Removal of Boron from Simulated Iraqi Surface Water by Electrocoagulation Method

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Abstract

The removal of boron from aqueous solution was carried out by electrocoagulation (EC) using magnesium electrodes as anode and stainless steel electrodes as cathode. Several operating parameters on the removal efficiency of boron were investigated, such as initial pH, current density, initial boron ion concentration, NaCl concentration, spacing between electrodes, electrode material, and presence of carbonate concentration. The optimum removal efficiency of 91. 5 % was achieved at a current density of 3 mA/cm² and pH = 7 using (Mg/St. St.) electrodes, within 45 min of operating time. The concentration of NaCl was o. 1 g/l with a 0.5cm spacing between the electrodes.

First and second order rate equation were applied to study adsorption kinetics. The adsorption process follows second order kinetic model with good correlation. The energy consumption was evaluated for the optimum operating conditions. It was in the range of 1.296 to 1.944 Kwh/m^3. The overall expected increase in the operating cost of water treatment using membrane desalination facility (for example) will be about 20%.

Keywords: Boron removal, Electrocoagulation, Water treatment, Mg electrode, pH.

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الخلاصة

في هذا البحث استخدمة طريقة التخثير الكهربائي لمعالجة الماء الملوث بالبورون وباستخدام اقطاب انود من المغنسيوم كمخثر واقطاب كاثود من الحديد المقاوم . خلية تخثير بحجم 1.5 لتر قد استخدمت ايضا في العمل.

تم دراسة العديد من العوامل المؤثرة على عملية معالجة المياه الملوثة بالبورون وكانت هذه العوامل كالتالي: الدالة الحامضية، كثافة التيار، التركيز الاولي للبورون، تركيز كلوريد الصوديوم، المسافة بين الاقطاب، نوعية الاقطاب، وتركيز ايونات الكاربونيت. ومن النتائج تبين ان الازالة المثلي كانت 91.2% و قد تحققت بزمن 45 دقيقة بدالة حامضة تساوي 7 و كثافة تيار قدرها 3ملي امبير لكل سنتيميتر مربع وباستخدام اقطاب المغنسيوم. وبتركيز كلوريد صوديوم يساوي 0.1 ملغرام / لتر ومسافة 0.5 سنتيميتر بين الاقطاب. كما وقد تم دراسة كلفة استهلاك الطاقة في الضروف المثلى للازالة ، وقد كان استهلاك الطاقة يتراوح بين 1.296 الى 1.944 كيلو واط .ساعة/ متر مكعب

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1. Introduction

Recently boron has come to the forefront as a possible drinking water contaminant (WHO, 2008). Due to the increasing demand of water in both potable use and for Irrigation, coupled with a decrease of suitable water sources, suppliers have to use alternatives such as sea water. By using those alternative sources more trace contaminants start to appear in the final product. One of these is boron. Boron (B), an inorganic compound, is a non-volatile metalloid that is ubiquitous in the environment in compounds called borates. Common borates include boron oxide, boric acid, and borax (USEPA, 2008). Boron could be released into the environment by both natural weathering processes and anthropic sources (Yonglan et al, 2008)**.**

Concentrations of boron vary widely and depend on the surrounding geology and wastewater discharges. Boron compounds are used in the manufacture of a variety of industrial products such as glass and porcelain, wire drawing, leather production, carpets, cosmetics, detergents, food preservatives, photographic chemicals, fireproofing fabrics and weather proofing for woods (Nihal et al, 2004)**.** During these processes many boron compounds are introduced into the environment as a waste.

Short and long term oral exposures to boric acid or borax in laboratory animals have demonstrated that the male reproductive tract is a consistent target of toxicity (WHO, 2008). According to the WHO and Iraqi drinking water guideline for 2009, boron levels in drinking water have to be limited to 0.5 mg/l.

There is no easy or simple method for the removal of boron from water and wastewater (Kabay, 2010). Technologies for boron removal include conventional and advanced treatment technologies such as, chemical precipitation, ion exchange, reverse osmosis (RO), adsorption with Activated carbon (AC), Electrodialysis (ED) and etc. These methods have disadvantages such as using large quantities of chemicals and generating large volume of sludge causing disposal problems. Therefore there is a need to find more efficient method to remove boron from water.

In recent years, the level of fresh water flowing into Shatt Al-Arab from Euphrates and Tigris rivers has declined, so salt and boron containing-water from the Arabian Gulf has increasingly poured in. This resulted in a problem to the water treatment plants in the south of Iraq because they use conventional methods for water treatment which are not effective for boron reduction as previously mentioned.

