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Removal of Sulfate and Iron from a Water Solution Using a New Flow Pattern in an Electrocoagulation Reactor

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- *River and rejected RO water purification.*
- *Removing sulfate and iron from water.*
- *Effect of number of electrodes and flow rates.*

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Abstract: This research examines the application of electrocoagulation (EC) by employing two water sources: river water and rejected water from a reverse osmosis system. To assess the impact of numerous factors on the removal efficiency of sulfate and iron, continuous flow experiments were conducted using bipolar and monopolar aluminum electrodes. The parameters studied included the number of electrodes (2, and 4) and flow rates (600, and 1000 L/h). The experimental findings revealed that increasing the number of electrodes improved the removal efficiency. Conversely, an increase in flow rate resulted in a decrease in removal efficiency for both water sources. For concentrated water, the best sulfate removal reached 47% (for four plates with 600L/h), whereas for the river, the highest sulfate removal was 50% (for four plates and a flow rate of 1000 L/h). For river water samples, the best iron removal was 56% (for four plates and 600L/h), whereas for concentrated water samples, the most significant removal was 79% (for four plates and 600L/h).

ازالة الكبريتات والحديد من محلول مائي باستخدام نمط جريان جديد في مفاعل التخثر الكهربائي

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الخلاصة

هذا البحث يدرس استخدام عملية التخثر الكهربائي لنوعين مياه مختلفة: مياه نهر ومياه الرفض لمنظومة التناضح العكسي. لتقييم تأثير عوامل مختلفة على ازالة الكبريتات والحديد تم اجراء تجارب ذات جريان مستمر باستخدام اقطاب الالمنيوم على شكل الواح مربوطة بطريقة احادية وثنائية القطب. العوامل التي تمت دراستها هي: عدد الاقطاب (٢و٤) ومعدل جريان (٦٠٠و١٠٠٠) لتر /ساعة. اظهرت النتائج ان زيادة عدد الاقطاب ساهمت في تحسين كفاءة الازالة. بالمقابل زيادة معدل التدفق ادت الى تقليل كفاءة الازالة لكل من مصادر المياه. بالنسبة لمياه المرفوضة المركزة بلغت أعلى نسبة ازالة للكبريتات ٤٧٪ (عند استخدام اربعة ألواح مع معدل جريان ٦٠٠ لتر/ساعة) بينما بلغت اعلى نسبة ازالة للكبريتات في مياه النهر ٥٠٪ عند استخدام ٤ الواح وبمعدل جريان ١٠٠٠ لتر/ساعة. وكانت ازالة الحديد لمياه النهر بنسبة ٥٦٪ باستخدام ٤ الواح ومعدل سرعة جريان ٦٠٠ لتر /ساعة بينما كانت نسبة ازالة الحديد للمياه المرفوضة من منظومة فلاتر التناضح العكسي ٧٩٪ باستخدام ٤ ألواح وبمعدل جريان ٦٠٠ لتر/ساعة.

الكلمات الدالة: الكاربون المنشط، الإمتزاز، النفط الخام، الفناديوم.

1.INTRODUCTION

Physical and chemical treatments are used to treat water [1]. Coagulation, electro-oxidation, electroflotation, precipitation, adsorption, and settling are examples of pollutant removal mechanisms [2]. One of these methods is electrochemical coagulation, the electrochemical synthesis of destabilizing agents that results in charge neutralization for pollutant elimination [3]. During water treatment procedures, electrocoagulation is a popular approach for removing various contaminants. Recent studies have shown that electrocoagulation significantly affects drinking water quality [4]. EC is an electrolytic process where the wastewater serves as the electrolyte [17] by applying a current to electrodes immersed in a solution, EC enables removing pollutants from a solution [37]. Typically, the electrodes are constructed of either iron or aluminum [37-41, 17]. Table 1 shows some of the research on removing different species and ions. The idea behind the electrocoagulation process is that the coagulants are produced in situ as the sacrificial metallic anode dissolves under the influence of the applied current, and the cathode produces hydrogen gas that floats the contaminants [42]. Removal of coagulated pollutants by sedimentation or by electro-flotation by evolved H2. Electro-flotation can disperse the coagulated particles via the bubbles of H2 gas produced at the cathode from the water reduction reaction, transporting the solids to the top of the solution [43]. Fig. 1 shows a schematic representation of the EC process. Numerous chemical reactions occur at the electrode surfaces throughout the EC process, particularly the dissolution of aluminum by anode oxidation, which also results in the simultaneous reduction of water to generate hydrogen gas. The result of the breakdown of water is [45]:

At cathode:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad \textbf{(1)}$$

At anode:

$$Al \rightarrow Al^{3+}_{(aq)} + 3e^- \quad \textbf{(2)}$$

$$2H_2O \rightarrow 4H^+ + O_2 + 4e^- \quad \textbf{(3)}$$

The developing Al^{3+} ions are effective coagulants for flocculating particles. However, the hydrolysed aluminium ions can create large Al-O-Al-OH networks that chemically adsorb pollutants [46].

