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Keywords: electrocoagulation, electricity, electrode consumption, desalination, static anode reactor

Abstract:

Saline water treatment has become increasingly important for drinking water supplies. The aim of this study was to evaluate the ability of the electrocoagulation (EC) process with combined aluminum electrodes in removing various types of salt from water samples collected at Sawa Lake, Al-Muthanna, Iraq. The targeted types of salt include total dissolved solids (TDS), chloride salt (Cl⁻), bromine (Br⁻), and sulphate (SO₄²⁻). A bench scale consisting of combined EC configurations with static electrodes was employed under combined electrical connections. The effect of the six variables factors, such as applied current density (I), reaction time (RT), pH, temperature (T), stirring speed (Mrpm) and inter electrode distance (IED) were observed to achieve a higher removal of TDS, Cl⁻, Br⁻ and SO₄²⁻. Initial results showed the following optimum operating conditions: I = 2 mA/cm², RT = 80 min, pH = 8, T = 25 °C, IED = 1 cm and Mrpm = 500. The maximum removal efficiency of TDS, Cl⁻, Br⁻ and SO₄²⁻ were 91%, 93%, 92% and 90%, respectively. It can be concluded that the EC method applied in the present study was effective to removing salts from lake water.

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Article

Treatment of Saline Water Using Electrocoagulation with Combined Electrical Connection of Electrodes

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Abstract: Saline water treatment has become increasingly important for drinking water supplies. The aim of this study was to evaluate the ability of the electrocoagulation (EC) process with combined aluminum electrodes in removing various types of salt from water samples collected at Sawa Lake, Al-Muthanna, Iraq. The targeted types of salt include total dissolved solids (TDS), chloride salt (Cl^-), bromine (Br^-), and sulphate (SO_4^{2-}). A bench scale consisting of combined EC configurations with static electrodes was employed under combined electrical connections. The effect of the six variables factors, such as applied current density (I), reaction time (RT), pH, temperature (T), stirring speed (M_{rpm}) and inter electrode distance (IED) were observed to achieve a higher removal of TDS, Cl^- , Br^- and SO_4^{2-} . Initial results showed the following optimum operating conditions: $I = 2 \text{ mA/cm}^2$, $RT = 80 \text{ min}$, $\text{pH} = 8$, $T = 25 \text{ }^\circ\text{C}$, $\text{IED} = 1 \text{ cm}$ and $M_{rpm} = 500$. The maximum removal efficiency of TDS, Cl^- , Br^- and SO_4^{2-} were 91%, 93%, 92% and 90%, respectively. It can be concluded that the EC method applied in the present study was effective to removing salts from lake water.

Keywords: static anode reactor; desalination; electrode consumption; electricity; electrocoagulation

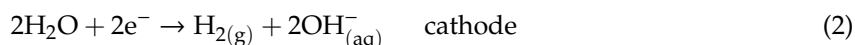
1. Introduction

In recent years, freshwater resources have been declining worldwide, thus requiring treatment and recycling of water. Since seawater represents more than 98 percent of the water available on Earth, desalination is an important process to treat the freshwater scarcity problem [1,2]. Desalination indicates the exclusion of minerals and salts from used water. Some technologies like thermal distillation, reverse osmosis (RO), adsorption, flotation, nanofiltration and electrochemical, electrocoagulation (EC), electrodeposition, electrooxidation (EO) and electroflotation (EF) can be used to treat polluted water. However, most of the discussed methods have some major drawbacks [3–9].

In regards to thermal desalination, the energy consumption and costs are approximately 13 kWh/m^3 for water desalination at 45% retrieval [10]. Thus, thermal desalination produced approximately 40%

of the water desalinated in the world annually [11]. RO is the dominant technology in the last three decades for many countries, particularly due to its low energy cost [12]. RO can work at approximately 3.0 kWh/m³ for desalination of saline water at 40% retrieval [13]. However, the presence of chlorine in saline water damages the polyamide membrane [14,15], and the membrane replacement process is very costly [16]. Adsorption and nanofiltration methods are not constantly sufficient to reach the discharge limits [17], while the electrodeposition process requires a long reaction time for the treatment [17,18]. Thus, utilization of the electrocoagulation (EC) method is one of the best alternatives for water desalination.