Researches had been done to remove boron from water and wastewater using EC method with different types of electrodes materials. Nihal et al., 2004 investigated the feasibility of boron removal from wastewater using EC with aluminum electrodes. The process was examined under various factors in order to assess optimal operating conditions. They showed that boron removal by EC process depends on current density, initial concentration and time. Removal of 92-96% was achieved under a current density of 30 mA/cm². Erden et al., 2007 provided a quantitative comparison of EC and Chemical Coagulation (CC) approaches based on boron removal. Comparison was done with the same amount of coagulant between EC and CC processes. Although, all operational parameters were same in EC and CC, boron removal efficiency between each treatment processes was a quite different. Boron removal efficiency for EC and CC were 94% and 24%, respectively. Subramanyan et al., 2010 optimized the process parameters for the removal of boron from drinking water by EC. The results showed that a maximum removal efficiency of 97.3% was achieved at a current density of 50 A/m² and pH of 7 using magnesium at the anode and stainless steel as the cathode.

This work aims to evaluate the effectiveness of EC process to remove boron from water using Magnesium/ St.St. electrodes and determine the effects of various operating parameters on the removal efficiency such as initial boron concentration, initial pH, current density, gap

between electrodes, sodium chloride concentration, and time.

2. Electrocoagulation Mechanism

EC is a complicated process involving many chemical and physical phenomena that use sacrificial electrodes, such as Al, Fe and others, to supply ions into the water (Christos et al, 2010). In the EC process the coagulants generates in situ by dissolving electrically the consumable electrodes (Fe/Al). The metal ions generation takes place at the anode; hydrogen gas is released from the cathode (Lawrence, et al, 2007). In the EC process current is passed through a metal electrode, oxidizing the metal (M) to its (M^{n+}) at the anode.

$$M \longrightarrow M^{n+} + ne^- \qquad (1)$$
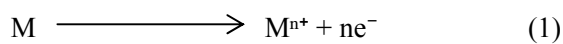
At the cathode side, hydrogen gas (H_2) and the

hydroxyl ion (OH^-) are generated by reducing

the water.

$$2H_2O + 2e^- \longrightarrow 2\,OH^- + H_2(g) \qquad (2)$$
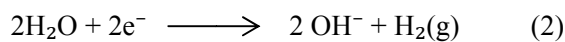
(Holt et al, 1999).

In the solution, the metal cations resulted from the anode oxidation combine with

hydroxyl ion (OH^-) resulting from water to form

highly charged coagulant. In the case of Aluminum anode, the Al^{3+} reacts with H_2O to form $Al(OH)_3$, and in the case of magnesium anode the Mg^{2+} reacts with H_2O to form $Mg(OH)_2$.

The destabilization mechanism of the contaminant, particulate suspension, and breaking of emulsion may be summarized as follows (Christos et al, 2010).

1- Compression of the diffuse double layer around the charged species by the interactions of ions generated by oxidation of the sacrificial anode.
2- Charge neutralization of the ionic species in the wastewater by counter ions produced by the electrochemical dissolution of the sacrificial anode.
3- Floc formation.

3. Experimental Apparatus & Procedures

Fig 1 shows the EC cell consisted of a 1.5-L Plexiglas vessel. Magnesium electrodes of surface area (240 cm²) acted as the anode. The cathodes were stainless steel of the same size as the anode. A regulated direct current (DC) was supplied from a rectifier (5 A, 0–25 V).
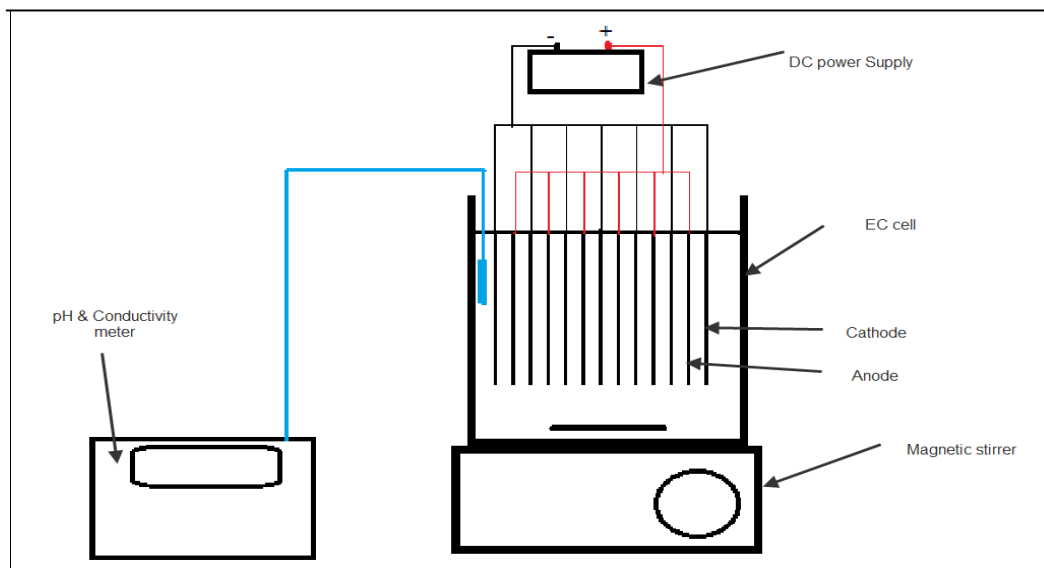
Figure 1: Schematic diagram of EC cell.