Table 1 Removal of Different Species Using Electrocoagulation.

Parameter	Reference No.
Hardness, Fluoride	[5]
Arsenic	[6]
BOD, P, FC, COD	[7]
Calcium, Turbidity	[8]
Polymer Types: polyamide (PA), Polyethylene (PE), Polyethylene Terephthalate (PET) and Polypropylene (PP)	[9]
Turbidity, COD, BOD	[10]
Hardness, SO4 , and Manganese	[11]
Total Phosphorous, COD	[12]
Chlorella Vulgaris	[13]
Dye, COD	[14]
Perfluorooctanoic Acid, Microcystins	[15]
TDS	[16]
TSS, Oil Grease	[17]
Total Pohosphorus, Total Nitrogen, TOC, Turbidity	[18]
Turbidity	[19]
COD	[20]
TDS, TSS, HCO3, CL, Ca	[21]
DFZ436, COD, DFZ437, COD, Conductivity, chloride, TDS	[22]
Fe, Turbidity, KMnO4	[23]
Turbidity	[24]
Calcium, Magnesium, Silica	[25]
TTHM, NOM, DOC	[26]
Color, Turbidity	[27]
Hardness	[28]
TDS, Cl, Br, SO4	[29]
Hardness, Alkalinity, TDS	[30]
Arsenic	[31]
Arsenic	[32]
Phosphate	[33]
Chromium (VI)	[34]
Iron	[35]
Fluoride	[36]
Sulfate, Iron	This study

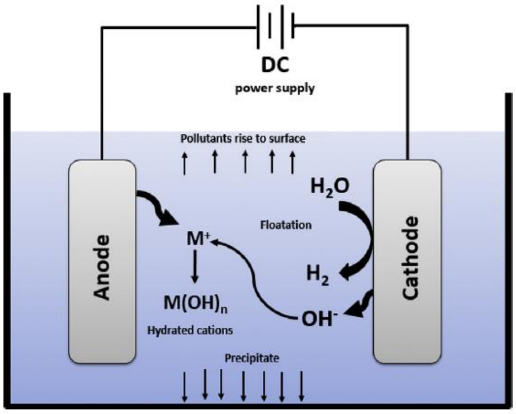


Fig. 1 A Schematic Description of the EC Cell. [44].

It chemically induces the aluminum and its hydroxide film, and it is represented by [47,48]:

$$2Al+6H_2O+2OH^- \rightarrow 2Al(OH)^-_4+3H_2 \quad \textbf{(4)}$$

$$2Al+3H2O \rightarrow 2Al+3+\frac{3}{2}H_2+3OH^- \quad \textbf{(5)}$$

Additionally, the $Al(OH)_{-4}$ ions are released during a chemical reaction, and they can interact with cationic species to minimize the pollutants from effluent. So, they neutralize their charges and decrease their solubility [45].

$$[Al(OH)_{-4}] \rightarrow Al(OH)_3+OH^- \quad \textbf{(6)}$$

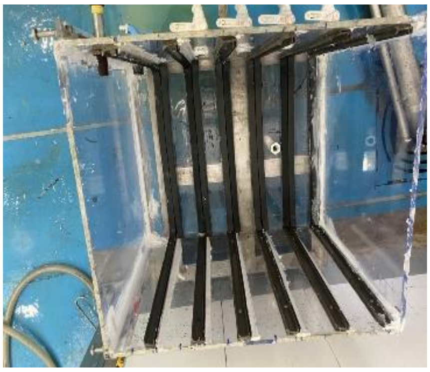
The electrocoagulation method consists of three steps: the destabilization of pollutants, the suspension of particles, and the de-emulsification, as well as the aggregation of unstable phases and floc-forming [49-51] , all of which contribute to the synthesis of coagulant [51,52]. The migration of the produced cations to the oppositely charged electrode (electrophoresis) destabilizes the negatively charged pollutant by the double layer compression or charge neutralization, thus lowering the repulsive forces and promoting the particles' aggregation (coagulation) [53,54,37]. This step includes compression of the diffuse double layer (electrical double layer) around the charged species by the interaction of ions generated by oxidation of the sacrificial anode, charge neutralization (resulting in a zero net charge) of the ionic species presents in the media by counter ions produced by the electrochemical dissolution of the sacrificial anode and floc formation as a result of particle bridging [55]. This destabilization mechanism is quite simple, where the adsorption of counter-charged ions on the surface of colloidal particles neutralizes their surface charge so that repulsive forces are overcome, and Van der Waals attractive forces dominate. Eventually, colloidal particles approach each other and coagulate [6,44,56-58]. Also, the entrapment of particles in the sediment, called sweep coagulation, is often encountered when high metal salt concentrations are added. In such cases, the metal salts react with water, forming insoluble metal hydrates that precipitate, forming a sludge blanket. The formed precipitates eventually entrap colloidal particles during and after precipitation [44, 57, 58]. As a result, coagulation may occur due to the creation of flocs, which entrap and connect colloidal particles still present in the aqueous medium [59]. The electrocoagulation process has the following advantages over other chemical processescompared to other chemical procedures: effluent has fewer total dissolved solids, is easy to operate, and degrades organic waste more quickly and effectively than chemical coagulation, and bigger and more stable flocs are developed. Except in severe circumstances, controlling the pH of the water does not need chemicals, lowers residue, processes various contaminants simple to remove, and its operating costs are far lower than those of most current technologies [60]. The quantity of sludge produced by EC would be reduced since it does not need a chemical additive and removes pollutants quickly [61]. Due to these advantageous characteristics, EC is preferable to traditional physicochemical treatment methods [41]. It reduced maintenance costs, fewer labor requirements, and quick results [62]. This article studied removing of sulfate and iron for two and four-plate electrodes using two flow rates to treat river water and reverse osmosis rejected water.