Electrocoagulation is an electrochemically-driven technology that yields coagulant species in situ from the electrodisolution of sacrificial anodes, usually made of aluminum or iron [19]. An EC reactor mainly consists of an electrolytic cell made of Plexiglas [20], glass [20,21], an anode, and a cathode. The anode and cathode consist of metal plates, with the anode recognized as a ‘sacrificial electrode’. The electrodes are commonly made of the same or different materials, e.g., iron (Fe) [22,23], aluminum (Al) [24,25] or iron and aluminum combined [20,26]. Overall, the EC process utilizes a simple tool and generates a relatively low amount of sludge, and has been successfully employed to remove salts from saline water. This method is characterized by its simplicity, safety, ease of control, selectivity, versatility, amenability to automation, and environmental compatibility. In the EC process, when electrodes have been utilized, aluminum dissolves and serves as a coagulant agent, leading to the release of hydrogen gas at the cathode (represented in Equations (1) and (2)). During the dissolution of aluminum at the anode, different aquatic aluminum species are generated. These types of aluminum act as the coagulants by combining with contaminants to produce insoluble hydroxides, which may be removed from the solution [27], as shown in Equations (1) and (2) below:



where Al is the aluminum anode. The Al ions generated at the anode are soluble and will subsequently react with the hydroxide ions generated at the cathode. The aluminum hydroxides are created as shown in Equation (3) below:



Previous studies have shown that the EC method is an effective tool for treating various water contaminants in textile wastewater [12,20,21,28], chemical fiber wastewater [29], algae or microorganisms [30], oily wastewater [31–33], restaurant wastewater [34,35], sewage water [35], arsenic [36–38], fluoride [39–42], boron [43,44], and nitrite [45,46]. The EC process has also been utilized in the removal of different metal ions from water, e.g. Zn, Ag, Cu, Co, and Mn [47–51]. The aim of the present study is to evaluate the effectiveness of the electrocoagulation method for saline water treatment. Additionally, the study aims to determine the best operational parameters for the removal of total dissolved solids (TDS), chloride salt (Cl⁻), bromine (Br⁻), and sulfate (SO₄²⁻) using a bench scale with combined EC configurations—with static electrodes—under combined electrical connections.

2. Materials and Methods

2.1. Saline Water Samples

The saline water samples were collected from Sawa Lake, Iraq. Sawa Lake is located about 240 km south-west of Baghdad, in the Al-Muthanna province in southern Iraq between 31°18′ N (latitude) and 45°00′ E (longitude) (Figure 1). There is no surface resource feeding the lake, and therefore the main source of water for the lake is groundwater. The total surface area is about 10 km², the mean depth is 5.5 m [52] with an average length and width of 5 and 2 km, respectively. In 2018, the physico-chemical characteristics of saline water at Sawa Lake was reported by the Iraqi Ministry of

Health and Environment. Based on the report, it was found that the average values for pH, conductivity, temperature, turbidity, total dissolved solid (TDS), chloride salt (Cl^-), bromine (Br^-) and sulphate (SO_4^{2-}) were 8.25, 38,062 ($\mu\text{s}/\text{cm}$), 21.4 ($^\circ\text{C}$), 95 ntu, 26,263 mg/L, 8,498 mg/L, 6 mg/L, 6562 mg/L, respectively. Sawa Lake is considered to have the highest salinity among the Iraqi inland waters.

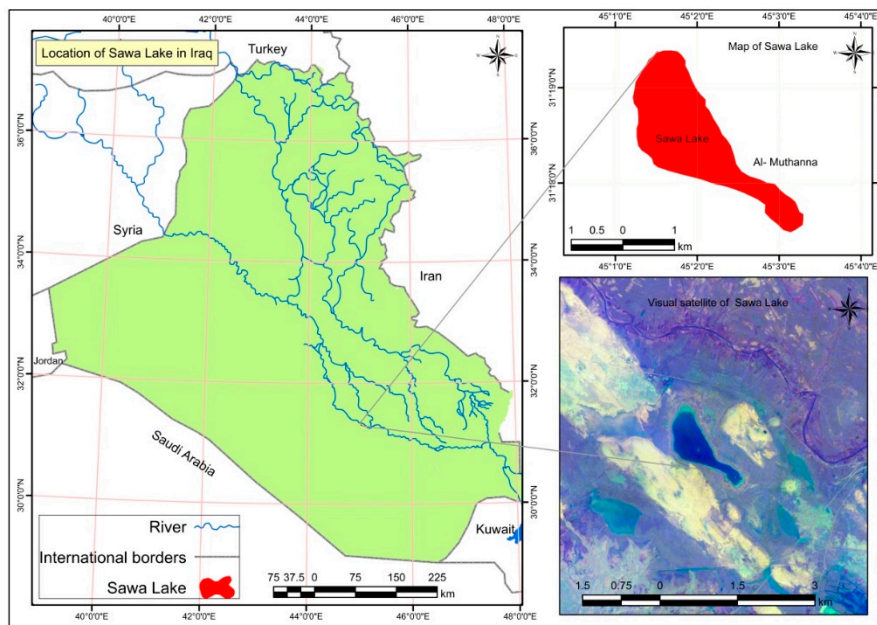


Figure 1. Map of Sawa Lake, Iraq.

