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Hardness Removal by A Continuous Flow Electrochemical Reactor from Different Types of Water

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Abstract

The present study focuses on the technique of hardness removal by using a novel reactor performing an electrocoagulation (EC) process. The variation of alkalinity is also recorded. Continuous flow experiments were conducted for Total Hardness (TH) removal using a transparent plastic reactor using aluminum plate electrodes that have holes so that the water flows through the plates in a zigzag way. The influence of various operating parameters such as the number of plates (two and four), flow rate (600, 1000 L/h), and water type (Tigris River & rejected water from Reverse Osmosis system RO) was investigated. The results showed that an increase in the number of electrodes led to an increase in the total hardness removal efficiency. In addition, the increase in the flow rate led to a decrease in the removal efficiency. For the rejected RO water type, the highest hardness removal rate was 16.16% for 4 plates electrodes and 600 L/h flow rate while for the river water was 29% for 4 plates electrodes and 1000 L/h.

Keywords: Aluminum; Electrocoagulation; Hardness; Reverse osmosis rejected water; River water

1. Introduction

High levels of hardness in water have been associated with a variety of health problems, including cardiovascular illnesses [1] and Nephritic diseases [2]. Water hardness causes several issues for life and industry. Some problems associated with water hardness in industrial and domestic use especially scale accumulation in the boilers, boiler power output, boiler thermal efficiency, and plugs in the pipeline.

Some of the treatments used for removing hardness are electro-dialysis, chemical precipitation, lime soda process, adsorption, and anti-scale magnetic treatment

Water hardness can also be removed using a variety of methods, including chemical compounds, ion exchange resins, reverse osmosis, and nanofiltration. Each of these methods has some drawbacks, including increased sludge production, permanently hard water, high annual running costs, silt buildup on membranes necessitating effluent post-treatment, and disposal of remaining sludge. Strict environmental restrictions in recent years have demanded new procedures for effective and appropriate water treatment with relatively low operational costs [3]-[4]. One of the most recent techniques for treating water is the electrocoagulation method [5].

Electrocoagulation is a method of electrochemical separation that applies electrical current between groups of metal sheets known as anodes and cathodes [6].

Electrodes are submerged in an electrolyte or conducting solution and linked to a current source and control device by an electrical circuit [7].

Due to their greater ability to remove the majority of pollutants from water media, increased wastewater treatment efficiency, lower cost, lack of hazardous chemicals, and ease of use, electrochemical technologies have drawn a lot of interest in the field of water and wastewater treatment [8].

The primary mechanisms for removing pollutants from the electrocoagulation process are described as "adsorption and charge neutralization, and enmeshment in the precipitate (also known as sweep floc coagulation)" and particle bridges [8]-[9].

There are three phases in the method: (1) Water molecule hydrolysis at the cathode, generating $H_{2(g)}$ bubbles and reacting OH^- ions with metal ions to form the coagulant; (2) electrolytic oxidation at the anode, and $O_{2(g)}$ in the solution; (3) contaminants adhering to the coagulant and floating to the surface for removal through sedimentation [10].

The electrocoagulation method is based on releasing active coagulant ions into solution by the corrosion of iron or aluminum anodes [11]. The use of electrocoagulation technology, which uses only a low-intensity electrical current and can therefore be powered by environmentally friendly methods like solar cells, wind turbines, and fuel cells, is a straightforward, dependable, economical, and promising method for treating various wastewaters without the use of additional chemicals and reducing the amount of produced sludge compared to chemical coagulation [12]-[13]. Because of their size and lower water binding, the flocs produced by electrocoagulation are readily removed by filtering. The lowest necessary solution conductivity limits the use of electrocoagulation to treat effluents with low concentrations of dissolved solids [13]-[14].

There are three sequential phases from ion production to floc formation: "Micro coagulant" is created when metal cations are separated from the anode by the action of an electric field,

interact with suspended water particles, and sink together by colliding flocs. Coagulation also creates a sludge layer for holding suspended solid particles [15].

When negatively charged hydroxide ions generated by the cathodes bond with positively charged ions, colloidal foci are created. When metal cations are hydrolyzed, positively charged hydrolysis products are created, which subsequently adsorb negatively charged contaminants, resulting in the amorphous state of the metal cations [16].