2.EXPERIMENTAL PROGRAM

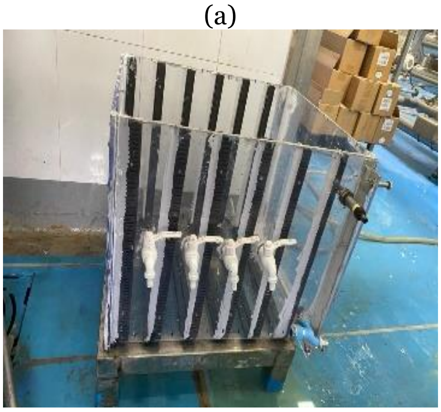
2.1.Experimental Sets

The EC experiments were conducted in continuous mode using a plastic reactor. A transparent plastic reactor is advantageous for observing the reaction process and monitoring flocks' formation and the pollutants' deposit. Additionally, using this non-conductive material ensures an appropriate setting for the reaction. The dimensions of the reactor were 50cmx50cmx60cm, with vertically placed aluminum plate electrodes for the anode and cathode. The plates consisted of holes (2.5 cm diameter, 5 cm spacing between the holes), as shown in Fig. 2. Two and four electrodes were used to determine the effects of electrode surface area. For the two plates experiment, the space between plates was 40 cm, while for the four plates experiment, the distance between the first plate and the second was 10 cm, as between the third and the fourth. The space between the third and the second plates is 20 cm. The plates were connected in monopolar and bipolar parallel connection modes. The surface area of the anode and cathode was 2500 cm^2 (0.25 m^2). Aluminum was chosen for the anode and cathode due to the low cost, reliability, and accessibility of the material, and it is better than iron for treating drinking water [60]. The experimental setup is shown in Fig. 3. The experiments were conducted in a controlled environment with accurate temperature regulation. The anode and cathode plates were connected to a DC power supply's positive and negative ports ('Model: S-480-48, DC output:48V,10A). Fig. 4 represents the pumps used for the treatment. The specifications of the pumps are shown in Table 2. Before each experiment, the electrodes were scraped using fine sandpaper, cleaned with (5%) hydrochloric acid solution for 5 min, rinsed with distilled water, dried, and finally weighed. The cleaning process prevents the material precipitation on the electrodes during long-term operation and induces a passivating effect that decreases treatment performance and increases power requirements. So, cleaning the electrodes was to remove and avoid a passivation film forming on the electrodes. Arranging of plates with holes could enhance the mixing and dispersion of contaminants in the water, thereby increasing the efficiency of the EC process. Alternating upward and downward flow paths created by the holes might lead to more effective contact between the electrodes and the contaminants, improving the coagulation and flocculation reactions.

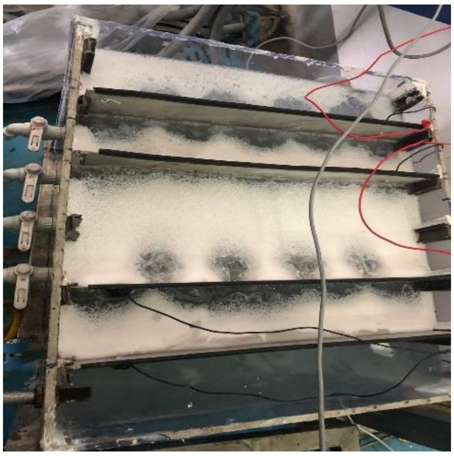
Fig. 2 Plate of Aluminum used for Cathode and Anodes.



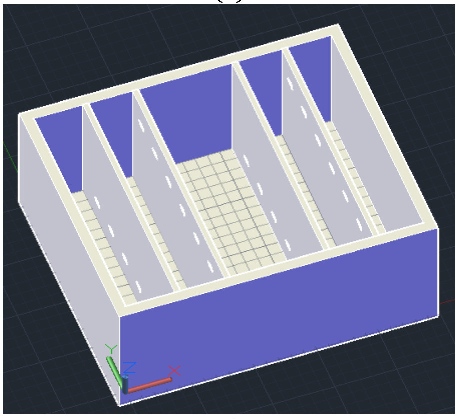
(a)



(b)



(c)



(d)

Fig. 3 The Box Used for the Treatment (a) Top View (b) Side View (c) During the Experiment(d) Diagram Shows the Distribution of the Holes in the Reactor.