2.2. Experimental Setup

Figure 2 illustrates the experimental setup of the EC process used in the present study. The desalination of saline water from Sawa Lake was performed in a batch reactor made of glass with the upper cover of the reactor being made of plexiglas. The saline water was tested for parameters such as TDS, Cl^- , SO_4^{2-} and Br^- . The reactor size used in the experiment had a width of 20 cm, length of 15 cm and depth of 15 cm. The volume of treatment water was 3 L for each run. Seven aluminum plates were connected in parallel with the main power supply: 4 monopolar (Mp) electrodes and 3 bipolar (Bp) electrodes, where the anode and cathode were both monopolar. A total of three inner aluminum electrodes were implemented in a bipolar form. The dimensions of the electrodes for the both anode and cathode plates were $10 \times 10 \text{ cm}^2$ and 0.1 cm thickness, with the distance between the inter-electrodes being 1 cm. Each electrode had a surface area of 100 cm^2 ($10 \text{ cm} \times 10 \text{ cm}$). The collection of electrodes was flooded into the saline water. The anode and cathode group were linked to negative and positive terminals with a DC power supply. A magnetic stirrer was used to mix the electrolyte. The anodic and cathodic reactions took place on the surface of the inner electrodes when the electric current was passed through the electrodes.

In the present study, the DC power supply used was made by YIZHAN, 0–40 V; 0–6 A, China. The effect of the applied current was investigated using different current densities (0.5, 1.0, 1.5, 2.0, 2.5 and $3 \text{ mA}/\text{cm}^2$) during 100 min of reaction time. The applied current was controlled to be stable and constant during the experiment using the programmable power supply by YIZHAN. After every run, the electrodes were completely washed with distilled water. The experiments were carried out in the laboratory and the temperature in the laboratory was set to approximately $26\text{--}28 \text{ }^\circ\text{C}$. The pH of the saline water was adjusted to the required value using concentrated hydrochloric acid and sodium hydroxide.

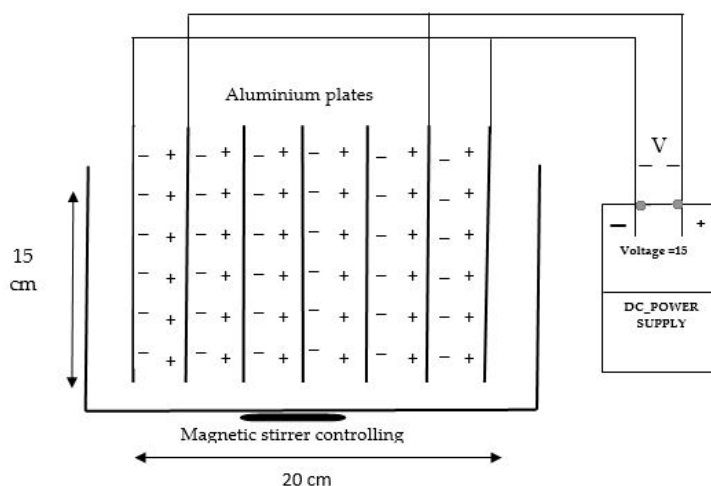


Figure 2. Experimental setup of the electrocoagulation (EC) process.

2.3. Parameters Analysis

Table 1 shows the parameters considered during the removal of TDS, Cl^- , Br^- and SO_4^{2-} from the lake water samples. The reaction time was 20 to 100 min, current density was 0.5 to 3 mA/cm^2 , pH ranged from 5 to 11, temperature was 25 to 45 $^\circ\text{C}$, inter-electrode distance (IED) ranged from 0.5 to 2 cm and stirring speed (S_s) was set at 250 to 750 (rpm). All parameter analyses were tested over 92 runs to obtain the optimal conditions, and the optimal conditions were repeated in 3 replicates to ensure the accuracy and reliability of the results.

