Kong, China). The differences in  $\text{K}^+$  and  $\text{Na}^+$  concentrations between real and synthetic Metlaoui tap water were neglected in this work. In fact, our previous work [7] revealed that these ions do not undergo adsorption on the aluminum hydroxide flocs ( $\text{Al}(\text{OH})_3$ ) formed during

the electrocoagulation process. Furthermore, due to their similar charge to the dissolved metals, they do not compete fluoride removal via coprecipitation. Additionally, these ions do not significantly influence water conductivity, which remained nearly the same at  $3.1 \text{ mS}\cdot\text{cm}^{-2}$  at  $20^\circ\text{C}$ , and are not involved in cathodic deposit compounds [7,12]. Hence, even with varying concentrations,  $\text{K}^+$  and  $\text{Na}^+$  ions generally do not exert a significant influence on the electrocoagulation treatment process.

Two different electrode configurations in the reactor were tested in this work to examine the effect of electrode positioning on fluoride removal efficiency by continuous EC. The electrodes were fixed either parallel or perpendicular to the direction of water flow. For the perpendicular electrode positioning, 6 electrodes (forming 5 electrochemical cells) were used, alternating as baffles on the left and right sides of the reactor. Figure 2 illustrates the two positioning configurations used.



**Figure 2.** Schematic representation of (a) parallel and (b) perpendicular electrode positioning (The arrows indicate the direction of the flow of fluorinated water in the reactor).

In this part of the experiment, a moderate water flow rate of  $20 \text{ L}\cdot\text{h}^{-1}$  was maintained, while the current density was adjusted (ranging from  $4.7$  to  $9.5 \text{ mA}\cdot\text{cm}^{-2}$ ) in both configurations. This variation aimed to assess the influence of this parameter on aluminum dissolution, fluoride removal yield, and energy consumption to identify the most effective configuration.

Following this, the electrodes were positioned perpendicular to the flow direction (Figure 2b) to investigate the impact of electrode connection mode (bipolar (BP), parallel monopolar (P-MP), and serial monopolar (S-MP)) on the efficiency of fluoride removal by electrocoagulation. Three alternative scenarios were examined to compare the three electric connection modes: the first one involved identical current ( $I = 0.5 \text{ A}$ ) and the same number of electrodes ( $N_{\text{el}} = 6$  electrodes), the second one involved identical current density ( $i = 4.7 \text{ mA}\cdot\text{cm}^{-2}$ ) and the same number of electrodes ( $N_{\text{el}} = 6$  electrodes), and the third one involved identical current density and the same number of electrochemical cells ( $N_{\text{cell}} = 3$  and  $5$ ) for the three connection modes. The operating conditions for comparing the connection modes are summarized in Table 2.

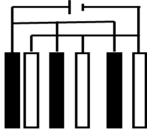
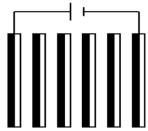
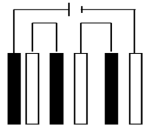
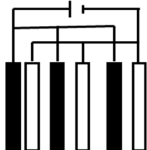
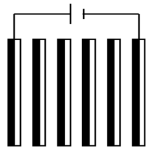
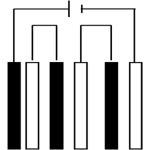
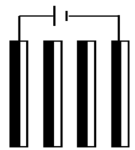
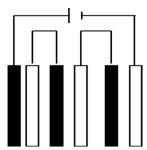
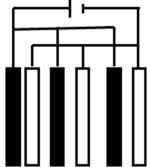
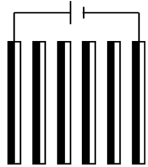
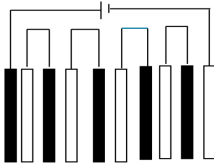
In all experiments, samples were collected at various intervals (every  $2.5 \text{ min}$  for the first  $20 \text{ min}$  and every  $5 \text{ min}$  for the subsequent  $40 \text{ min}$ ), then filtered through a  $0.45 \mu\text{m}$  polyvinylidene difluoride syringe filter (PVDF) to analyze the concentration of residual fluoride in water. This was accomplished using a specific electrode (Metrohm 6.0502.750F) with a ( $\text{Ag}/\text{AgCl}$ ) reference electrode. The detection limit of the device was  $0.016 \text{ mg}\cdot\text{L}^{-1}$ .

The fluoride removal yield ( $Y\%$ ) was calculated by the following expression (Equation (1)):

$$Y(\%) = 100 \frac{[\text{F}^-]_i - [\text{F}^-]_t}{[\text{F}^-]_i} \quad (1)$$

where  $[\text{F}^-]_i$  and  $[\text{F}^-]_t$  are fluoride concentration ( $\text{mg}\cdot\text{L}^{-1}$ ) at initial time and during the EC treatment, respectively.

**Table 2.** Operating conditions used for comparing electrical connection modes.

| Scenario 1: Same current and number of electrodes.<br>$N_{el} = 6$ electrodes, $I = 0.5$ A, $Q = 20$ L·h <sup>−1</sup>                            |                                                                                     |                                                                                       |                                                                                       |
|---------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------|
| Connection mode<br>$i$ (mA·cm <sup>−2</sup> )<br>$N_{cell}$                                                                                       | P-MP<br>0.95<br>5                                                                   | BP<br>4.7<br>5                                                                        | S-MP<br>4.7<br>3                                                                      |
| Electrical assembly                                                                                                                               |    |    |    |
| Scenario 2: Same current density and number of electrodes.<br>$N_{el} = 6$ electrodes, $i = 4.7$ mA·cm <sup>−2</sup> , $Q = 20$ L·h <sup>−1</sup> |                                                                                     |                                                                                       |                                                                                       |
| $I$ (A)<br>$N_{cell}$                                                                                                                             | 2.5<br>5                                                                            | 0.5<br>5                                                                              | 0.5<br>3                                                                              |
| Electrical assembly                                                                                                                               |    |    |    |
| Scenario 3: Same current density and same number of electrochemical cells.<br>$i = 4.7$ mA·cm <sup>−2</sup> , $Q = 20$ L·h <sup>−1</sup>          |                                                                                     |                                                                                       |                                                                                       |
| $N_{el}$<br>$I$ (A)                                                                                                                               | 4<br>1.5                                                                            | 4<br>0.5                                                                              | 6<br>0.5                                                                              |
| $N_{cell} = 3$<br>Electrical assembly                                                                                                             |  |  |  |
| $N_{el}$<br>$I$ (A)                                                                                                                               | 6<br>2.5                                                                            | 6<br>0.5                                                                              | 10<br>0.5                                                                             |
| $N_{cell} = 5$<br>Electrical assembly                                                                                                             |  |  |  |

The hydrodynamics study of the treatment was conducted using a pulsed injection of 10 mL of 0.1 M KCl solution at the reactor inlet in two different configurations while maintaining a water flow of 20 L·h<sup>−1</sup> of deionized water. Water conductivity was measured at the outlet of the reactor using a conductivity meter (Mettler Toledo SevenExcellence S470, Mettler Toledo, Hong Kong, China) to determine the concentration of KCl.