(a)

(b)

Fig. 4 Pumps Used (a) 1000L/h (b) 600L/h.

Table 2 The Specification of the Submerged Pumps that were Used.

Parameter	First pump	Second pump
AC	220-240 V 50Hz, 12W	220-240V 50Hz, 18W
Qmax	600L/h	1000L/h
Hmax	1.6 m	2.0 m

Table 3 Operating Parameters Values of the Present Work.

Experiment No.	Water Properties				No. of Electrodes	Electrode Configuration	Flow rate, L/h	Water Type
	Conductivity	AL, ppm	SO_4, ppm	Iron, ppm				
e1	3309	0.12	710	0.4	2	monopolar	1000	Concentrated
e2	3475	0.09	770	0.08	4	bipolar	1000	Concentrated
e3	3406	0.09	700	0.3	2	monopolar	600	Concentrated
e4	3345	0.034	210	0.3	4	bipolar	600	Concentrated
e5	890.3	0.034	210	0.314	2	monopolar	600	River
e6	1009	0.02	280	0.08	4	bipolar	600	River
e7	1039	0.11	200	0.169	2	monopolar	1000	River
e8	929.9	0.11	240	0.027	4	bipolar	1000	River

2.2. Water Samples

The water samples used in this work were collected from the Tigris River, and the rejected water was from the reverse osmosis system (membrane), which consisted of the salts' main concentration. A continuous process was done using submerged pumps with a flow rate of (600 L/h and 1000 L/h) the treatment time was chosen to be (60-50 minutes) with an applied voltage of 36 V and 3-2 A current for the plates. Table 3 shows the condition for each experimental run.

3. RESULTS AND DISCUSSION

The removal percentage of the sulfate and iron and the increasing percentage of the aluminum in the final solution were recorded. Sulfate, iron, and aluminum were measured using a Spectrophotometer (HACH DR6000).

3.1. Effect of the Number of Plates on the EC Process

The number of electrodes used in the EC process is an essential factor affecting the process; the electrode area influences the current density and can directly impact contaminants' removal efficiency. Electrocoagulation involves using electrodes to generate coagulant species, such as metal hydroxide flocs, that aid in removing contaminants. The surface area accessible for electrochemical reactions is increased by increasing the number of electrodes, which raises the rate at which coagulant species are generated; expanding the generation of coagulants may improve the efficacy of pollution clearance. When there are more electrodes, there are more places of interaction between the coagulant and the water's contaminants; increasing interaction between the coagulant and contaminants increases the probability of coagulation and subsequent clearance. The coagulant species generated at the anode electrode may interact with and neutralize other pollutants. Increasing the number of electrodes strategically within the electrocoagulation reactor will enhance flocculation and mixing. The electrodes may improve the dispersion and distribution of coagulant species throughout the water by creating flow patterns and turbulence. Enhanced mixing facilitates producing larger flocs by bringing the pollutants into touch with the coagulant. Increasing the number of electrodes increases the possibility of contact with pollutants and guarantees that the coagulant is dispersed uniformly. It improves the overall removal efficiency by lowering the chance of dead zones where the coagulant may not reach. Figs. (5-8) compare the results of studies conducted with two and four plates for aluminum, sulfate, and iron. These figures illustrate that as the number of electrodes increases, the removal percentage increases for Fe and SO_4. These findings are the same results as removing cadmium by Khaled et al. [63], COD removal by Elnenay et al. [64], TDS and turbidity by Gusa et al. [65], and non-sugar removal by Noersatyo et al. [66]. Thus, as the number of electrodes increased, the large surface area of the electrodes (the cross-sectional area for the current supply) led to excellent current efficiency [67]. The results are shown in Fig. 5 for concentrated water and a flow rate of 1000 L/h. The number of plates needed to achieve the best removal, and the quantity of aluminum released into the solution was raised. Fe and SO_4 were removed at 9.4% and 77%, respectively, and Al increased by 66%. Fig. 6 displays the results with concentrated water and a flow rate of 600 L/h. The SO_4 and Fe removal rates were 47% and 79%, respectively, while the increasing percentage for Al was 79%. Fig. 7 shows the result for the river water and a flow rate of 600 L/h; The removal rate for SO_4 and Fe was 32% and 56%, respectively, while for Al, the increasing percentage was 90%. Fig. 8 shows the removal percentages for (Fe and SO_4) and the amount of released AL as the number of plates grows for river water flowing at a flow rate of 1000L/h. Fe and SO_4 removal rates were 29% and 48%, respectively, while the rate at which Al increased was 59%.