Reactions on both electrodes (anode and cathode) are represented by equations 1, 2, and 3: [17]

$$\mathrm{Al} \rightarrow \mathrm{Al3} + (\mathrm{aq}) + 3\mathrm{e} - \qquad (1)$$

Cathode

$$\mathrm{2H2O} + 2\mathrm{e} - \rightarrow \mathrm{H2} + \mathrm{2OH} - \qquad (2)$$

$Al3++4OH- \rightarrow Al(OH)4-$ + monomeric or dimeric $Al \rightarrow Al13$ polymer + $OH- \rightarrow$ colloid and precipitated Al + $OH- \rightarrow Al(OH)3$ (3)

Both the cathode and the anode electrodes are in contact with the water when using the traditional equipment currently in use for hardness reduction. Both producing alkalinity and acting as a surface for the deposition of scale are tasks carried out by the cathode. The cathodic and anodic environments are not isolated by any media. In contrast to the majority of the water, which has a normal pH level, high pH circumstances only exist at a narrow border layer close to the cathodic surface. As a result, only the water layer next to the cathodic surface experiences the precipitation process. Electrolytic scale removal setups must regularly remove the scale that has accumulated on the cathode to function properly [18].

High pH provoked the precipitation of Hardness around the cathode, this environment acts to convert the HCO3- ion into the CO3-2 form. the ensuing high supersaturation level of CaCO3 promotes its precipitation. as indicated in the following equations 4, 5, and 6 [19]

$$\mathrm{HCO3} - + \mathrm{OH} - \rightarrow \mathrm{CO32} - + \mathrm{H2O} \qquad (4)$$

$$\mathrm{CO32} - + \mathrm{Ca2} + \rightarrow \mathrm{CaCO3} \qquad (5)$$

Also, the high pH conditions promote the precipitation of magnesium hydroxide:

$$\mathrm{Mg} + 2 + \mathrm{2OH} - \rightarrow \mathrm{Mg(OH)2} \qquad (6)$$

By precipitating divalent ions on the cathode and adhering to Al(OH)3n flocs generated by the EC cell under near-neutral pH (for example, pH 6–8), the hardness of the water is removed [20]-[23]. Fig. 1 shows a schematic description of the separation process of the pollutants [24].

The bicarbonate ion is the one that contributes to the alkalinity of river water. Bicarbonates, which are the main kind of alkalinity in natural waterways, are produced when carbon dioxide from the atmosphere is partitioned and when carbonate minerals in rocks and soil weather [25].

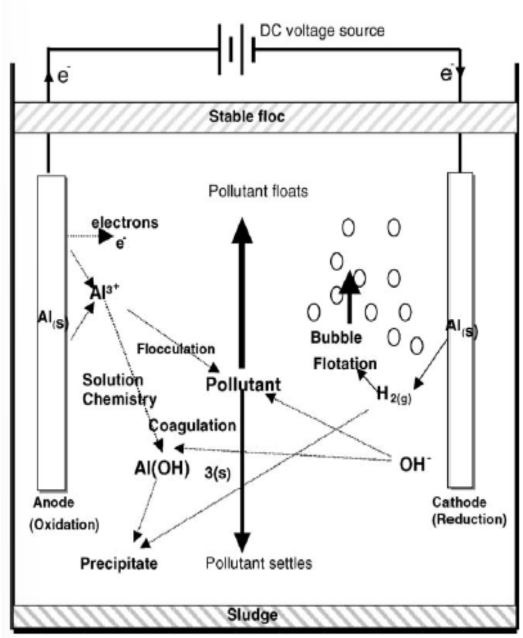

Figure 1 schematic for pollutant separation during the EC process[24].

Total alkalinity, in its simplest form, is an indicator of the extent to which water can withstand pH changes [25]. Alkalinity (Alk), or the ability of water to neutralize acids, is attributed to the presence of bases in salt water, which may absorb a proton from the addition of acid [26].

Numerous advantages of electrocoagulation treatment include flexibility, cheap capital expense, energy economy, and compatibility with the environment. By adding parallel units, it can readily scaled up [27]. Since pollutants are eliminated without the use of chemicals, there is no longer a need to carry, handle, or store chemicals. This results in cost savings, automation, more convenient operations, and a better environment for the health and safety of the operators. Additionally, electrocoagulation creates less hydrophobic sludge than coagulation and flocculation treatment, necessitating less dewatering [27]-[28]. In addition, molecules like hydrogen peroxide, which are created when water oxidizes, can oxidize harmful species into non-toxic ones [28].

In this study, a novel electrolytic cell was prepared to remove hardness from the Tigris River and reject water from the reverse osmosis system (water that is concentrated with salts), using two different numbers of electrodes for 1000 and 600 L/h flow rates. The change in alkalinity was investigated during each experiment.

2. Materials and Methods:

The EC experiment was conducted using a plastic reactor the total volume of the electrolytic cell was 120,000cm3 (0.12 m3) (120 Liter) operating in continuous mode as shown in Fig. 2. Aluminium plates, positioned vertically, were used as electrodes for both the anode and cathode. These plates had dimensions of 50 cm in length, 50cm in width, and 0.5mm in thickness, as depicted in Fig. 3. Before each experiment, the electrodes underwent a cleaning process. After scraping the plates with fine sandpaper and cleaning them for five minutes with a 5% hydrochloric acid solution, they were washed with distilled water and dried.