Boric acid H_3BO_3 was dissolved in distilled water for the required concentration. The solution of 1.5 L was used for each experiment as the electrolyte. The pH of the electrolyte was adjusted, if required, with 1 mol/L HCl or NaOH solutions before adsorption experiments. Electrolytic cell with monopolar electrodes in parallel connections has been used with an interelectrode distance varying from 0. 5 cm to 1 cm depending on each experiment conditions.

The magnesium electrode was weighed before and after each experiment to determine the amount of magnesium loss of each experiment. Before each experiment, the conductivity of the solution was adjusted with NaCl. During the experiments, the solution in the EC unit was stirred at 120 rpm by a magnetic stirrer with an Initial temperature of 22 ℃.

Each run was timed starting with the DC power supply switching on. During the experiments, anodic dissolution occurred and hydrogen gas was produced at the cathode. Samples of 10 ml were taken during each experiment at different time to be filtered and then analyzed using atomic absorption spectrometer. Samples were analyzed at the civil Engineering labs at Dalhousie University in Canada. Each sample was taken from a distance between two electrodes and at fixed depth. After each run, the electrodes were washed and brushed then cleaned by ethanol to remove any solids accumulated on the electrode surface.

The operating conditions for each test run are summarized in Table 1. These operating conditions were optimized from preliminary tests and found out that they are the best conditions that give acceptable removal efficiency.

Table 1: Test runs plan summary for operating parameters.

Test run number	Test run description	pH	Boron conc. (mg/l)	Current density (mA/cm²)	NaCl conc. (g/l)	Spacing between electrodes cm	Carbonate conc. (mg/l)	Time (min)
1	Eff. of pH	3-10	5	3	0.1	0.5	0	0-120
2	Eff. of current	7	5	1-4	0.1	0.5	0	0-90
3	Eff. of B conc.	7	3-7	3	0.1	0.5	0	0-90
4	Eff. of NaCl conc.	7	5	3	0.01-0.2	0.5	0	0-90
5	Eff. of spacing	7	5	3	0.1	0.5-1	0	0-90
6	Eff. of carbonate	7	5	3	0.1	0.5	50-250	0-90

From other related research, it has concluded that the best stirring speeds are from 100 – 150. So, stirring speed of 120 rpm has kept constant during all the experiments.

4. Results and Discussion

4.1 Effect of Initial pH

pH of the solution plays an important role in the electrochemical and chemical coagulation process (Christos et al, 2010)**.** In this study a set

of experiments were performed with initial pH in the range of (3-9), the test operating conditions are shown in Table 1. The maximum boron removal efficiency (99.59%) was obtained at pH of 7 within 120 min, which is neutral, from this it can be concluded that the majority of Mg coagulants are formed at this pH. The major

complex that is formed at this pH maybe due to formation of $Mg(OH)_2$ which could be less soluble in water at pH of 7. However, that could be the reason of high boron removal percentage at this pH. This result is in agreement with the work of (Vasudevan et al, 2010). In which he indicated that the highest boron removal was obtained at pH of 7 because boron was present as $B(OH)_3$ and the formation of $Mg(OH)_2$ was high.

The results that show the effect of different pH values on removal efficiency of boron are shown in Fig 2
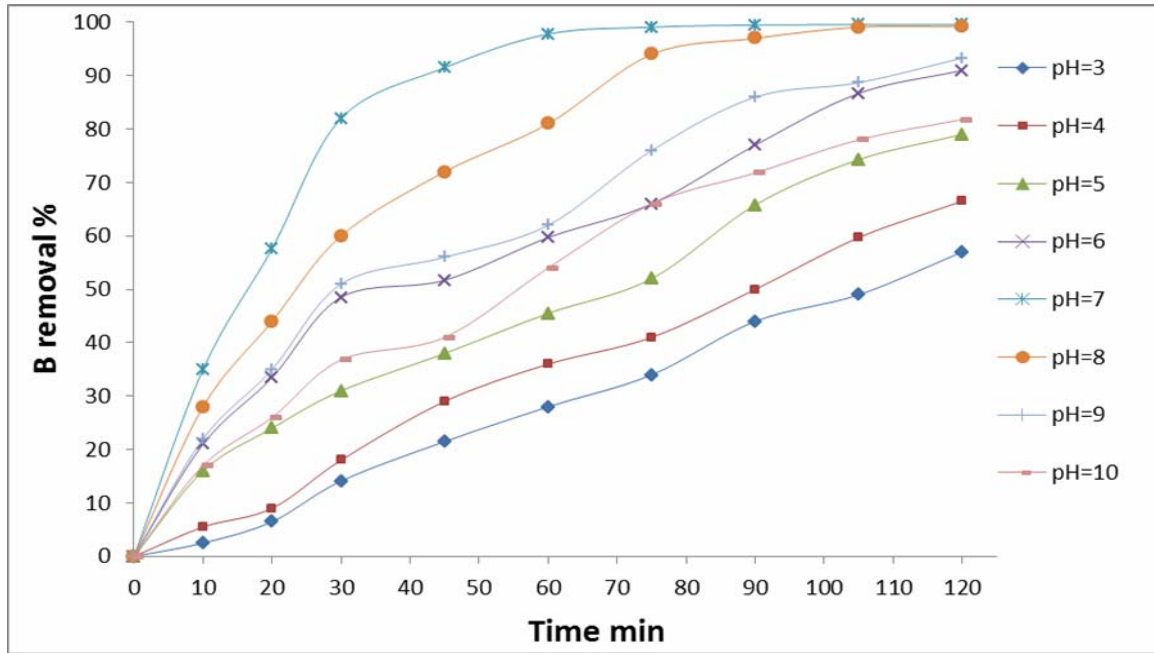
Figure 2: Boron removal efficiency with different pH values (Current density = 3mA/cm², [B]= 5 mg/l, [NaCl] = 0.1 g/l, gap = 0.5 cm, T = 22 °C, and stirring speed = 120 rpm).