Table 1. Parameters included for removing total dissolved solids (TDS), chloride salt (Cl^-), bromine (Br^-) and sulphate (SO_4^{2-}) from lake water.

Parameters	Unit	Range	Run	Replicated Optimal Conditions
Current density	mA/cm^2	0.5 to 3.0	6	3
Reaction time	min	20 to 100	5	3
PH solution	-	5 to 11	8	3
Temperature	$^\circ\text{C}$	25 to 45	5	3
IED	cm	0.5 to 2	4	3
S_s	rpm	250 to 750	3	3

The meter and/or method used for the analyses of parameters during the experiment are listed in Table 2.

Table 2. Parameters and methods of the analyses.

Parameter	Meter/Method
pH	pH meter-pHM84
Turbidity	HACH 2100P
Electric conductivity	HANNA HI-99301
TDS	Gravimetric method
Cl^-	Titration method
Br^-	ion chromatography
SO_4^{2-}	APHA Gravimetric

3. Results and Discussion

3.1. Effect of Applied Current and Time

Applied current (I) is an important parameter, playing a critical role in regulating the reactor rate during the EC process [21]. Specifically, the removal rate of the EC process increases with an increase in reaction time [20,53]. The effect of current (I) not only sets the coagulant dosage, but also the mixing rate of the EC process [26]. In the present study, experiments were conducted over various reaction times, before the optimization runs were carried out using constant optimum operating parameters. The variation in percentage removal of TDS, Cl^- , Br^- and SO_4^{2-} was measured over a fixed time (80 min) at various applied current densities ($I = 0.5, 1.0, 1.5, 2.5, 2.5$ and 3). The results of such investigations into the applied current density (mA/cm^2) are presented in Figure 3a–d.

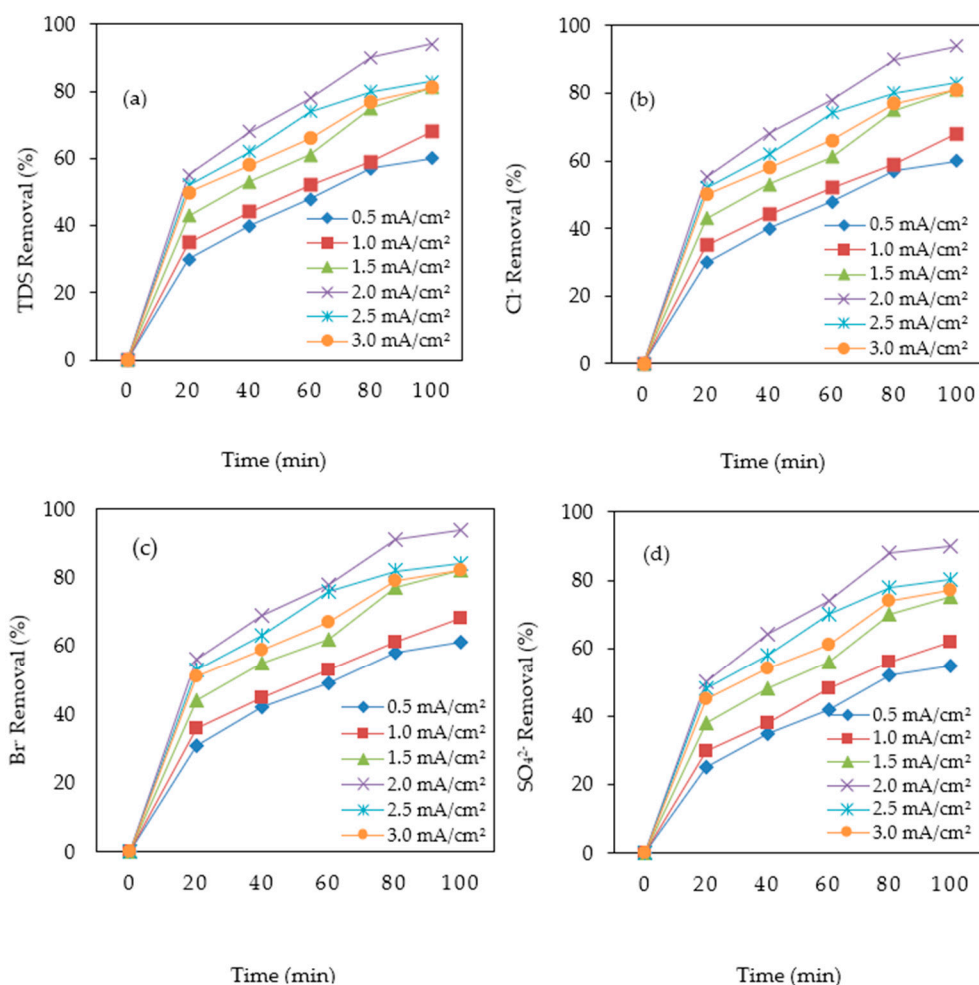


Figure 3. Effect of applied current and reaction time on removal of salts. (a) TDS; (b) Cl^- ; (c) Br^- ; and (d) SO_4^{2-} .