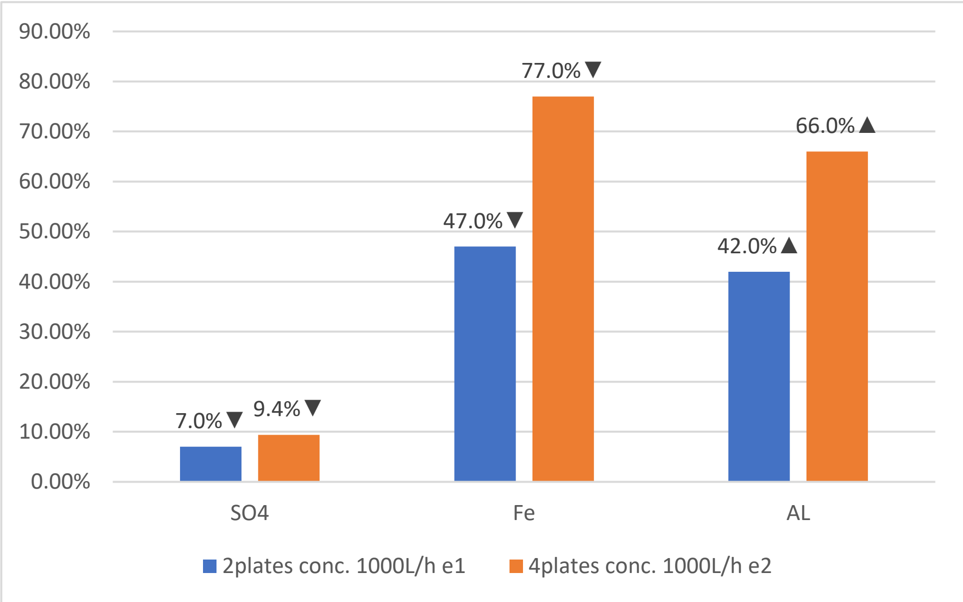


Fig. 5 The Effect of the Number of Plates Used for Concentrated Water for Flow Rates of 1000 L/h.

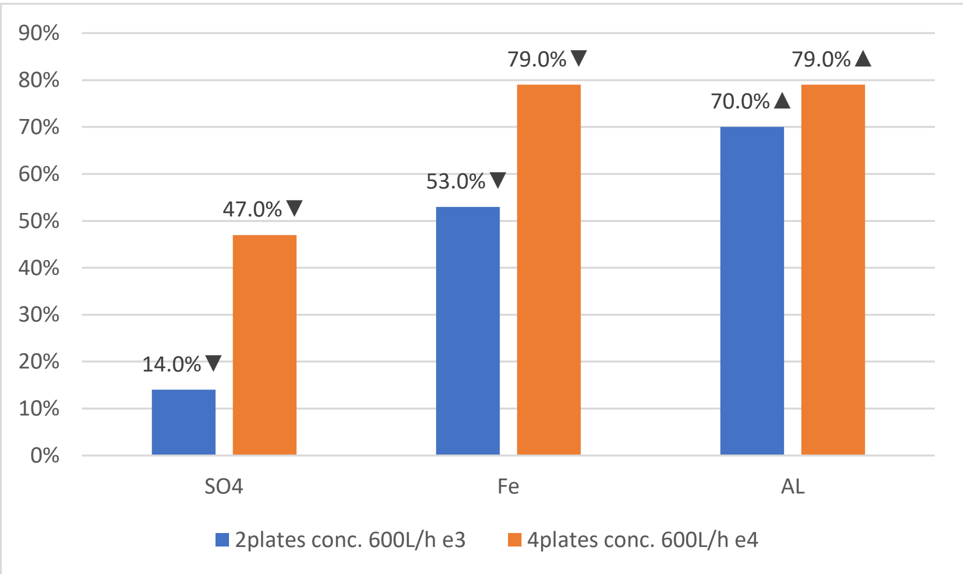


Fig.6 The Effect of the Number of Plates Used for Concentrated Water for Flow Rates of 600 L/h.