4.2 Effect of Current Density

Current density is very important parameter that affects the EC process because it directly determines both coagulant dosage and bubble generation rates and strongly influences both solution mixing and mass transfer at the electrodes. So current density is the key operational parameter that affecting the system's response time and also influencing the dominant pollutant separation mode (Christos et al, 2010)**.**

To examine the effect of current density on boron removal efficiency, a series of experiments were carried out with the current density being varied from 1 – 4 mA/cm². The test operating conditions are shown in Table 1. The results are depicted in Figure 3.

From the Figure it can be seen that the boron removal efficiency increases with the increase of current density. At high current densities, the generation of Mg ions increases due to the increase of anodic dissolution, resulting in a greater boron removal rates, indicating that the

Adsorption depends up on the availability of binding sites for boron.

From Fig 3, it can be indicated that a current of 3mA/cm² gives B removal efficiency of 94%

which effectively reduces boron concentration to less than 0.5 mg/l. Any other increase in current density leads to increase in removal efficiency but it is not economically efficient due to high power consumption. A comparison between experimental and theoretical sacrificial magnesium anode consumption has been achieved by calculating the theoretical Mg loss from the anode using Faraday's equation and compared with those obtained experimentally. (Christos et al, 2010).

$$w = \frac{itM}{Nf},$$ Faraday's law   (3)

Where:- w is the quantity of electrode material dissolved (g of M/cm²), i is the current density (A/cm²), t is the time in seconds, M is the relative molar mass of the electrode concerned, (M =56 for Fe, 27 for

Al, and 26. 3 for Mg), N is the number of electrons in oxidation/reduction reaction, ($N=2$ for Fe and Mg , $N= 3$ for Al), And f is the Faraday's constant, 96, 500 C/mol.

The comparison between theoretical and experimental amount of Mg dissolved in the electrolysis is shown in Figure 4.
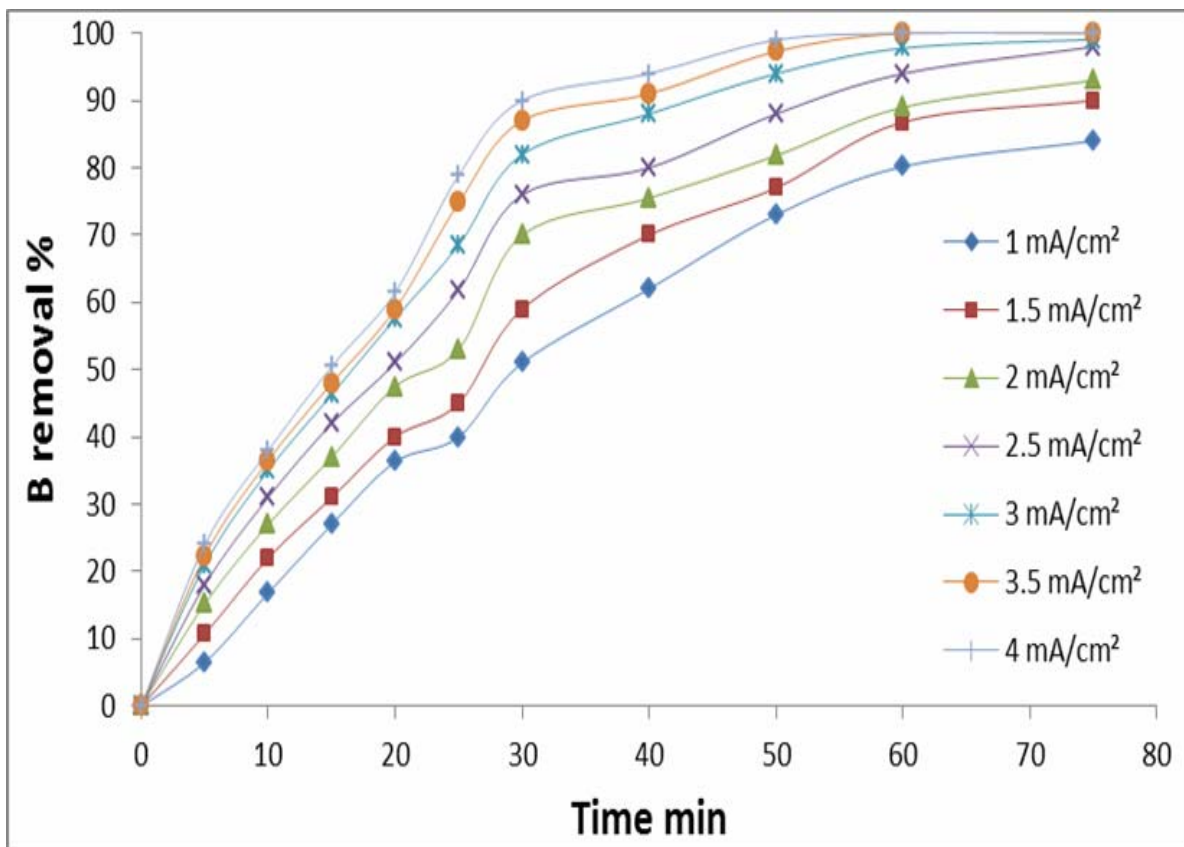