To mitigate the issue of white deposits observed on the cathodic surfaces during experiments, necessitating halts for cleaning and reactor maintenance, we investigated the effectiveness of electrode polarity reversal. Six electrodes were positioned perpendicular to the water flow direction and connected in the parallel monopolar mode for this purpose. This mode ensured the connection of all electrochemical cells to the switch box, enabling simultaneous polarity reversal in all electrochemical cells. In this study, we employed a switch box (CWT-250ATX12, Channel Well Technology (CWT), Taoyuan, Taiwan) controlled by a computer to manipulate the half-cycle duration (HCD) of polarity reversal. Inversions were performed every 0.5, 1, 2, and 5 min. Polluted water was discharged at 20 L·h<sup>−1</sup> and treated with a continuous current density of 4.7 mA·cm<sup>−2</sup>.

The measurements of the experimental quantity of dissolved aluminum were carried out by measuring the mass of the electrodes before and after the EC treatment. For each experiment, the electrodes were cleaned with paper wipes, rinsed with deionized water, and dried for 5 min in an oven at 105 °C. Afterwards, they were weighed using a RADWAG precision balance (AS220/C/2) (RADWAG Balances & Scales, Radom, Poland).

The dissolved aluminum flow rate ( $\dot{m}_{(Al)exp}$  ( $g \cdot h^{-1}$ )) was estimated from the difference between the initial and final total mass of all plates ( $m_{(electrodes)_0}$ ) and ( $m_{(electrodes)_f}$ ) during a specific period of EC treatment ( $\Delta t$ ), as expressed by Equation (2).

$$\dot{m}_{(Al)exp} (g \cdot h^{-1}) = \frac{m_{(electrodes)_0} - m_{(electrodes)_f}}{\Delta t} \quad (2)$$

where  $m_{(electrodes)_0}$  and  $m_{(electrodes)_f}$  are the total masses of the electrodes before and after treatment, respectively.

The theoretical rate of dissolved aluminum,  $\dot{m}_{(Al)th}$ , by electrocoagulation was calculated by Faraday's law, as expressed by Equation (3).

$$\dot{m}_{(Al)th} (g \cdot h^{-1}) = \frac{I_{cell} \cdot N_{cell} \cdot M_{Al}}{n \cdot F} \times 3600 \quad (3)$$

where  $I_{cell}$ ,  $N_{cell}$ ,  $M_{Al}$ ,  $n$ , and  $F$  are the current in an electrochemical cell, the number of electrochemical cells, aluminum molar mass, the number of exchanged electrons, and Faraday's constant ( $F = 96,485 \text{ C} \cdot \text{mol}^{-1}$ ), respectively.

The Faradaic yield represents the ratio of the experimentally dissolved quantity of aluminum ( $\dot{m}_{(Al)exp}$ ) to the theoretical quantity calculated by Faraday's law ( $\dot{m}_{(Al)th}$ ), as expressed by Equation (4). It is calculated to measure the efficiency of the EC process.

$$\eta = \frac{\dot{m}_{(Al)exp}}{\dot{m}_{(Al)th}} \quad (4)$$

The electrical energy consumption ( $P$ ) of EC treatment is calculated by Equation (5).

$$P (kWh \cdot m^{-3}) = \frac{U \cdot I}{Q} \quad (5)$$

where  $I$ ,  $U$ , and  $Q$  are current, overall cell potential, and water flow rate, respectively.

The total cost (TC) was calculated in this study as the sum of the electricity cost ( $C_{energy}$ ) and the cost of dissolved aluminum during the treatment of 1 m<sup>3</sup> of water ( $C_{electrodes}$ ), as expressed in Equation (6).

$$TC = C_{energy} + C_{electrodes} \quad (6)$$

The average price of one kilowatt of electricity in Tunisia is EUR 0.09, and the price of one kilogram of aluminum is EUR 2.3 [32,33].

### 3. Results and Discussions

#### 3.1. Effect of Electrode Configuration

Electrode configuration has a significant impact on electrocoagulation efficiency, as well as energy consumption [30,34,35]. In this section, we study the impact of this parameter, specifically the choice between parallel and perpendicular electrode arrangements, on fluoride removal through electrocoagulation treatment.

##### 3.1.1. Effect of Electrode Positioning on Aluminum Dissolution

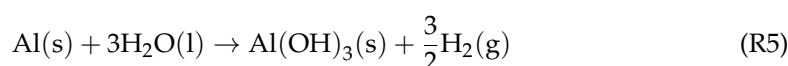
The experimental and theoretically dissolved aluminum quantities ( $\dot{m}_{(Al)exp}$  and  $\dot{m}_{(Al)th}$ ) after one hour of treatment, using three current density values (4.7, 6.6, and 9.5 mA·cm<sup>-2</sup>), are presented in Table 3. Faradaic yields are also provided for both positions.

**Table 3.** Quantities of total dissolved aluminum after one hour of treatment and Faradaic yield ( $\eta$ ) at different current densities in the two configurations ( $T = 20\text{ }^{\circ}\text{C}$ ; 6 electrodes connected in parallel bipolar (BP) electrical mode;  $Q = 20\text{ L}\cdot\text{h}^{-1}$ ).