With a set reaction time of 80 min, it was found that an increase in current density from 0.5 to 2.0 mA/cm^2 led to an increase in the removal percentage of TDS, Cl^- , Br^- and SO_4^{2-} from 57 to 90%, 60 to 92%, 58 to 91% and 52 to 88%, respectively. A low removal percentage was obtained with an increase in current density from 2.0 to 3.0 mA/cm^2 for the same reaction time. The increase in the current density up to 2.5 and 3.0 mA/cm^2 (above the optimum current value of 2.0 mA/cm^2) led to the production of a high number of hydroxyl radicals, which caused a rise in the pH of the solution. It is well known that the coagulant breaks down in high pH solutions, which then leads to a reduction in the performance of the EC process. According to the results, the performance of the EC process improves

with an increase in reaction time from 20 to 80 min, where the removal percentage increases from 55 to 90% (TDS), 59 to 92% (Cl^-), 56 to 91% (Br^-), and 50 to 88% (SO_4^{2-}) at 2.0 mA/cm² current density.

The running costs involved in the desalination of water include the electrical, membranes, maintenance, equipment and labor costs, while in the electrochemical process, the most important factors affecting the running costs are the cost of the consumed electrical energy and the electrode materials used [19]. The costs increase with increasing operating time. As shown in Figure 3, there is a minor increase in the removal process when the reaction time is increased from 80 min to 100 min. Conversely, that increase in reaction time (RT) led to an increase in costs, with more electricity consumed and electrode materials used. Therefore, 80 min was selected to reduce power consumption and lower operational costs. Furthermore, increasing the reaction time over 100 min led to a decline in the removal percentage due to the release of metal ions in the solution [54]. Therefore, 2.0 mA/cm² and 80 min were selected as the best operational factors that were also in accordance with results obtained in previous studies [20,55]. The energy consumption per unit of TDS mass required to reduce the salinity of the solution was estimated using Equation (4) [56].

$$\text{Energy consumption (kWh g}^{-1} \text{ TDS)} = \frac{E_{\text{cell}} I t}{V_s \Delta(\text{TDS})} \quad (4)$$

where E_{cell} is the average cell potential (V), I is the applied current (A), t is the treatment process time (h), V_s is the solution volume (m³), and $\Delta(\text{TDS})$ is the difference between the initial and final salinity value.

The optimal parameters—i.e., the 2.0 mA/cm² current density and 80 min treatment process time—were used to estimate the energy consumption. The result showed that the energy required to achieve the optimal removal percentage of TDS (91%) was just 0.21 kWhm⁻¹. Interestingly, this value of energy consumed is considered less than that of RO treatment. The values associated with RO treatment have a wide range of required energy to perform the treatment process, and depend on the membrane materials, technology and the nature of the treated water. In general, the required energy of a RO treatment process is between 3 and 10 kWhm⁻¹ [57], and this value is higher than that required to achieve the treatment of saline water by EC technology.