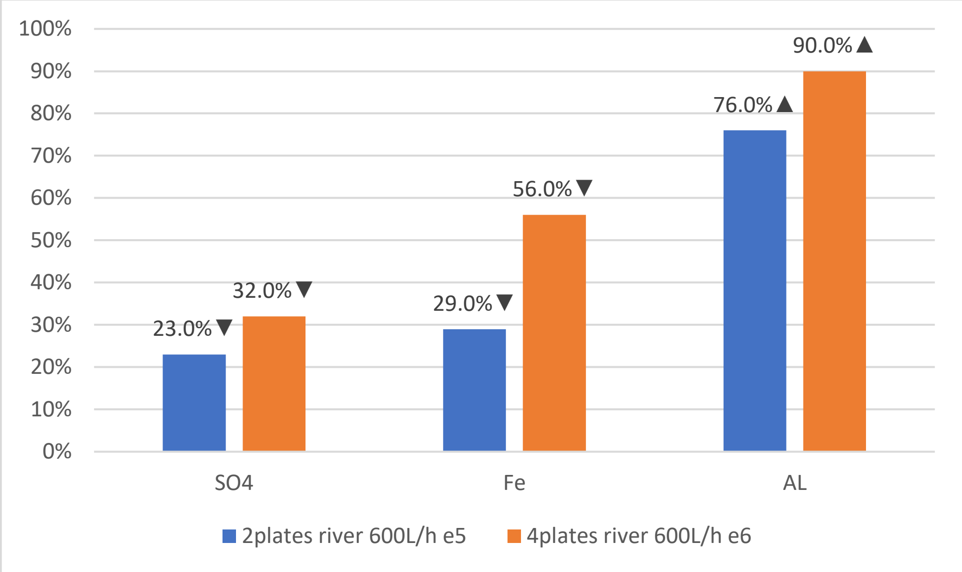


Fig. 7 The Effect of the Number of Plates Used for River Water for Flow Rates of 600 L/h.

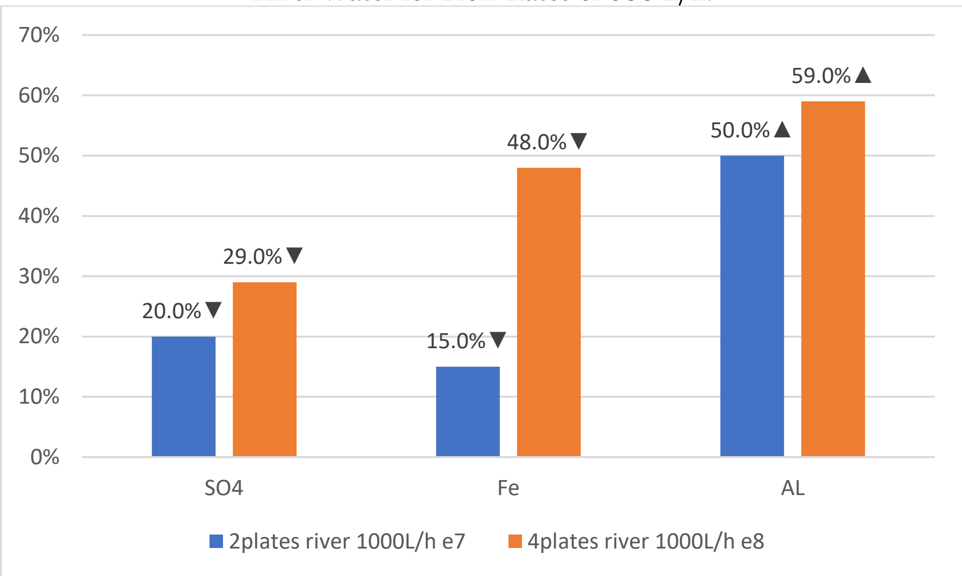


Fig. 8 The Effect of the Number of Plates used for River Water for Flow Rates of 1000 L/h.

3.2.Effect of Flowrates on the EC Process

The electrocoagulation process can be affected by flow rates in several ways. The electrocoagulation process's effectiveness and productivity are mainly dependent on flow rates. It includes forming of flocks and reactions inside the reactor, eliminating iron and sulfate. and releasing aluminum ions. The residence time would be decreased as the flow rate increases; this might lead to insufficient removal of contaminants and incomplete coagulation. On the other hand, lowering the flow rate will increase the residence time, allow coagulation, and increase the contaminant's removal efficiency. The mixing process of the coagulant produced and the pollutant depends on the flow rate. A higher flow rate may enhance the mixing and the interaction between the coagulant and the contaminant. Still, a higher flow rate could reduce the coagulation by producing turbulence. An optimal flow rate must be determined to get sufficient mixing and dispersion while avoiding excessive turbulence. The flow rates affect the electrical current and the coagulant's generation between the electrodes. Also, the elevated flow rates may result in higher mass transfer of the coagulant and increase the electrode efficiency. Higher flow rates could lead to insufficient current distribution and incompatible coagulant production, reducing the efficiency of the process. So it is essential to integrate the two main factors (residence time and mixing). After all, a reduction in period leads to insufficient EC process at higher flow rates. On the other hand, mixing is better than electrocoagulation since it makes contaminants easier to remove. Nevertheless, it is crucial to maintain equilibrium as excessive mixing might result in a decline in overall efficiency. In conclusion, the significance of mixing in the electrocoagulation (EC) process must be considered. However, achieving an optimum residence duration is crucial to its overall efficacy in removing contaminants. Figs. (8-10) reveal that as the flow rate increases, the removal per cent decreases, and the amount of Al released decreases. It should be noticed that the removal percentage is inversely proportional to flow rates (during the high speeds, the retention time decreased compared with the lower rates) because of the effect of electrolysis time on the residual Fe and SO_4 concentration. The outcomes of these figures are matching [68-75]. Fig. 9 reveals that as the flow rate decreases, the removal rate of Fe and SO_4 increases, and the dissolution rate of aluminum increases. For concentrated water with two plates using the SO_4, the Fe removal percentage reached 14% and 53%, respectively, while for Al, the increase reached 70%. Fig. 10 shows the impact of the flow rate on the removal rate of Fe, SO_4 and the increase of the dissolution of aluminum. For concentrated water and four plates using the SO_4, the Fe removal percentage reached 47% and 79%, respectively, while for Al, the increase reached 79%. Fig. 11 shows that the removal rate for SO_4 and Fe for the lower speed is higher than that for the higher speed. As SO_4, Fe removal was 23% and 29% for 600 L/h speed, and the increasing percentage for Al was 76%. Fig. 12 shows that the removal rate for SO_4 and Fe for the lower speed is higher than that for the higher speed for four plates used in treating river water. As SO_4, Fe removal was 32% and 56% for 600 L/h speed, and the increasing percentage for Al was 90%.