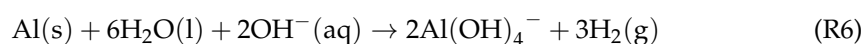
| $i\text{ (mA}\cdot\text{cm}^{-2})$                                    |                            | 4.7   | 6.6   | 9.5   |
|-----------------------------------------------------------------------|----------------------------|-------|-------|-------|
| $\dot{m}_{(\text{Al})\text{exp}}$<br>( $\text{g}\cdot\text{h}^{-1}$ ) | Parallel positioning       | 0.595 | 0.758 | 1.060 |
|                                                                       | Perpendicular positioning  | 1.026 | 1.338 | 1.790 |
| $\dot{m}_{(\text{Al})\text{th}}$ ( $\text{g}\cdot\text{h}^{-1}$ )     | Independent of positioning | 0.839 | 1.175 | 1.679 |
| $\eta$                                                                | Parallel positioning       | 0.71  | 0.64  | 0.63  |
|                                                                       | Perpendicular positioning  | 1.22  | 1.13  | 1.06  |

These results indicate a significant increase in dissolved aluminum rates when we change the electrode configuration from parallel to perpendicular positioning. Parallel positioning allows for dissolving approximately  $(58 \pm 2)\%$  of the amount of dissolved aluminum in perpendicular positioning. Moreover, in perpendicular positioning, dissolved aluminum concentrations exceed the theoretical values calculated by Faraday's law (Equation (3)). The excess metal dissolution is likely due to the following causes [36,37]:

- Pitting corrosion of the electrodes by chloride ions in the solution ( $[\text{Cl}^-] = 490\text{ mg}\cdot\text{L}^{-1}$ ).
- Aluminum corrosion by pitting at the anode (Reaction (5)).



- The attack of hydroxide ions (generated by water reduction) on aluminum cathodic electrode ((Reaction (6))



In parallel positioning, dissolved aluminum rates were lower than the expected theoretical values. This decrease can be attributed to differences in agitation and hydrodynamics within the reactor. In fact, with the parallel configuration, the inlet water flow is divided among the compartments of the reactor. Assuming uniform water distribution between the electrodes, the Reynolds number of the flow between two electrodes is approximately 11. This indicates a laminar flow regime and poor agitation between the electrodes. In laminar flow, the velocity profile of particles is parabolic and approaches zero near the electrode. Stagnation at the surface of the electrodes may cause an accumulation of dissolved aluminum in this area, slowing down the dissolution of aluminum during the process [38,39]. Moreover, at low agitation, the flocs could not mix homogeneously and settled between the electrodes. This increases the cell resistance at the electrode surface and hinders metallic dissolution. In the perpendicular configuration, water flow traverses through the same section with a higher velocity compared to the parallel electrode positioning. Additionally, the altered flow direction intensifies agitation within the reactor due to the presence of obstacles such as reactor sides and electrodes. This agitation facilitates the transfer of dissolved material from the electrode to the reactor, thereby promoting metal dissolution [40,41].

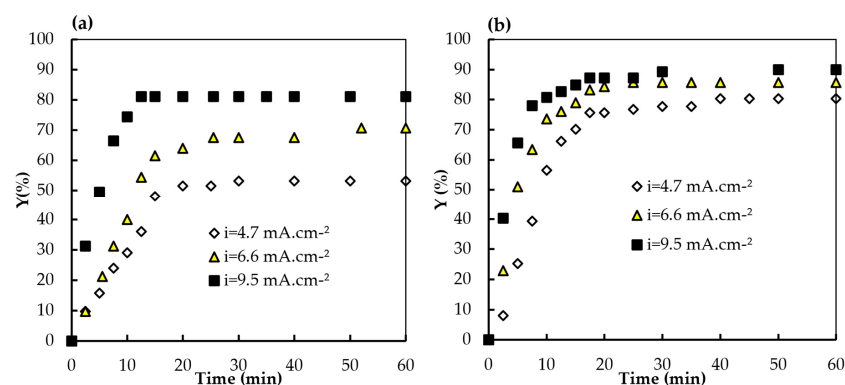
Moreover, we observed that the Faradaic yields decreased with increasing current density. In fact, the use of high current densities can lead to other secondary reactions near the anode, such as the direct oxidation of a pollutant or the formation of oxygen near the anode according to Reaction (7):



This reaction competes with aluminum dissolution reactions, consequently decreasing dissolution at higher current densities [42–44].

### 3.1.2. Effect of Electrode Positioning on Fluoride Removal Efficiency and Electrical Energy Consumption

Figure 3a,b will present the evolution of continuous electrocoagulation efficiency in fluoride removal over time using configurations both (a) parallel and (b) perpendicular to the flow direction ( $Q = 20 \text{ L}\cdot\text{h}^{-1}$ ). In this part of the experiment, a bipolar electrical connection mode was employed due to its simple electrical assembly and its low energy consumption (Cf Section 3.2.3).



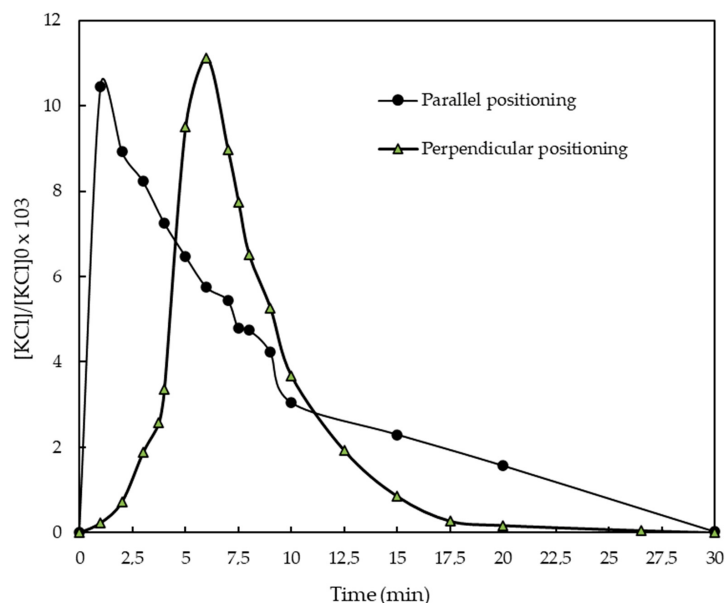
**Figure 3.** Evolution of continuous electrocoagulation fluoride removal efficiency in (a) parallel and (b) perpendicular positions ( $Q = 20 \text{ L}\cdot\text{h}^{-1}$ ;  $d = 1.5 \text{ cm}$ ; BP electrical connection mode).