3.2. Effect of Initial pH

The importance of the effect of pH on water treatment in the EC method has been reported in previous studies [25,58,59]. In the present study, the effect of pH on TDS, Cl^- , Br^- and SO_4^{2-} removal were investigated by adjusting the initial pH in the interval pH from 5 to 11 with optimum conditions (Mp Al–Bp Al, $I = 2.0 \text{ mA/cm}^2$) and a reaction time of 80 min. Figure 4 shows that at pH 8, the removal percentage of TDS, Cl^- , Br^- and SO_4^{2-} was enhanced from 26,263, 8498, 6.00 and 6562 to 2626.3, 859.8, 0.6 and 656 which represented 91%, 93%, 92% and 90%, respectively. The increases observed in the pH levels may be attributed to water electrolysis, which led to the production of hydrogen and (OH) ions. Afterward, pH was considered to be relatively stable, which could be attributed to the formation of the insoluble $\text{M}(\text{OH})_3$ flocs and metal hydroxide [20]. Higher efficiencies were recorded in the pH range of 5 to 9, which is close to the optimal pH for $\text{Al}(\text{OH})_{3(\text{s})}$ solid formation. Flocs of $\text{Al}(\text{OH})_{3(\text{s})}$ have large surface areas, which is useful for rapid adsorption of soluble organic compounds and trapping of colloidal particles. It has been established that at low pH, ranging from 5 to 6, cationic soluble species Al^{3+} and $\text{Al}(\text{OH})^{2+}$ are predominant [20]. When the pH is between 7 and 9, various aluminum-based monomeric and polymeric species are formed, which are finally transformed into insoluble amorphous $\text{Al}(\text{OH})_{3(\text{s})}$ through complex polymerization/precipitation kinetics [20,21]. When the pH is higher than 10, the soluble $\text{Al}(\text{OH})^{4-}$ anion concentration increases at the expense of $\text{Al}(\text{OH})_{3(\text{s})}$, according to the following reaction:



Furthermore, close examination of an activity-pH diagram for Al^{3+} species in equilibrium with solid phase $\text{Al}(\text{OH})_3$ —as shown in Figure 5—clearly indicates that the minimum solubility of aluminum hydroxide is recorded at pH 8 [21], which corresponds to the optimum pH value found in this study.

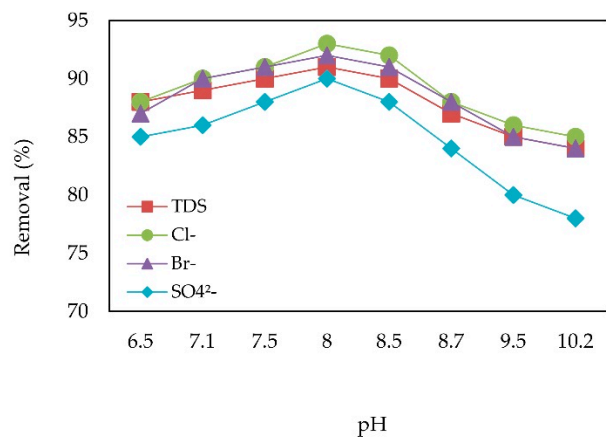


Figure 4. pH effect at optimal conditions (applied current density $[I] = 2.0 \text{ mA/cm}^2$, reaction time $[RT] = 80 \text{ min}$).

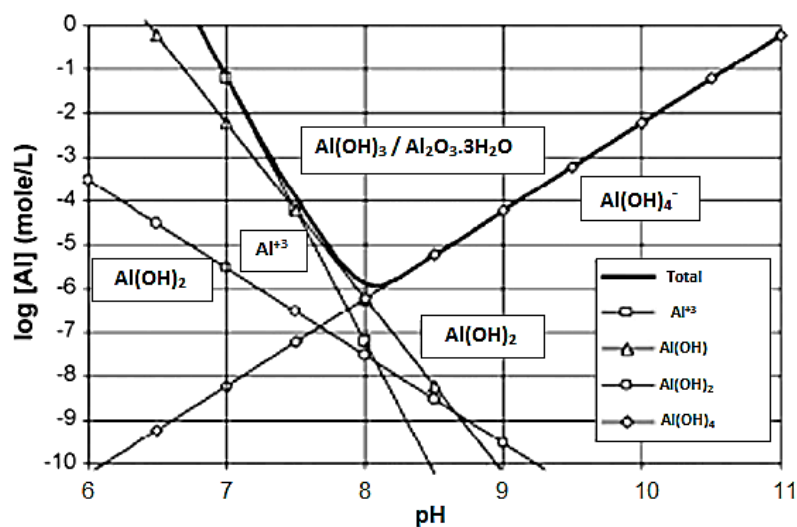


Figure 5. Aluminum hydrolysis solubility diagram.

3.3. Effect of Temperature

The temperature (t) of a solution can control the EC process during treatment [17–21]. An increase in temperature generates an increase in treatment efficiency due to the increased movement of ions. This movement promotes collision of ions with the coagulated material, thereby facilitating the removal process [60]. In order to investigate the varying effects of temperature on TDS, Cl^- , Br^- and SO_4^{2-} removal, experiments were carried out at temperatures between 25 and 45 °C, as shown in Figure 6. It has been observed that a rise in temperature causes an increase in the treatment process performance. This may be attributed to an increase in the mass transferred from the anode surface to the solution bulk in the form of Al^{3+} , and the increase of the rate of Al^{3+} hydrolyzed to $\text{Al}(\text{OH})_3$. This finding is in accordance with results of previous studies [61,62]. However, it was observed that increasing the temperature above 25 °C was uneconomic, as there was only a slight change in the rate of the reaction and the percentage of removal. As a result, a reaction temperature of 25 °C was selected as the optimal factor.