This cleaning procedure aimed to remove any passivation film formation on the electrodes. The electrodes were connected in a bipolar parallel connection configuration. whereby only the first electrode is connected to the anode and the last electrode is connected to the cathode. The positive and negative terminals of a DC power supply were linked to the anode and cathode plates (model: S-480-48, DC output: 48V- 10A). The applied voltage and current were 36 V and 2-3 A respectively.

The work was conducted at room temperature, the pumps used for this experiment were submerged pumps with two different flow rates. Fig. 4 illustrates the two pumps. Table 1 shows the operating variables used for each experiment used for the research.

Table 1 The operating variables used for each experiment

Experiment number	**Number of electrodes**	**Water type**	**Flow rate (L/h)**	**Alkalinity (mg/L) as CaCO3**	**Hardness**	**Conductivity μS/cm2**
e1	2	rejected	1000	180	430	3309
e2	4	rejected	1000	260	420	3475
e3	2	rejected	600	240	420	3406
e4	4	rejected	600	360	400	3345
e5	2	River	600	150	460	890.3
e6	4	River	600	650	140	1009
e7	2	River	1000	110	380	1039
e8	4	River	1000	850	443	929.9

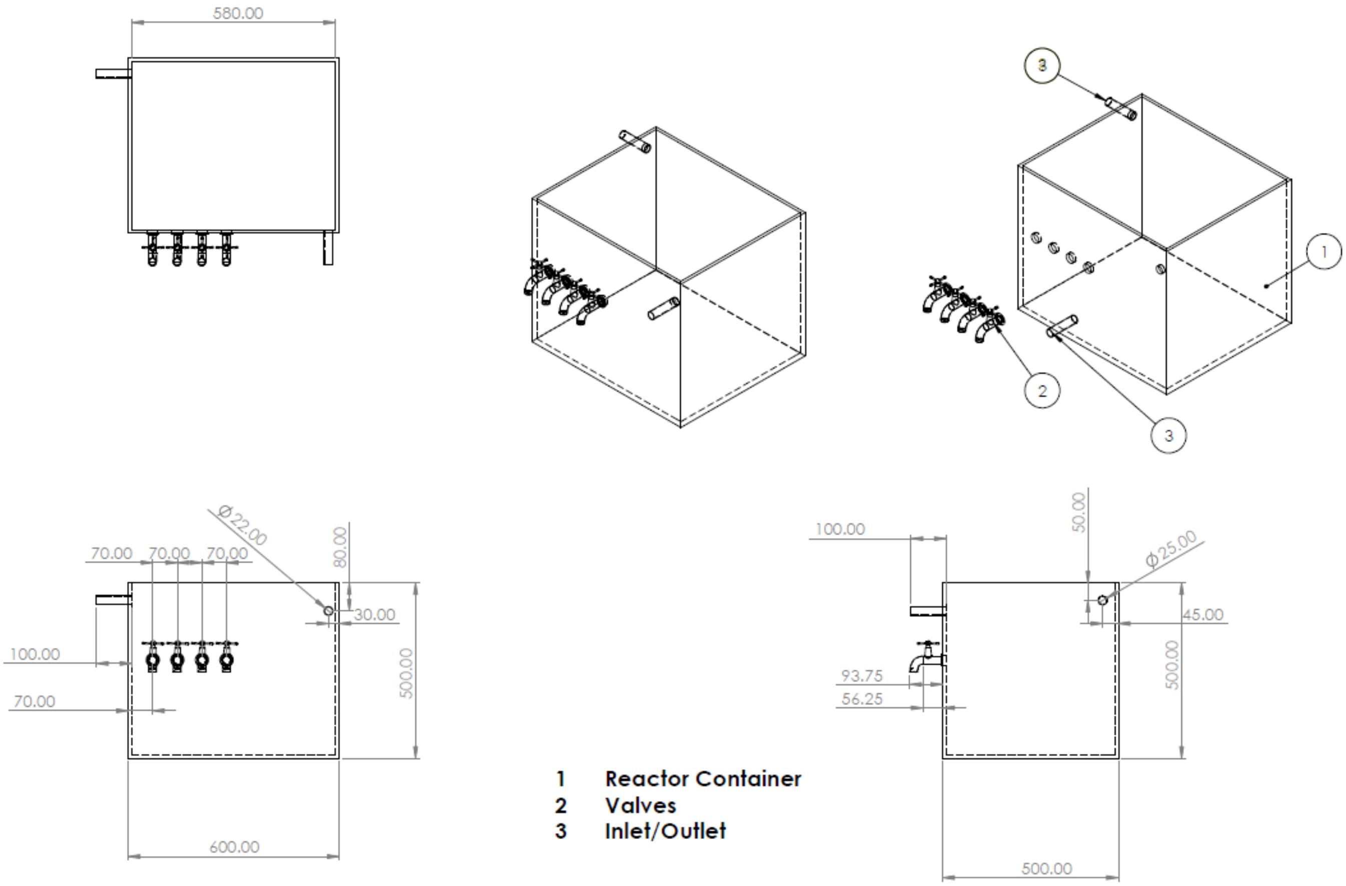

Figure 2 The schematic of the reactor

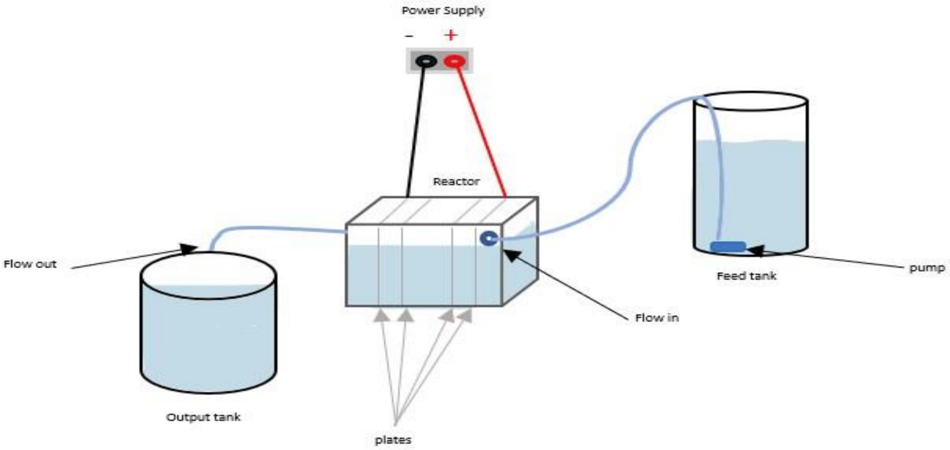

Figure 3 The schematic of the process

Figure 4 the plates of the anode and cathode

3. Results and discussion:

Total hardness and alkalinity were the main targets studied in this work. Two parameters were examined in this paper, the number of plates (2 and 4) used and the flow rates for the process (600 and 1000 L/h). The method used for measuring the TH and Alkalinity is titration. Total hardness was tested using the standard EDTA titration method [29]-]30] With aluminum coagulants, the creation of positively charged monomeric and polymeric particles results in the charge neutralization process at low metal concentrations and pH ranges of 4 to 7. At pH 7, these positive particles are transformed into $Al(OH)_3$ precipitate [31].

The change percent was calculated by equation 1:

$$C\% = \frac{(in-out)}{in} \times 100\% \qquad (7)$$

Where C is the change percent, for hardness it is considered as the removal percent while for alkalinity it is the change percent as the alkalinity sometimes increases and in others it decreases.