Figure 3: Effect of current density on boron removal efficiency. (pH = 7, [B]= 5 mg/l, [NaCl] = 0. 1 g/l, gap = 0. 5 cm, T = 22 °C, and stirring speed = 120 rpm).
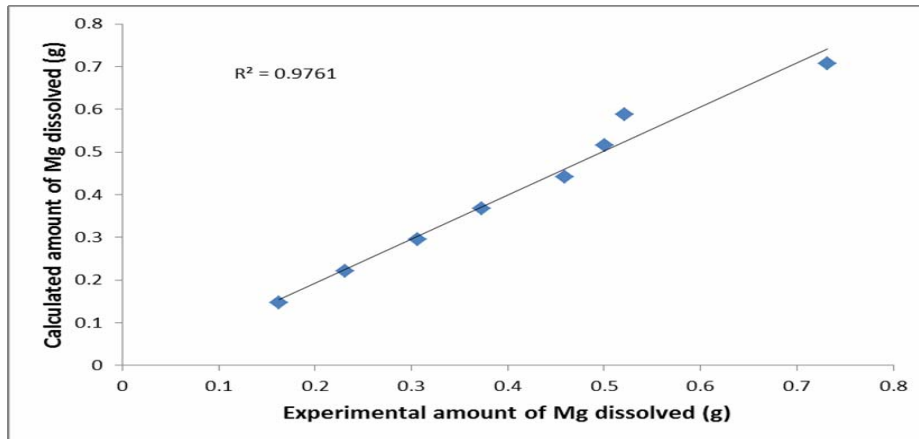
Figure 4: Measured Vs. calculated amount of Mg released from the anode

4.3 Effect of Initial Boron Concentration

The effect of initial boron concentration on boron removal was examined with solutions of boron concentrations ranging from 3-7 mg/l. The other test operating conditions are shown in Table 1. Fig 5 shows the boron removal efficiency with time for different initial boron Concentrations. From the results it can be seen That boron removal efficiency decreased with increasing boron concentration. This can be explained as follows; although the same amount Mg^{2+} passed to solution at the same current density for all boron concentration, Mg^{2+} was insufficient for solutions of higher B concentration.
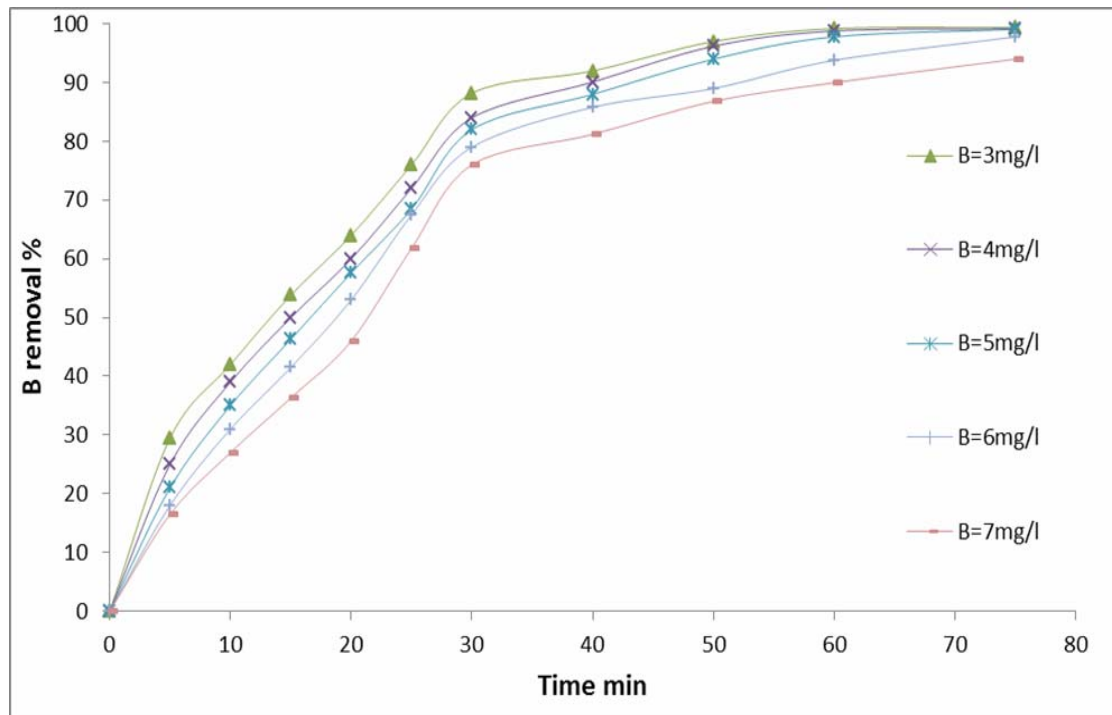
Figure 5: The Effect of initial boron concentration on Boron removal efficiency. (pH = 7, current = 3mA/cm², [NaCl] = 0. 1g/l, gap = 0. 5cm, T = 22 °C, and stirring speed = 120rpm).