The results show a significant difference in electrocoagulation efficiency between the two configurations. Indeed, the efficiencies obtained by electrodes in parallel positioning are lower than those obtained by electrodes in perpendicular positioning. Thus, the same residual fluoride concentration ( $[\text{F}^-] = (0.70 \pm 0.03) \text{ mg}\cdot\text{L}^{-1}$ ) was obtained using a current density of  $9.5 \text{ mA}\cdot\text{cm}^{-2}$  in the parallel configuration and a current density of  $4.7 \text{ mA}\cdot\text{cm}^{-2}$  in the perpendicular configuration. This was explained by measuring the quantities of dissolved aluminum using the two configurations presented in Table 3. It is also important to note that a fraction of the water passes through the extremities of the reactor without undergoing the fluoride removal reaction, whether by coprecipitation or adsorption, when the electrodes are arranged in parallel to the flow direction. This untreated portion represents approximately 33% of the total volume of untreated water. Furthermore, the observed differences could also be attributed to differences in hydrodynamic conditions.

To better understand these differences, an in-depth analysis of the reactor's hydrodynamics was conducted. Figure 4 presents the evolution of the ratio between the concentration of KCl at the reactor outlet and the initial concentration of the injected solution ( $[\text{KCl}]/[\text{KCl}]_0$ ) over time, allowing for a more precise evaluation of the impact of hydrodynamic conditions on the performance of the continuous electrocoagulation process.

The findings suggest that placing the electrodes perpendicular to the flow promoted uniformity in flow distribution. Indeed, the curves' peaks, indicating the moment when the majority of  $\text{K}^+$  and  $\text{Cl}^-$  ions exit simultaneously, are observed at 1 and 6 min for parallel and perpendicular positioning, respectively. Furthermore, the theoretical residence time ( $\tau = V/Q = 7.5 \text{ min}$ ) is closer to the actual residence time in perpendicular positioning (6 min). This configuration prolongs the tracer's presence within the reactor, thereby increasing the fluorinated water's residence time and improving fluoride treatment efficiency. Conversely, positioning the electrodes parallel to the flow leads to water short-circuiting, where a part of the water (estimated at about 33%) exits the reactor without treatment or with minimal treatment. Thus, in parallel positioning, we observe that a portion of  $\text{K}^+$  and  $\text{Cl}^-$  ions exit slowly from the reactor, indicating the presence of stagnant zones in the reactor with a decrease in water homogeneity and fluoride removal yield. These findings can be attributed to the off-centered water inlet (Figure 2a), which allows water to exit without undergoing complete treatment in the area in front of the reactor inlet.

Conversely, due to the lack of agitation in the other part, stagnant zones and dead volumes are exhibited.



**Figure 4.**  $[KCl]/[KCl]_0 \times 10^3$  vs. time in the continuous EC reactor in both electrode configurations ( $[KCl]_0$  and  $[KCl]$  are the KCl concentrations in the injected solution and at the outlet of the reactor, respectively).

The study of the energetic aspect is of paramount importance to determine the optimal electrode positioning. Table 4 presents the evolution of the total voltage in the electrochemical reactor and the energy consumption at different current densities in both configurations.

**Table 4.** The overall voltage ( $U$ ) and electrical energy consumption ( $P$ ) of EC processes in both configurations ( $Q = 20 \text{ L} \cdot \text{h}^{-1}$ ;  $d = 1.5 \text{ cm}$ ;  $T = 20 \text{ }^\circ\text{C}$ ; BP electrical mode).

|                           |                                                           | $i \text{ (mA} \cdot \text{cm}^{-2}\text{)}$ | 4.7   | 6.6   | 9.5   |
|---------------------------|-----------------------------------------------------------|----------------------------------------------|-------|-------|-------|
| Parallel positioning      | $U_{\text{par}} \text{ (V)}$                              |                                              | 12    | 15.2  | 19.4  |
|                           | $P_{\text{par}} \text{ (kWh} \cdot \text{m}^{-3}\text{)}$ |                                              | 0.299 | 0.532 | 0.970 |
| Perpendicular positioning | $U_{\text{per}} \text{ (V)}$                              |                                              | 16.2  | 21.1  | 27.1  |
|                           | $P_{\text{per}} \text{ (kWh} \cdot \text{m}^{-3}\text{)}$ |                                              | 0.405 | 0.739 | 1.354 |

The results reveal a 39% increase in the voltage of the five electrochemical cells when employing perpendicular positioning compared to parallel positioning. In parallel positioning, both the water flow and the voltage of each electrochemical cell ( $U_{\text{celpar}}$ ) remain constant across all cells. The total voltage measured during operation ( $U_{\text{par}}$ ) represents the sum of the voltages between each electrochemical cell ( $U_{\text{celpar}}$ ). In the perpendicular configuration, it is reasonable to expect that the voltages of the electrochemical cells increase as they progress towards the last cell. Indeed, the number of flocs gradually increases from the first cell to the last electrochemical cell. These aluminum hydroxide flocs are characterized by a porous structure [7], giving them significant ohmic drop. Consequently, this elevates the resistance of the electrolyte (water) and subsequently increases the voltages of the electrochemical cells, thereby raising the total voltage ( $U_{\text{perp}}$ ) [40,41].

The identical 39% increase observed between the different voltages also applies to the various energy consumptions for all current densities in both configurations. This consistency is attributed to the proportionality between energy consumption and cell voltage, as described by Equation (5).

### 3.2. Effect of Electrical Connection Mode

The previous section demonstrated the effectiveness of positioning electrodes perpendicular to the direction of polluted water flow. Consequently, we chose to use this configuration to study the effect of electrode connection mode on fluoride removal by EC. This factor has not been extensively explored in the literature, as existing studies typically involve batch electrocoagulation reactors using the same current density and electrode number for all three connection modes [35,45–48]. In this study, we varied the current density, the number of electrochemical cells, the number of electrodes, and their connection modes to determine the optimal connection mode for EC. A constant flow rate of 20 L·h<sup>−1</sup> was used for all experiments.