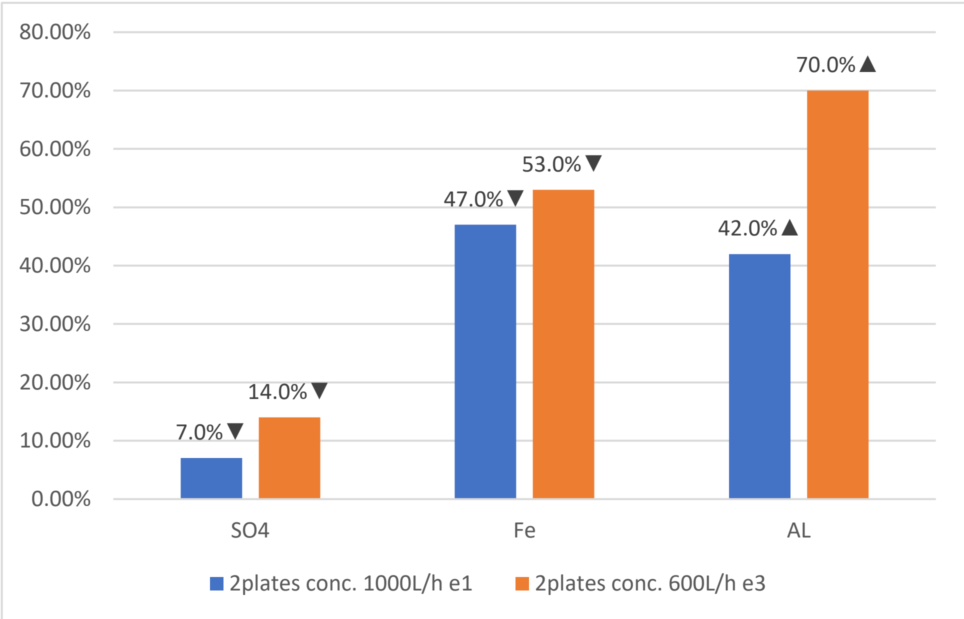


Fig. 9 The Effect of Flow Rates for Two Plates for Concentrated Water.

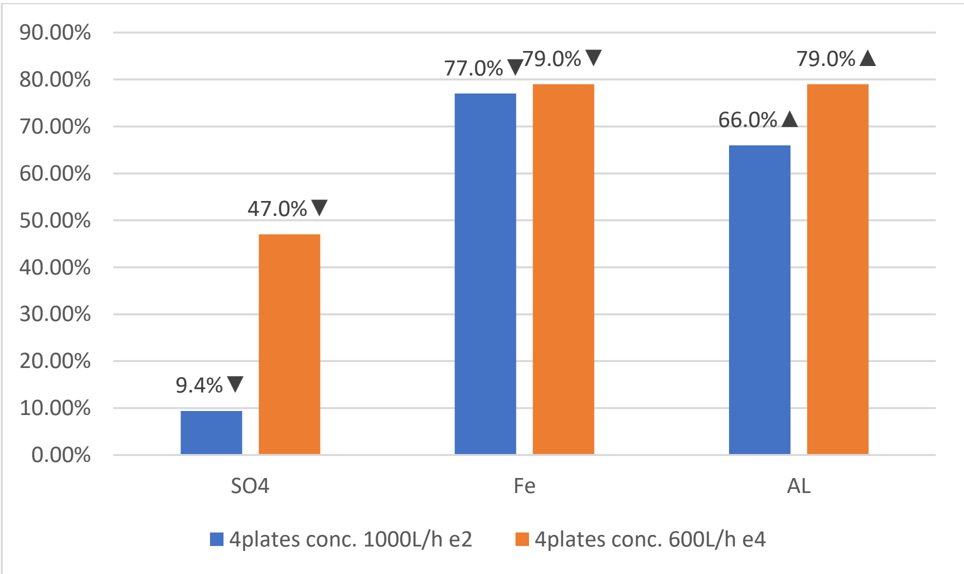


Fig. 10 The Effect of Flow Rates for Four Plates for Concentrated Water.