4.4 Effect of Sodium Chloride Concentration

The effect of NaCl concentration on boron removal was examined with NaCl concentrations ranged between 0. 01 – 0. 2 g/l. the other test operating conditions is shown in Table 1. As expected, the obtained results showed that boron removal increased with increasing concentration of sodium chloride because of the increase in solution conductivity.

In addition, the energy consumption decreased with increasing concentration of sodium chloride due to potential decrease under constant current density. The results obtained from the experimental work are shown graphically in Fig 6.
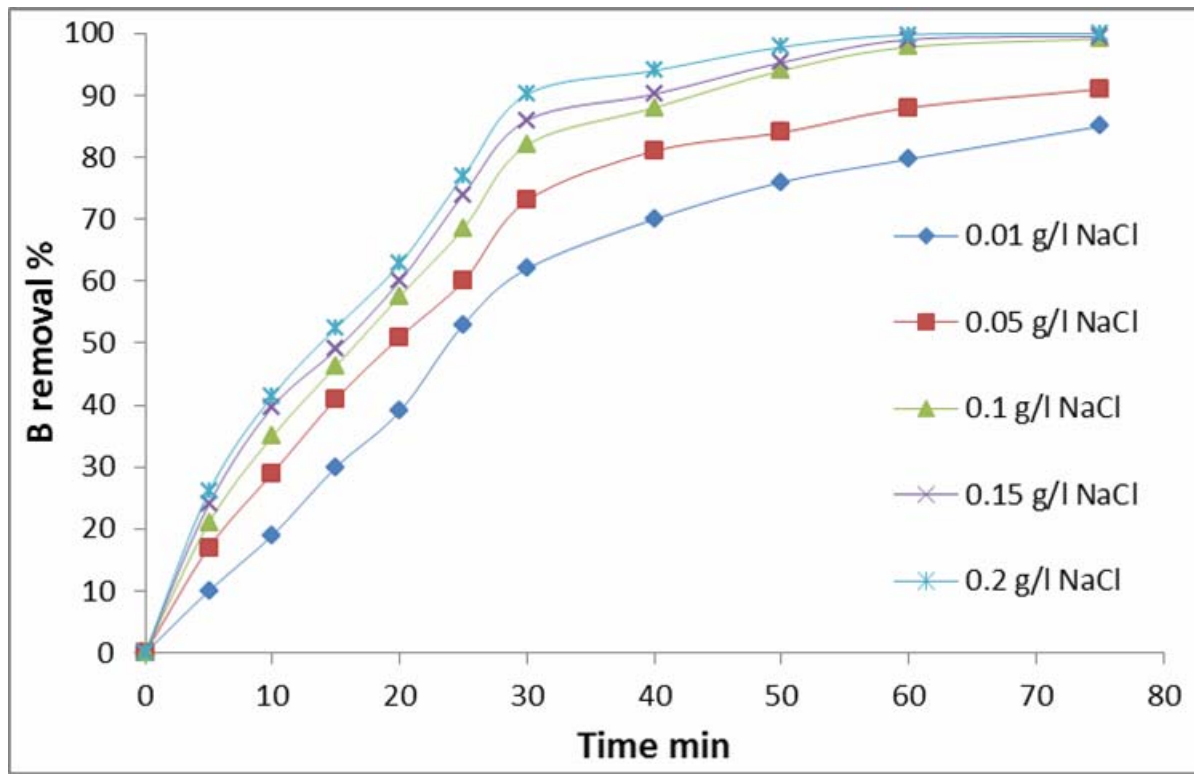
Figure 6: Relationship between B removal efficiency and time with different NaCl concentrations. (pH = 7, current = 3 mA/cm², [B] = 5 mg/l, gap = 0.5 cm, T = 22 °C, and stirring speed = 120 rpm).