#### 3.2.1. Effect of Electrode Connection Mode on Total Dissolved Aluminum

Following the three scenarios outlined in Table 2, electrodes were connected using three different electrical connection modes, each operating at varying current intensities, current densities, and numbers of electrochemical cells. Table 5 presents the quantities of total dissolved aluminum after one hour of EC treatment using the three connection modes. These measurements provide crucial insights into how each connection mode influences aluminum dissolution during EC, which is essential for evaluating the efficiency and overall performance of each connection mode for fluoride removal by electrocoagulation.

**Table 5.** Quantities of experimental and theoretical dissolved aluminum and Faradaic yield ( $\eta$ ) after one hour of EC treatment at different electrical connection modes (T = 20 °C, Q = 20 L·h<sup>−1</sup>).

| Connection Mode                                                          |                  |                                          | P-MP   | BP    | S-MP  |
|--------------------------------------------------------------------------|------------------|------------------------------------------|--------|-------|-------|
| Scenario 1: $N_{el} = 6$ ,<br>$I = 0.5$ A                                |                  | $\dot{m}_{(Al)exp}$ (g·h <sup>−1</sup> ) | 0.252  | 1.026 | 0.714 |
|                                                                          |                  | $\dot{m}_{(Al)th}$ (g·h <sup>−1</sup> )  | 0.168  | 0.839 | 0.504 |
|                                                                          |                  | $\eta$                                   | 1.50   | 1.22  | 1.42  |
| Scenario 2: $N_{el} = 6$ ,<br>$i = 4.7$ mA·cm <sup>−2</sup>              |                  | $\dot{m}_{(Al)exp}$ (g·h <sup>−1</sup> ) | 1.045  | 1.026 | 0.714 |
|                                                                          |                  | $\dot{m}_{(Al)th}$ (g·h <sup>−1</sup> )  | 0.8392 | 0.839 | 0.504 |
|                                                                          |                  | $\eta$                                   | 1.24   | 1.22  | 1.42  |
| Scenario 3: $i = 4.7$ mA·cm <sup>−2</sup> ,<br>$N_{cell} = 3$ or 5 cells | $N_{cell} = 3$ : | $\dot{m}_{(Al)exp}$ (g·h <sup>−1</sup> ) | 0.723  | 0.713 | 0.714 |
|                                                                          |                  | $\dot{m}_{(Al)th}$ (g·h <sup>−1</sup> )  | 0.504  | 0.504 | 0.504 |
|                                                                          |                  | $\eta$                                   | 1.44   | 1.42  | 1.42  |
|                                                                          | $N_{cell} = 5$ : | $\dot{m}_{(Al)exp}$ (g·h <sup>−1</sup> ) | 1.045  | 1.026 | 1.095 |
|                                                                          |                  | $\dot{m}_{(Al)th}$ (g·h <sup>−1</sup> )  | 0.839  | 0.839 | 0.839 |
|                                                                          |                  | $\eta$                                   | 1.24   | 1.22  | 1.30  |

The results indicate a significant influence of electrical connection mode on aluminum dissolution in the EC process, particularly in the first and the second scenarios. In fact, in the first scenario, aluminum dissolution is the most significant when the electrodes are connected in the bipolar mode, with a value of 1.026 g·h<sup>−1</sup>. Subsequently, the serial monopolar mode shows a dissolution of 0.714 g·h<sup>−1</sup>, while the parallel monopolar mode shows the lowest dissolution, with only 0.252 g·h<sup>−1</sup> of aluminum. This variation is explained by the differences in the number of electrochemical cells and current densities, as presented in Table 2. The bipolar mode exhibited higher dissolution due to configuration with a large number of electrochemical cells (five cells) and a high current density ( $i = 4.7$  mA·cm<sup>−2</sup>). Conversely, the parallel monopolar mode led to the lowest aluminum dissolution due to the reduced number of electrochemical cells (three cells) and current density ( $i = 0.95$  mA·cm<sup>−2</sup>). In the second scenario, the results show that both the P-MP and BP connection modes dissolved the same amount of aluminum during EC. This is attributed to the involvement of the same number of electrochemical cells combined with the same current density. However, the serial monopolar mode showed the lowest aluminum dissolution. This decrease was attributed to its reduced number of electrochemical

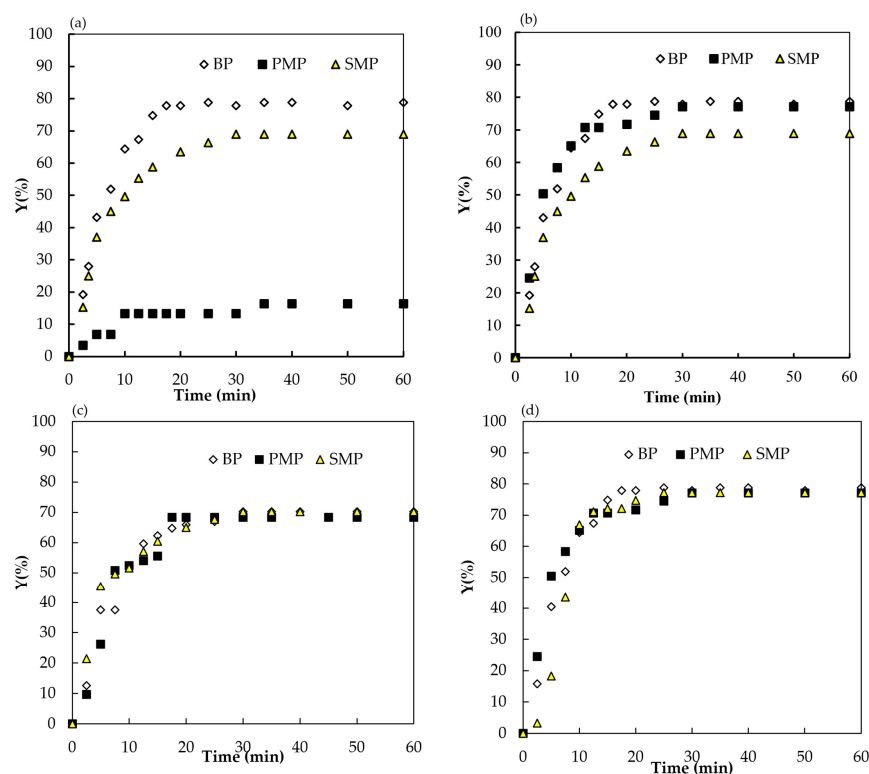
cells (three cells) compared to the other two connection modes (five cells). These findings underscore the importance of considering both the number of electrochemical cells and current density in the electrocoagulation process, as they directly influence aluminum dissolution and, consequently, treatment efficiency.