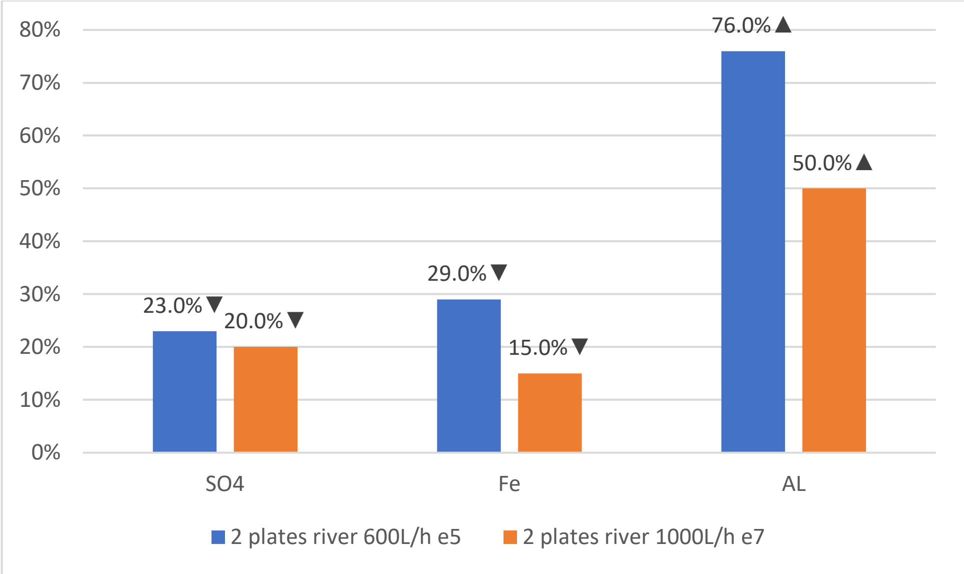


Fig. 11 The Effect of Flow Rates for Two Plates for River Water.

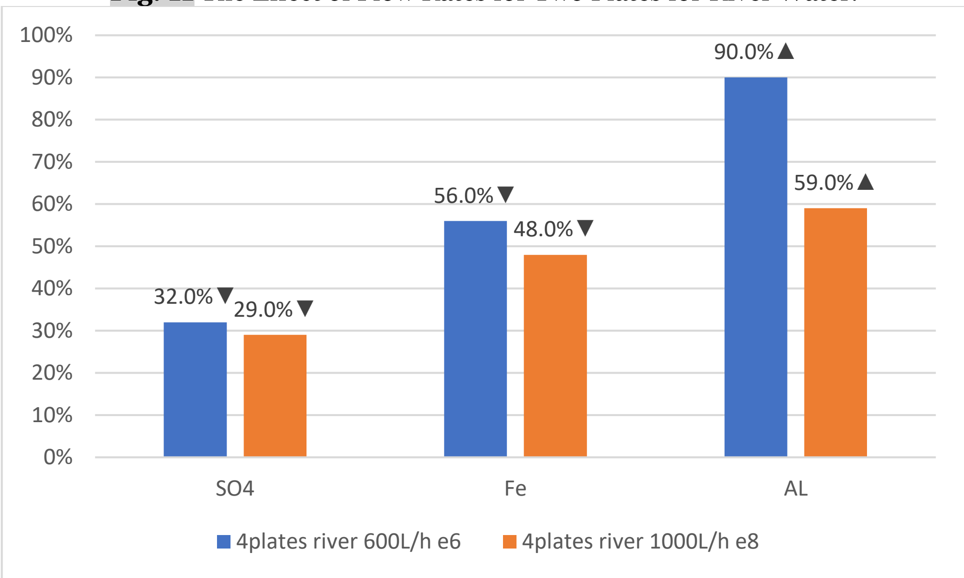


Fig. 12 The Effect of Flow Rates for Four Plates for River Water.

4.CONCLUSIONS

The removal of sulfate and iron from two types of water (river water and rejected water from a reverse osmosis system) was significantly influenced by the number of electrodes and flow rate, as demonstrated in continuous experiments employing monopolar and bipolar aluminum electrodes. The study outcomes highlighted the effectiveness of a continuous flow electrocoagulation reactor equipped with Al plates that have strategically placed holes, facilitating the distribution of coagulants in water samples through an innovative approach. These results are similar studies of Elnenay et al. [64] and Apshankar and Goel [68]. The highest sulfate removal for river water was 50% (for four aluminum plates and 1000 L/h flowrate), and the lowest was 20% (for two plates and 1000 L/h). While for concentrated water, the best removal reached 47% (for four plates with 600L/h), and the minimum reduction was 7.04% (for two plates and 1000L/h). The best removal for iron reached 56% (for four plates and 600L/h) for river water samples, while the minimum reduction reached 15% (for two plates and 1000L/h). For concentrated water samples, the best removal reached 79% (for four plates and 600L/h), and the lowest was 47% (for two plates and 1000L/h). For river water samples, the best iron removal achieved 56% (for four plates and 600L/h), while the lowest removal achieved 15% (for two plates and 1000L/h). The best removal for concentrated water samples was 79% (for four plates and 600L/h), while the lowest was 47% (for two plates and 1000L/h).

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