4.5 Effect of Spacing between Electrodes

The effect interelectrode spacing on boron removal was investigated with spaces of 0.5, 0.75, and 1cm. The other operating conditions are shown in Table 1. It can be seen from Figure 7 that the removal efficiency increases with the decrease of space between electrodes while the highest removal efficiency of 99.1% was obtained at gap of 0.5 cm after 75 min. This can be explained that decreasing the space between electrodes results in low resistance through the solution which in effect results in increasing the rate of magnesium dissolution and

Mg²⁺ releases and consequently leads to more boron removal from the solution. On the other hand, decreasing the space could enhance the flotation process by limiting the generated bubbles in a narrow space which results in higher removal efficiencies.
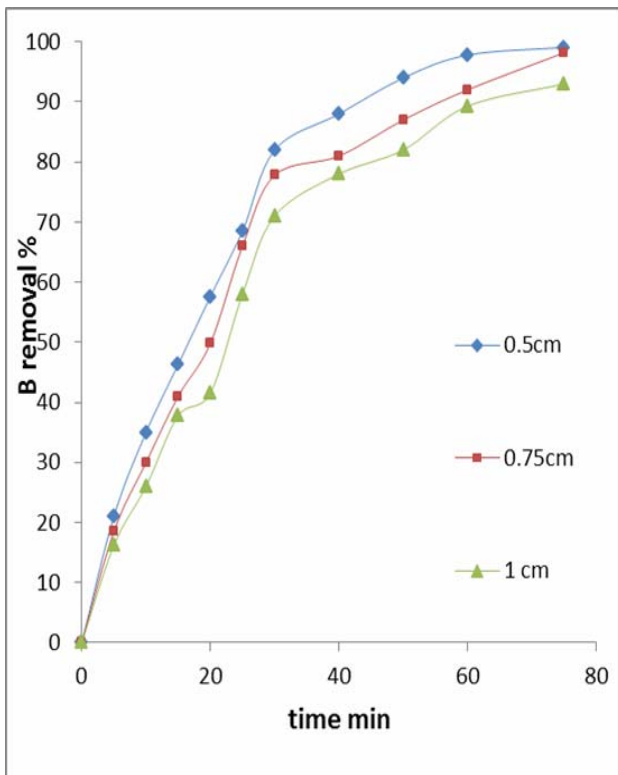
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Figure 7: Effect of interelectrode spacing on boron removal efficiency

4.6 Effect of Electrode Material

A set of experiments were carried out using aluminum electrodes with the same dimensions and active area of the magnesium electrodes to

investigate the effect of different electrode material on boron removal efficiency. The operating conditions were maintained the same

As when magnesium electrodes were used in the previous experiments. The operating conditions were: current density of 3 mA/cm², pH = 7, boron concentration of 5 mg/l, NaCl concentration of 100 mg/l, and interelectrode spacing of 0.5 cm.

Figure 8 shows that using Al electrodes is more efficient than Mg electrodes for the same operating conditions, although both electrodes reach the same maximum efficiency within 80 min. This could be happened because that the positive charge of Al ions (Al³⁺) is higher than the charge of Mg ions (Mg²⁺) which leads to more sorption affinity on pollutant colloids for Al ions.

The reason of using Mg electrodes instead of Al electrodes in the boron removal from drinking water is the high residual of aluminum in treated water, which can create health problems. In the case of Mg electrodes, there is no such disadvantage. The maximum allowed concentration of Al in drinking water is 0.2 mg/l while it is 100 mg/l for Mg **(**Iraqi specifications for drinking water, 2009)**.**

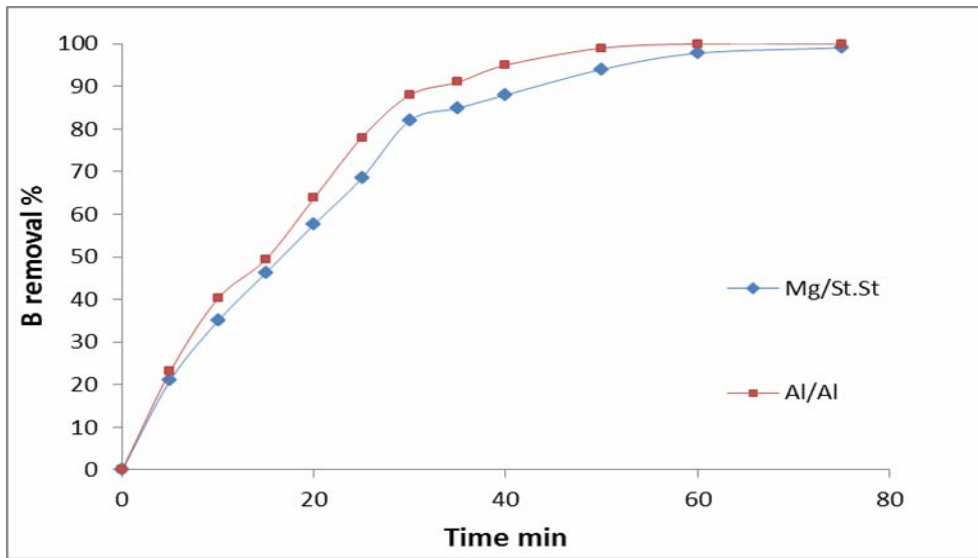Figure 8: Effect of electrode material on boron removal efficiency