The results obtained using the third scenario demonstrate that, whether with three or five electrochemical cells, the quantities of dissolved aluminum by the three connection modes (BP, P-MP, and S-MP) during EC were very similar when the current density and the number of electrochemical cells were kept constant. Indeed, quantities of  $(0.716 \pm 0.07) \text{ g} \cdot \text{h}^{-1}$  and  $(1.014 \pm 0.06) \text{ g} \cdot \text{h}^{-1}$  were dissolved using three and five electrochemical cells, respectively, with a current density of  $4.7 \text{ mA} \cdot \text{cm}^{-2}$ . Thus, on average, each electrochemical cell dissolves approximately  $(0.22 \pm 0.02) \text{ g} \cdot \text{h}^{-1}$  of aluminum.

Table 5 demonstrates that the dissolved aluminum quantities exceed the theoretical amounts (calculated by Faraday's law). Thus, an average Faradaic yield of  $(1.3 \pm 0.2)$  was found. The excess metal dissolution is due to the electrodes pitting during electrocoagulation [36,37]. To sum up, these observations indicate that the connection modes seem to function similarly with the same number of electrochemical cells and the same current density. This finding will be explored in more detail in the following section, where the evolution of fluoride removal efficiency will be studied.

### 3.2.2. Effect of Electrode Connection Mode on Fluoride Removal Yield

In this section, we present the temporal evolution of the fluoride removal yield of synthetic water, with the same composition as Metlaoui's tap water, using EC across three scenarios (presented in Table 2). By comparing the fluoride removal efficiency trends among these scenarios, we can determine the optimal configuration for fluoride removal by EC. The progression of fluoride removal yield over time for the three scenarios is depicted in Figure 5.



**Figure 5.** Evolution of fluoride removal efficiency over time for different connection modes using (a) 6 electrodes and a constant current  $I = 0.5 \text{ A}$ ; (b) 6 electrodes and the same current density of  $i = 4.7 \text{ mA} \cdot \text{cm}^{-2}$ ; (c) a current density of  $i = 4.7 \text{ mA} \cdot \text{cm}^{-2}$  and 3 electrochemical cells; and (d) a current density of  $i = 4.7 \text{ mA} \cdot \text{cm}^{-2}$  and 5 electrochemical cells ( $Q = 20 \text{ L} \cdot \text{h}^{-1}$ ;  $d = 1.5 \text{ cm}$ ;  $T = 20^\circ \text{C}$ ).

Figure 5a demonstrates that, at the same current and number of electrodes, the bipolar mode (BP) exhibited the highest efficiency with a fluoride removal of  $(79 \pm 1)\%$ . The serial monopolar mode (S-MP) also yielded a good efficiency of  $(69 \pm 1)\%$ , meeting the WHO standard for fluoride ions ( $1.5 \text{ mg}\cdot\text{L}^{-1}$ ), although this efficiency remained lower than that achieved by the bipolar mode. This decrease is attributed to the reduction in total dissolved aluminum from  $1.026$  to  $0.714 \text{ g}\cdot\text{h}^{-1}$ , as presented Table 2. The parallel monopolar mode yielded a very low efficiency  $(17 \pm 1\%)$  compared to the other connection modes. This is explained by the decrease in the number of electrochemical cells (three cells) and the current density ( $i = 0.95 \text{ mA}\cdot\text{cm}^{-2}$ ) using this electrical mode, thus affecting the aluminum dissolution ( $\dot{m}_{(\text{Al})\text{exp}} = 0.252 \text{ g}\cdot\text{h}^{-1}$ ). These results are obtained due to variations in current density and the number of electrochemical cells, leading to differences in aluminum dissolution (Scenario 1 in Table 5), which is the key parameter in the electrocoagulation (EC) process.

On the other hand, Figure 5b demonstrates that, at the same current density and electrode number, both the bipolar (BP) and parallel monopolar (P-MP) modes exhibited the highest efficiencies, achieving fluoride removal rates of  $(79 \pm 1)\%$ . The serial monopolar mode (S-MP) also demonstrated a good efficiency of  $(69 \pm 1)\%$ , but remained lower than the efficiency provided by the other two connection modes. This reduction is attributed to the decrease in total dissolved aluminum (with  $\dot{m}_{(\text{Al})\text{exp}} = 0.714 \text{ g}\cdot\text{h}^{-1}$ ) compared to the other connection modes (with  $\dot{m}_{(\text{Al})\text{exp}} = 1.026$  and  $1.045 \text{ g}\cdot\text{h}^{-1}$  for BP and P-MP, respectively). This reduction is related to the reduced number of electrochemical cells in the S-MP mode (three cells) compared to the other modes (five cells).

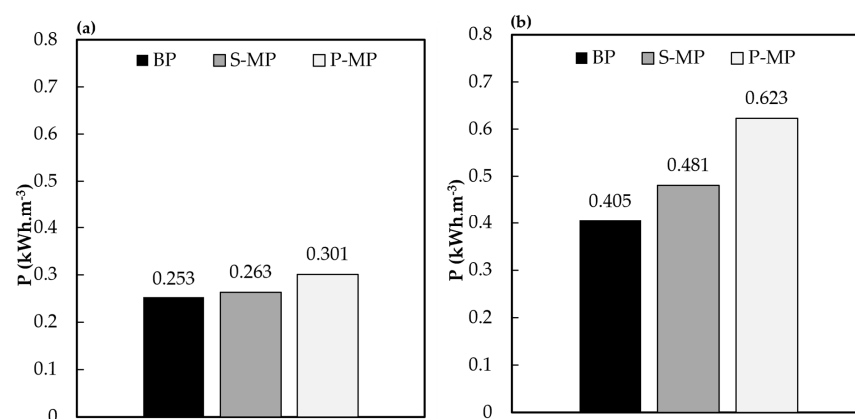
The results obtained by the third scenario, as depicted in Figure 5c,d, reveal an interesting finding: when maintaining a constant number of electrochemical cells and current density, the efficiency of fluoride removal remains constant using the three connection modes. This uniformity in efficiency is explained by the equivalent amounts of dissolved aluminum produced by the three connection modes in this scenario (Cf Table 5). Since aluminum dissolution plays a crucial role in the electrocoagulation process by co-precipitating with fluoride ions and by forming coagulant species that adsorb pollutant ions, the similar levels of aluminum dissolution across different connection modes contribute to the constant efficiency observed [7,12]. In addition, it should be noted that after varying the operational conditions, we did not observe a significant difference in the water pH, particularly during the steady state. This was due to the bicarbonate ions in the water acting as a buffer, maintaining the pH at  $7.0 \pm 0.3$  despite the changes in operating conditions. Overall, these findings highlight the importance of considering factors such as aluminum dissolution by fixing the number of electrochemical cells and current density when optimizing the electrocoagulation process for fluoride removal, regardless of the electrical connection mode.