The energy consumption is represented by the equation:

$$energy\ consumption = A \times V \qquad (8)$$

Where A is the current and V is the voltage, the energy consumption during the process is equal to 108 W.

The flow of water would be across the holes of the plate so that the plates' arrangements with holes might improve the water's ability to mix and disperse pollutants, improving the effectiveness of the EC process.

The presence of holes created alternating upward and downward flow paths, facilitating improved contact between the electrodes and contaminants, thereby enhancing the coagulation and flocculation reactions.

Different researchers have opinions when it comes to changes in alkalinity. Some argue that it decreases, while others claim that it increases. As a result, there are findings regarding the fluctuations in alkalinity.

According to the results of Foudhaili et al., electrocoagulation works better than chemical coagulation. This improved performance is partly due to the hydroxyl ions (OH_(aq) produced at the cathode and the lack of sulfate anions [32].

However, in the study of Saiba et al, it is well noted that the alkalinity decreases after the EC process [33]-[34]. On the other hand, the EC procedure had no noticeable impact on the water's alkalinity, which is essential since some alkalinity is required to avoid sudden pH fluctuations as mentioned in Franco et al. [35].

3.1 Effect of number of plates (surface area):

The number of electrodes used in the electrocoagulation process can have a significant impact on its efficiency and effectiveness. Two and four plates were used in this work. The purpose of using different numbers of electrodes was to examine the impact of electrode surface area.

The electrodes dissolving and producing a lot of aluminum metal ions have a significant effect on the EC efficiency [36]-[37], This is enhanced by adding more plates to have a greater active surface. It is crucial to keep in mind that the number of plates should be raised in a way that prevents the development of considerable ohmic resistance, and this relies on the kind of electrode layout and the number of electrodes employed [38]-[41].