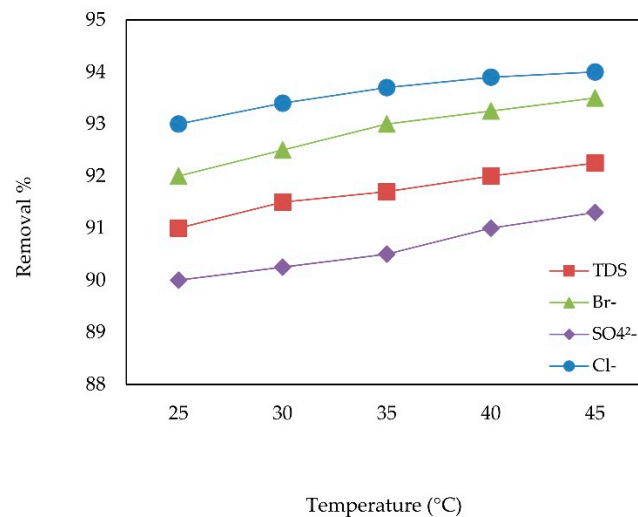


Figure 6. Temperature effect at optimal conditions ($I = 2.0 \text{ mA/cm}^2$, $RT = 80 \text{ min}$, $\text{pH} = 8$).

3.4. Effect of Inter-Electrode Distance

Assembly of the electrodes is crucial to both an effective electrode surface area and the inter-electrode distance (IED) [59]. An increase in the distance between the two electrodes leads to an increase in the resistance between the electrodes, requiring greater potential to overcome and therefore causing an increase in the treatment costs. However, an increase in distance between the electrodes theoretically leads to less interaction between the ions and hydroxide polymers [59]. Literature studies reinforced the importance of the effect of the IED on removal efficiency in the EC method [21,27,63]. The variation in voltage drop (ηIR) is governed by the following equation [21]:

$$\eta IR = I \cdot \frac{d}{S \times K} \quad (6)$$

where I represent the electric current, measured in amperes (A), d is the length of distance between the two electrodes in meters (m), S is the active anode surface area in square meters (m^2), and k is the specific conductivity in micro-siemens per meter ($\mu\text{S/m}$). This equation implies that the voltage drop will increase with an increase of inter-electrode distance (IED) at constant anodic surface area and conductivity of the solution.

In the present study, the effect of the IED was examined using 0.5, 1 and 2 cm by using Mp Al–Bp Al, $I = 2.0 \text{ mA/cm}^2$, $t = 80 \text{ min}$, and $\text{pH} = 8$, as the optimum conditions (Figure 7). The best performances were achieved with 1 cm as the middle distances. For a modification of the distance from 0.5 to 1 cm, the results were 90 to 91% for TDS, 92 to 93% for Cl^- , 90 to 92% for Br^- and 88 to 90% for SO_4^{2-} , respectively. On the other hand, an expansion of the distance from 1.5 to 2 cm led to a decline in the treatment removal rate, with a drop of 90 to 88% for TDS, 91 to 88% for Cl^- , 90 to 87% for Br^- and 87 to 82% for SO_4^{2-} , respectively. The influence of the IED on the performance of contaminants removal has been explained in previous studies [20,21], finding that an expansion of the inter-electrode distance (IED) leads to a decline of electrodes attraction. This was observed on the generated aluminum polymers, adversely affecting the EC process.

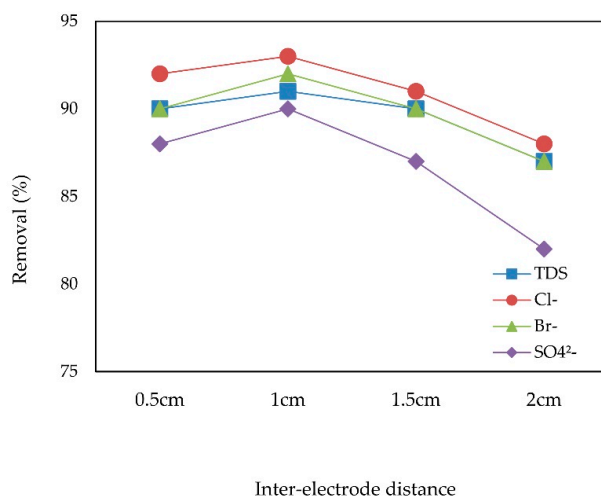


Figure 7. Effect of inter-electrode distance under optimal conditions ($I = 2.0 \text{ mA/cm}^2$, $RT = 80 \text{ min}$, $\text{pH} = 8$, $t = 25 \text{ }^\circ\text{C}$).