### 3.2.3. Effect of Electrode Connection Mode on Energetic Consumption

Energy consumption is of paramount importance in selecting the optimal operating conditions for the process. Given that similar efficiencies are achieved with the same number of electrochemical cells and current density, the choice of connection mode cannot be made without considering its implications in terms of energy consumption. Figure 6 illustrates the electrical energy consumption rates for the fluoride removal of  $20 \text{ L}\cdot\text{h}^{-1}$  of polluted water across different connection modes using a current density of  $4.7 \text{ mA}\cdot\text{cm}^{-2}$  and (a) three and (b) five electrochemical cells.

Figure 6a,b illustrate that, when using the same current density with the same number of electrochemical cells, we observe that the parallel monopolar connection mode (P-MP) results in the highest energy consumption due to the high applied current intensity ( $1.5 \text{ A}$  and  $2.5 \text{ A}$  for three and five electrochemical cells, respectively). The bipolar connection mode (BP) exhibited the lowest electrical energy consumption. Specifically, only  $0.253$  and  $0.405 \text{ kWh}\cdot\text{m}^{-3}$  were consumed by EC treatment using three and five cells, respectively. This is explained by the current division among the different electrochemical cells and

the low measured voltage. Thus, using this electrical connection mode, we can achieve a fluoride concentration of  $0.7 \text{ mg}\cdot\text{L}^{-1}$  in Metlaoui's tap water with a total cost of only EUR 0.154 per  $\text{m}^3$  of water (calculated by Equation (6)).



**Figure 6.** Energy consumption of (a) three and (b) five electrochemical cells for the three electrical connection modes ( $i = 4.7 \text{ mA}\cdot\text{cm}^{-2}$ ;  $Q = 20 \text{ L}\cdot\text{h}^{-1}$ ; EC duration = 1 h).

Therefore, it can be concluded that the optimal electrical connection mode for the fluoride removal by electrocoagulation process is the bipolar mode. This conclusion is based on the process performance, considering both operational efficiency and total cost.

Finally, to ensure that no more pollutants were added by the EC process, an analysis of the residual aluminum in the water was undertaken. These levels did not surpass the limit of detection of the utilized colorimeter, set at  $0.013 \text{ mg}\cdot\text{L}^{-1}$ . As a result, the level remains within the WHO's recommended limit for residual aluminum content in drinking water ( $0.2 \text{ mg}\cdot\text{L}^{-1}$ ).

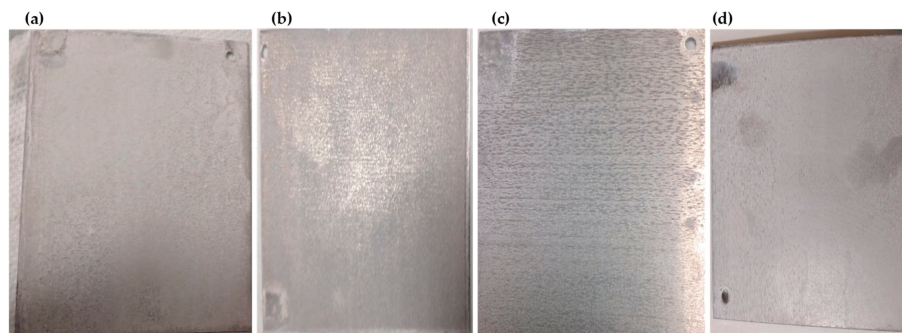
### 3.3. Efficiency of Polarity Reversal in Deposit Removal

With the presence of bicarbonate ions in water, calcium leads to the formation of a white cathodic deposit, primarily composed of  $\text{CaCO}_3$  [7,12]. This deposit prevents corrosion and poses a problem for reactor maintenance. Table 6 presents the amounts of dissolved aluminum after one hour of EC with different half-cycle durations of polarity reversal (0.5, 1, 2, and 5 min) and without polarity reversal. This represents the sum of mass losses from all electrodes after one hour of treatment.

**Table 6.** The quantity of dissolved aluminum and the number of inversions after one hour of EC with and without polarity reversal ( $i = 4.7 \text{ A}\cdot\text{cm}^{-2}$ ,  $N_{\text{el}} = 6$ ,  $Q = 20 \text{ L}\cdot\text{h}^{-1}$ ).

| Half-cycle duration (HCD) (min)                                | $\infty$ | 0.5   | 1     | 2     | 5     |
|----------------------------------------------------------------|----------|-------|-------|-------|-------|
| Number of inversions in one hour (NI)                          | 0        | 119   | 59    | 29    | 11    |
| $\dot{m}_{(\text{Al})\text{exp}} (\text{g}\cdot\text{h}^{-1})$ | 1.026    | 0.751 | 0.844 | 0.958 | 0.964 |

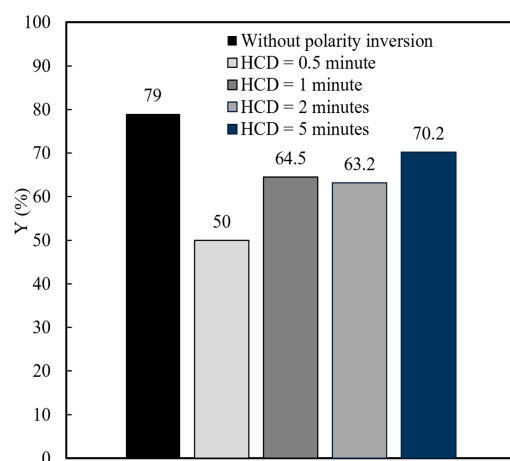
The amount of total dissolved aluminum in water with polarity reversal is consistently lower than that without reversal. This quantity increases with the half-cycle reversal duration, with a notable increase between 0.5 and 1 min of HCD, followed by a less pronounced increase at longer reversal durations. These results are also linked to the number of polarity inversions (NI). In fact, reducing the half-cycle reversal duration increases the number of inversions. Each reversal leads to a decrease in current density and cell voltage before initial values are restored [49], thereby reducing the quantity of dissolved aluminum. The effect of current reversal on cathodic deposit removal is observed and illustrated using the photos presented in Figure 7a–d.