The outcomes support the idea that increasing the number of electrodes improves the removal efficiency, i.e., the more electrode surface area there is per reactor volume, the better the reactor is at removing pollutants from a solution. This is demonstrated in Fig. 5 through 8 of the study. These findings match those obtained by eliminating cadmium [41], COD removal [42], Total dissolved solids and turbidity [43], and non-sugar removal [44] Consequently, high current efficiency was caused by the large surface area of the electrodes (the cross-sectional area for the current supply) as the number of electrodes rose [45]. Additionally, the W/m^2 Pd power density (Pd) needed to give any constant coagulant dosage decreases as the electrode surface area increases [46].

The change percent was calculated by Equation 9:

$$change\ \% = \frac{(in-out)}{in} \times 100\% \qquad (9)$$

For hardness, the change is basically positive since it is removing, while in alkalinity calculation in some cases it is increasing percent and in others, it is decreasing.

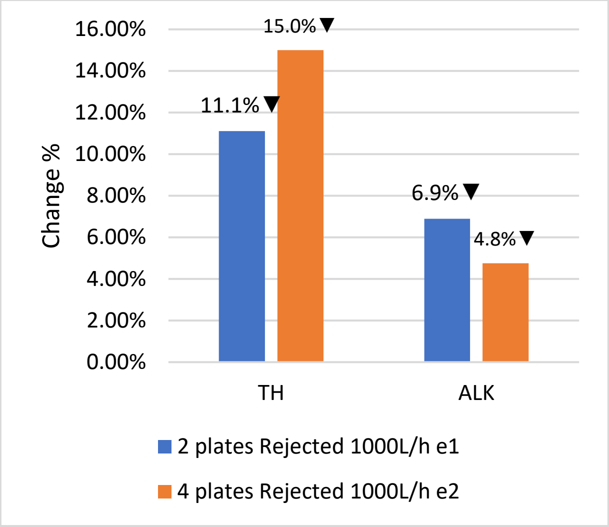

Figure 5 The effect of the number of plates used for RO rejected water for flow rates of 1000 L/h.

Fig. 5 displays a result for the RO-rejected water at a flow rate of 1000 L/h. The hardness removal rates for the two and four-plate processes, respectively, are 11.11% and 15%. In terms of alkalinity, the operations involving two plates and four plates showed declining rates of 6.9% and 5%, respectively.

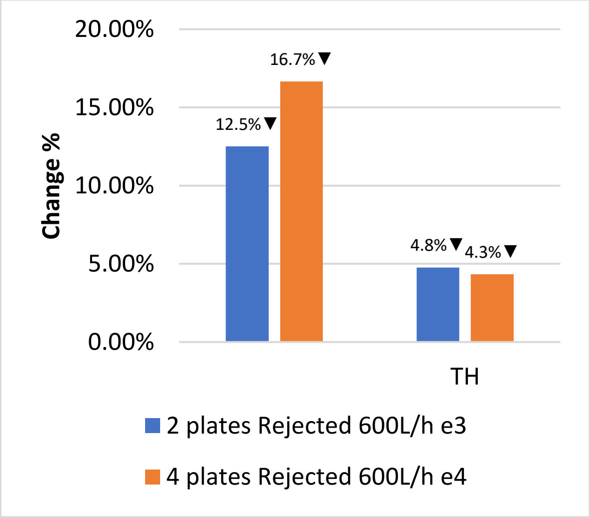

Figure 6 the effect of the number of plates used for rejected RO water for flow rates of 600 L/h.

The results for the RO-rejected water with a flow rate of 600 L/h are displayed in Fig. 6, where the hardness removal rates for the two and four-plate processes are 12.5% and 16.66%, respectively. In terms of alkalinity, the operations using two plates and four plates showed declining rates of 4.76% and 4.34%, respectively.

For river water experiments, it is clear that there are some differences regarding the amount of removal and this is a result of the conductivity differences between the river water and rejected water, as the rejected water has higher conductivity than that of the river.

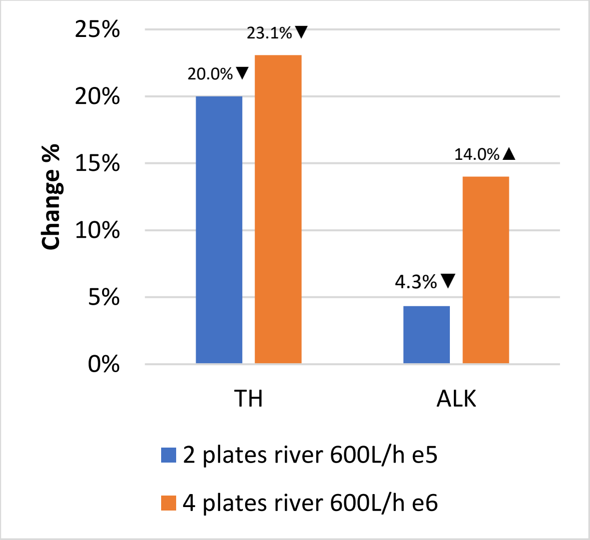

Figure 7 the effect of the number of plates used for river water for flow rates of 600 L/h.