3.5. Effect of Stirring Speed

Stirring speed (S_s) is an influential factor in the EC process [20]. When the stirring rate is raised, pollution forms, combines and attaches together, and thus precipitation and mass transfer occur more easily. The reverse effect occurs when the stirring rate is increased more than needed, with the contaminants formed within the reactor getting degraded, creating smaller flocs that are difficult to separate from solution. This action causes a decrease in treatment efficiency [20,54,64]. In order to investigate the effect of the agitation speed on TDS, Cl^- , Br^- and SO_4^{2-} removal percentage in the EC process, stirring speeds of 250, 500 and 750 rpm were performed using the optimal conditions of: Mp Al-Bp Al, $I = 2.0 \text{ mA/cm}^2$, $t = 80 \text{ min}$, and $\text{pH} = 8$. The highest treatment efficiency was obtained when the speed was set to 500 rpm. The results show the TDS removal rate increased from 85 to 91%, Cl^- from 86 to 93%, Br^- from 85 to 92% and SO_4^{2-} from 80 to 90% for an increase in stirring speed of 250 to 500 rpm. Lower results were obtained when the agitation speed was raised from 500 to 750 rpm, where the TDS removal rates decreased to 87%, Cl^- to 87%, Br^- to 86% and SO_4^{2-} to 80%, as shown in Figure 8. These results can be interpreted by considering the fact that excessive stirring speed can lead to the breakup of flocs. Furthermore, the higher the stirring speed, the greater the energy the agitator consumed, and therefore the greater the operational costs. Consequently, 500 rpm was chosen as the best stirring speed.

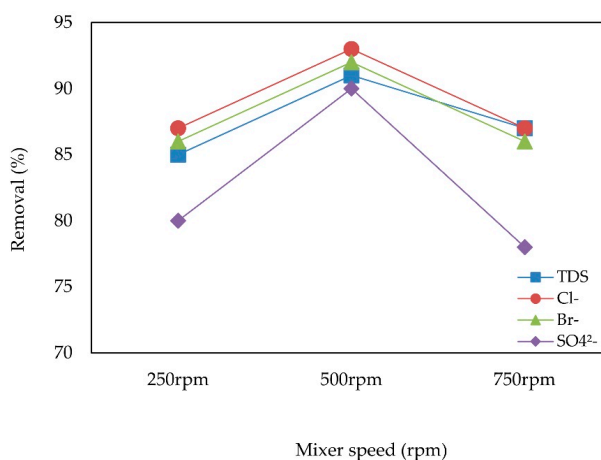


Figure 8. Effect of mixer speed under optimal conditions ($I = 2.0 \text{ mA/cm}^2$, $RT = 80 \text{ min}$, $\text{pH} = 8$, $t = 25 \text{ }^\circ\text{C}$, $\text{IED} = 1 \text{ cm}$).

4. Conclusions

Water treatment is essential in ensuring access to a clean water supply [65–72]. The present study evaluated the applicability of the EC method in the removal of salts (TDS, CL, Br and SO_4^{2-}) from aqueous environments using aluminum electrodes. Furthermore, the effects of current density (I), reaction time (RT), pH, temperature of the solution (T), distance between the electrodes (IED) and stirrer speed (rpm) on the removal of salts were investigated. The optimal conditions achieved were: $I = 2.0 \text{ mA/cm}^2$, $RT = 80 \text{ min}$, $\text{pH} = 8$, $T = 25 \text{ }^\circ\text{C}$, $IED = 1 \text{ cm}$ and $M_{\text{rpm}} = 500$. EC, under the combined electrical connection of aluminum electrodes, was shown to be very efficient for removal of salts from saline water. Using optimal conditions of the EC process, the removal percentages were 91%, 93%, 92% and 90% for TDS, Cl^- , Br^- and SO_4^{2-} , respectively. The findings of the present study reveal the technical feasibility of electrocoagulation as a reliable technique for removal of salts from aqueous environments.

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Conflicts of Interest: The authors declare no conflict of interest.

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