**Figure 7.** Surface aspect of electrodes after one hour of treatment at different HCDs: (a) without polarity reversal; (b) 0.5; (c) 1; and (d) 2 min HCD ( $i = 4.7 \text{ mA}\cdot\text{cm}^{-2}$ ,  $N_{\text{el}} = 6$ ,  $Q = 20 \text{ L}\cdot\text{h}^{-1}$ ).

Polarity reversal decreases the electrode passivation induced by cathodic deposits with an optimal polarity reversal cycle. In fact, we observed that the deposit was eliminated with polarity reversals every minute or less (Figure 7b,c), and persisted on the electrodes when inversions occurred every 2 min or more (Figure 7d). The electrodes were alternately reversed to dissolve the aluminum in the tap water through oxidation, thereby eliminating the deposits on the electrodes [43,50]. In fact, when an electrode switches polarity from cathode to anode, the electrochemical reaction reverses: the new anode begins to oxidize (R1), thereby dissolving the aluminum and the deposits that had accumulated on it. These deposits, primarily composed of calcium carbonate ( $\text{CaCO}_3$ ) [7], become more soluble and easier to detach in the acidic environment that is created locally around the anode during oxidation [43,49,50].

In this section, the effect of polarity reversal on fluoride removal efficiencies over time with varying HCDs is studied. Figure 8 presents fluoride removal efficiencies with and without polarity reversal.



**Figure 8.** Fluoride removal yields after one hour of electrocoagulation treatment with different half-cycle durations of polarity reversal ( $i = 4.7 \text{ mA}\cdot\text{cm}^{-2}$ ,  $N_{\text{el}} = 6$ ,  $Q = 20 \text{ L}\cdot\text{h}^{-1}$ ).

These results highlight that fluoride removal efficiency with polarity reversal remains lower than that obtained after one hour of treatment without polarity reversal. However, by increasing the cycle duration, we observe an increase in the fluoride removal rate, despite the persistence of deposits. This observation is explained by the increase in the quantity of dissolved aluminum, as indicated in Table 6. In fact, as the half-cycle duration increases from 0.5 to 5 min, aluminum dissolution improves, nearing the total dissolved aluminum without polarity reversal. This leads to an increase in the final fluoride removal yield, approaching the yield achieved by EC without polarity reversal (Figure 8). In addition, during the cathodic half cycle, an alkaline layer is formed on the electrode surface. Dissolved aluminum precipitates in this layer as aluminum hydroxides, exhibiting

distinctive passivating characteristics, and resulting in a subsequent decrease in fluoride removal yield. A longer half-cycle duration allows the formation of an acidic boundary layer at the anode that reduces metal precipitation [43,51].

Indeed, upon analyzing the data, we also observed that, despite the decrease in efficiency linked to polarity reversal, the quantity of aluminum required to remove the same amount of fluoride remains constant, whether polarity reversal is employed or not. Specifically,  $(0.40 \pm 0.03) \text{ g} \cdot \text{h}^{-1}$  represents the amount of aluminum required to eliminate  $1 \text{ g} \cdot \text{L}^{-1}$  of fluoride ions using a water flow rate of  $20 \text{ L} \cdot \text{h}^{-1}$ . Additionally, the cell voltage remains constant at  $U = (16.2 \pm 0.2) \text{ V}$ , ensuring uniform energy consumption regardless of polarity reversal. The uniform dissolution of aluminum per gram of treated fluoride and the unchanged energy consumption suggest that polarity reversal is an effective method for electrode deposit removal without disrupting treatment. This approach eliminates the need to stop treatment for electrode cleaning and avoids the use of additional chemicals or mechanical friction, which can lead to aluminum loss during washing and increase the overall treatment cost.

Polarity reversal every minute was chosen as the optimal half-cycle inversion duration to ensure efficient fluoride removal, achieving residual fluoride concentrations below the WHO standard of  $1.5 \text{ mg} \cdot \text{L}^{-1}$  while preventing the formation of cathodic deposits.

#### 4. Conclusions

In this work, the impact of electrode positioning on fluoride removal through electrocoagulation (EC) was explored. Two electrode positioning configurations were examined: one where the electrodes were aligned parallel to the water flow direction, and the other where perpendicular electrodes were staggered on both sides of the EC reactor. The findings revealed that the parallel positioning only managed to dissolve 58% of the aluminum quantity dissolved by the perpendicular positioning during EC. This discrepancy significantly affected the fluoride removal by EC process. Therefore, the perpendicular positioning was deemed optimal due to its notable effectiveness in fluoride removal from water. Adopting this positioning, the study of the effects of electrical connection modes was conducted. The subsequent analysis of various scenarios demonstrated that all three connection modes yielded similar efficiencies when current density and the number of electrochemical cells were kept constant. Among these, the bipolar mode emerged as the optimal connection mode due to its low energy consumption, ease of implementation, and the absence of residual aluminum in the treated water. Finally, polarity reversal demonstrated good fluoride removal efficiency when the half-cycle duration was greater than or equal to 1 min, and an effective elimination of cathodic deposits for half-cycle durations less than or equal to 1 min. Polarity reversal every minute was chosen to ensure both good efficiency and cathodic deposit removal without stopping the treatment, and without the need for chemical or mechanical interventions.

In summary, this work has demonstrated that optimizing operational parameters enhances fluoride removal efficiency and reduces operational costs, promoting the sustainable management of energy and water resources.

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