As can be shown in Fig. 7, the removal rate of hardness for river water at a flow rate of 600 L/h is equivalent to 20% for two plates and 23.07% for four plates, respectively. In terms of alkalinity, there was a 14% rise for four plates compared to a 4% decrease for two.

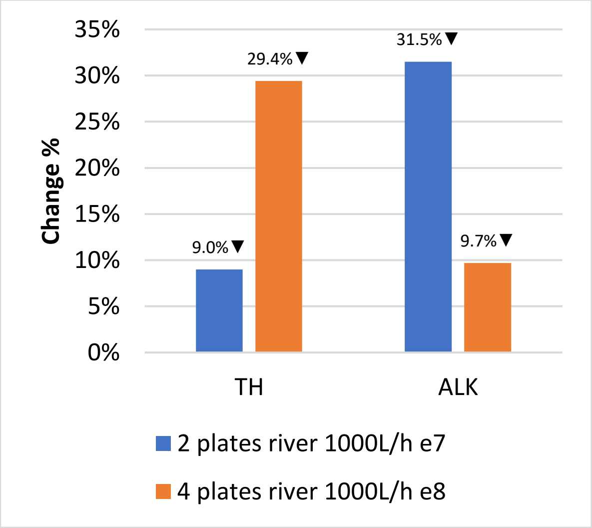

Figure 8 The effect of the number of plates used for river water for flow rates of 1000 L/h

The results for river water at a flow rate of 1000 L/h are displayed in Fig. 8, where the hardness reduction rates for the two and four-plate processes are 9% and 29.4%, respectively. In terms of alkalinity, the operations using two plates and four plates showed declining rates of 32% and 9.7%, respectively.

Since they enable increasing the active area of contact with the medium and reducing electrical consumption, the number of electrodes and agitation were crucial factors for enhancing efficiency [47]-[48].

The findings indicated that increasing the number of electrodes had a positive impact on improving removal efficiency, requiring the usage of many electrodes to achieve a given efficiency in a short time [49].

3.2 The effect of flow rate:

The amount of aluminum released to a system using aluminum electrodes should theoretically depend on the current and time of the electrolysis, according to Faraday's law. Aluminum will thus be released into the system at a higher rate as a result of the interaction between current and residence duration (charge)[50].

To calculate the theoretical quantity of total aluminum, Faraday's law is utilized to connect the current flow (I for time t) to the quantities (m) of aluminum and hydroxide ions produced inside the reactor. The equation shows the faraday’s law.

$$m = \frac{I \times t \times M}{ZF} \tag{9}$$

where M is the molecular weight (g/mol^{-1}), Z is the number of electrons transported in the process at the electrode, and F is Faraday's constant (96486 C mol^{-1}) [51].

Equation 1 which is derived from Faraday’s law shows that at constant current, the input flow rate Q has an inverse relationship with the theoretical Al concentration in the liquid phase. This shows that shortening the residence period of the liquid phase at constant cell volume dilutes the coagulant even if it has no effect on the rate of coagulant formation. Nevertheless, despite the fact that the input concentration of pollutants does not change with the amount of time the liquid phase is present, the flow rate of pollutants increases in accordance with Q. Due to that, the ratio of coagulant to pollutant concentrations and the ratio of pollutant to coagulant residence time scale drop in the same way.

$$C_{th}=\frac{(\Delta \mathrm{mth}/\Delta t)}{Q}=\frac{M}{Q}\ \frac{I}{ZF} \tag{10}$$

M is the metal's molar mass, Z is the number of electrons transferred, F is the Faraday constant, and Q is the liquid phase's volumetric flow rate. The experimental mass loss of the electrodes per unit of time ($\Delta \mathrm{mth}/\Delta t$) outlet concentration of aluminum (Cth) in the coagulant species. This is derived in Eq. 2 from Faraday’s law[52].

With increasing flow, the Faradaic efficiency shows a minor drop in voltage and power per coagulant dosage [53].

Hydraulic retention time depends on flow rate, which also impacts how quickly coagulant ions dissolve in the solution, which in turn affects how effectively pollutants are removed [24]. Low flow rates allow for more coagulant dosages to be used for the same amount of contaminants, which increases removal effectiveness. the removal efficiency fell as flow rates rose for a given voltage or current density.in each of these studies [50], [54]-[60].

Since slower flow rates result in longer residence times, it is predicted that removal efficiency will decline as the flow rate rises. This may be because insufficient amounts of aluminum hydroxide complexes are formed. A longer residence time indicates that the pollutant solution is allowed to remain in the electrocoagulation cell for a longer time, giving the reaction more opportunity to occur. A result, Consequently, the longer the time that pollutants spend in contact with aluminum hydroxides, the slower the flow rate at the constant cell and current, the longer the liquid phase's residence time at constant coagulant production rate, the higher the mass ratio of coagulant to pollutant, and the longer the time that pollutants spend in contact with coagulants, the more likely it is that the pollutants will be adsorbed and be neutralized [50],[52],[61]-[64]. Because they may promote either turbulent flow or collisions between contaminant molecules and active species [64]-[65].
At the lowest inlet flow rate, the influence of current density at a constant flow rate was more noticeable [52].

The coagulant produced by electrochemical oxidation of the anode material and the pollutant molecules can properly combine at the slower flow rate, increasing the rate of coagulation. If not, a flow reactor's residence time decreases at increasing flow rates, thereby slowing down the coagulation rate [62].

The experiment results show that as the intake flow rate was increased, it took less time to reach steady state conditions. This is explained by the fact that the flow rate is inversely proportional to the change in residence time. Because it is known that operating time increases with decreasing current density under steady-state circumstances which are the same results found by Merzouk et al. and Kobya et al. [52].

Fig. 9 through Fig. 12 show the same result as removing seawater organic matter [52], Copper, lead, and cadmium removal efficiency [66], fluoride removal efficiency [50], organic matter removal efficiency [64], DOC removal efficiency [52], arsenic [67].

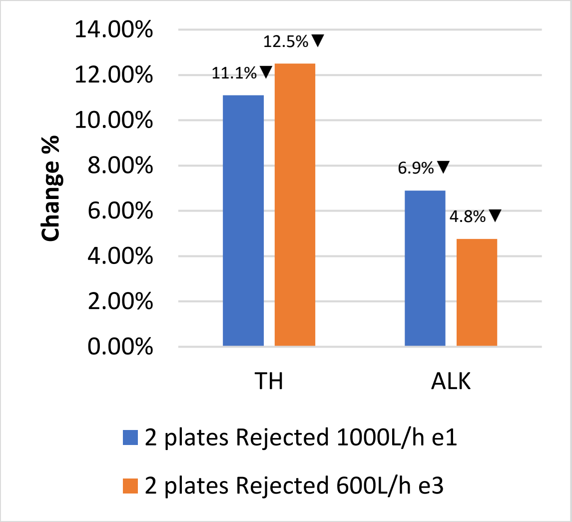

Figure 9 The effect of flow rates for 2 plates for RO rejected water

The impact of flow rate on the hardness removal rate is seen in Fig. 9. For two plates of RO-rejected water, the hardness removal rate at 1000 L/h flow (11.11%) was less than that at 600 L/h flow (12.5%). The rate of decrease in alkalinity is 6.9% for 1000 L/h and 4.76% for 600 L/h.

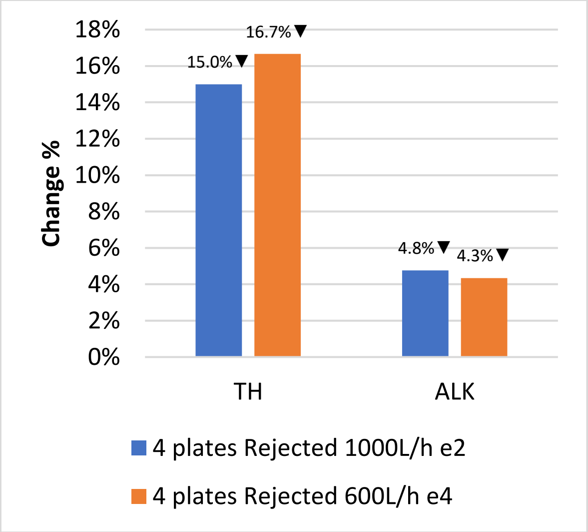

Figure 10 The effect of flow rates for 4 plates for RO rejected water

The impact of flow rate on hardness removal rate is seen in Fig. 10. The removal rate of hardness for RO-rejected water for four plates was lower at a 1000 L/h flow rate of 15% than at a 600 L/h flow rate of 16.66%. The rate of decrease for alkalinity is 5.34% for 1000 L/h and 4.34% for 600 L/h.

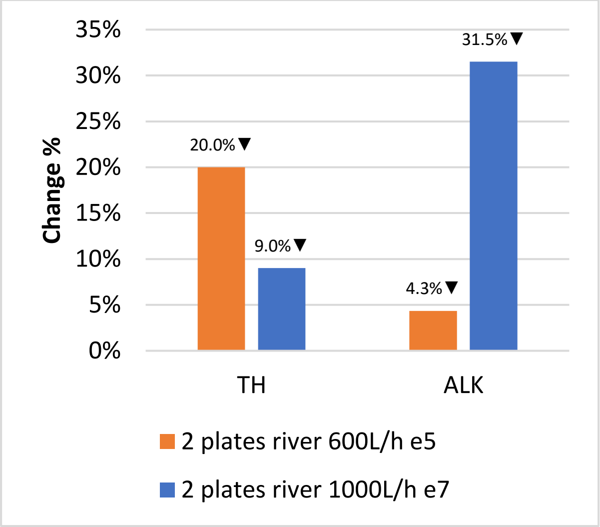

Figure 11 The effect of flow rates for 2 plates for river water

Fig. 11 shows the effect of the flow rate on the removal rate of hardness. For RO rejected water for 2 plates where the removal rate for hardness at 1000L/h flow 9% was lower than that for 600 L/h 20%. For alkalinity, the decreasing rate for 1000 L/h is 32 % and for 600 L/h is 4 %.

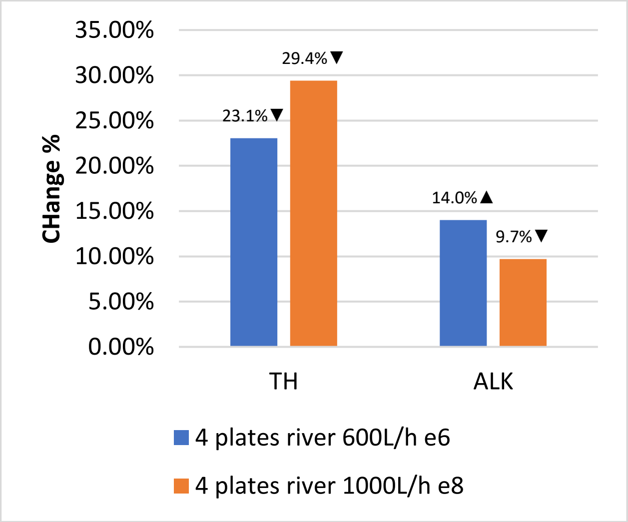

Figure 12 The effect of flow rates for 4 plates for river water

The impact of flow rate on hardness removal rate is seen in Fig. 12. For two plates of RO-rejected water, the hardness removal rate at 1000 L/h flow was 29.4%, but it was 23.07% at 600 L/h flow. In terms of alkalinity, the rate of decrease is 9.7% for 1000 L/h while the rate of increase is 14% for 600 L/h.

4. Conclusions

In chemical coagulation, Polyelectrolyte was generated by using alum. The health risks associated with the presence of residual aluminum ions in the water after treatment, and the issues that arose in the water supply networks as a result of the aluminum compounds. Fouling reduces the service life of the RO membrane, increases operating pressure, lowers filtrate flow, and thus increases the cost of generating water, all of which have a detrimental effect on membrane performance. The quality of the feedwater has a direct bearing on the useful life and performance of a membrane. Fortunately, these issues may be resolved and the RO process maintained by treating the feedwater prior.

Continuous experiments with bipolar and monopolar aluminum electrodes showed that the number of electrodes and flow rate had a substantial impact on the removal of hardness from two types of water (river water and rejected water from a reverse osmosis system). The study's findings demonstrated the use of an electrocoagulation reactor with continuous flow that is fitted with Al plates that have holes positioned at specific intervals to facilitate the dispersion of coagulants in water samples. The results show that as the number of electrodes increased the removal efficiency increased while for the flow rate, the experiments reveal that as the flow rate increased the removal of hardness decreased. The highest hardness removal for river water was 29.4% (for 4 aluminum plates and 1000 L/h flowrate), and the lowest was 9% (for 2 plates and 1000 L/h). While for concentrated water, the best removal reached 16.66% (for 4 plates with 600L/h), and the minimum removal was 11.11% (for 2 plates and 1000L/h). The alkalinity during the experiments was varied and decreased for all experiments except for the river water that used 4 plates with a flow rate of 600L/h the alkalinity increased.

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Conflict of interest:

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

Author Contribution Statement

Shahad and Naseer proposed the research problem.

Hussein and Aiman developed the theory and performed the computations.

Shahad and Dhuha verified the analytical methods and investigated [a specific aspect].

Naseer supervised the findings of this work.

All authors discussed the results and contributed to the final manuscript.

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