4.7 Effect of Coexisting Anions (Carbonate).

To investigate carbonate anions on boron removal a set of experimental series were conducted with different carbonate concentrations of 50, 100, 150, 200, and 250 mg/l. the other operating conditions are shown in Table 1. Figure 9 shows the effect of different carbonate concentrations on boron removal efficiency. The results indicate that carbonate anions have a significant effect on boron removal efficiency. This could be due to passivation of the anode and reducing dissolution of the anode.
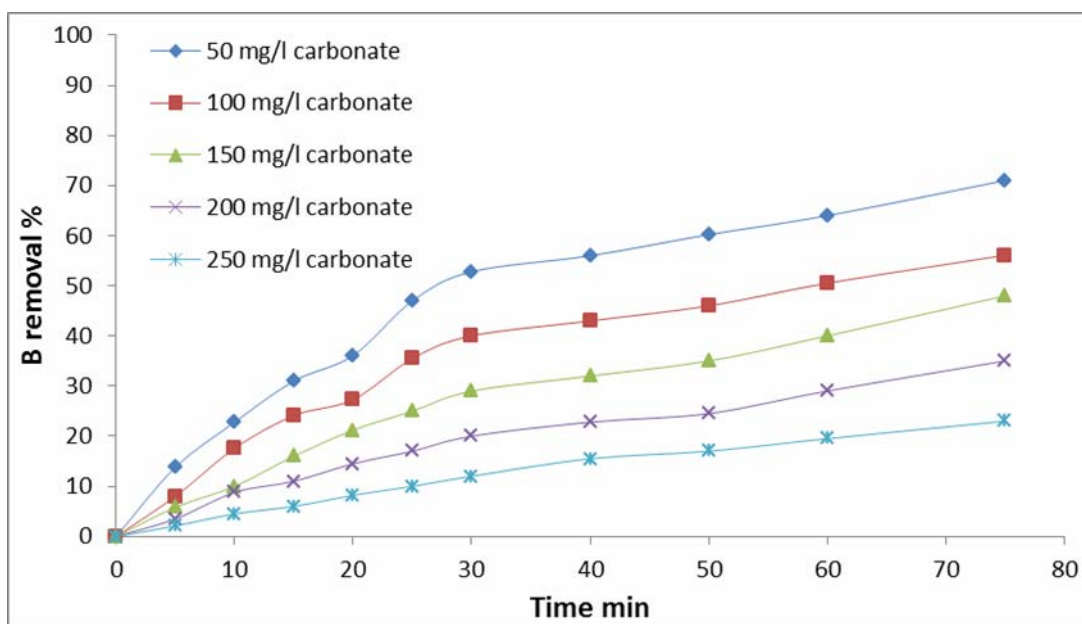

Figure 9: Effect of different carbonate concentration on B removal efficiency

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4.8 Studies of Adsorption Kinetics

The kinetics of adsorption of boron is analyzed using first-order and second-order Lagergen rate equations. (Azizian, 2004)**.**
The pseudo-first-order rate equation is

$$\frac{dq}{dt} = k_1 \left(q_e - q \right) \tag{4}$$

Integrating Eq. (4) for the boundary conditions $t = 0$ to $t = t$ and $q = 0$ to $q = q$ gives

$$ln \frac{(q_e - q)}{q_e} = -k_1 t \tag{5}$$

Where: q and qe are the grams of solute sorbed per gram of sorbent at any time and at

equilibrium, respectively, and k_1 is the rate

constant of first-order sorption.
Another model for the analysis of sorption kinetics is pseudo-second-order. The rate law for this system is expressed as

$$\frac{dq}{dt} = k_2 \left(q_e - q \right)^2 \tag{6}$$

Integrating Eq. (6), for the boundary conditions $t = 0$ to $t = t$ and $q = 0$ to $q = q$, gives

$$\frac{1}{(q_e - q)} = \frac{1}{q_e} + k_2 t \tag{7}$$

Where: k_2 is the pseudo-second-order rate constant of sorption.

Equation (7), can be rearranged to obtain a linear form,

$$\frac{t}{q} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{8}$$

The amount of boron adsorbed at equilibrium and the first-order rate constant (k_1) were calculated from the slope of the plots of $ln \frac{qe - q}{qe}$ versus time (t). It was found that the calculated qe values do not agree with the experimental qe values.
To investigate the adsorption of boron using Lagergen second-order equation, t/qt versus time were plotted in Figure 10. The second order kinetic values of qe and k_2 were calculated from the slope and intercept of the plots t/qt vs. t.
Table 2 gives the computed results from first- and second-order kinetic models. For the second-order kinetics model the calculated experimental amount of boron adsorbed at equilibrium values agree better than those of the first order kinetic model, indicating that adsorption follows more closely the second-order kinetic model.
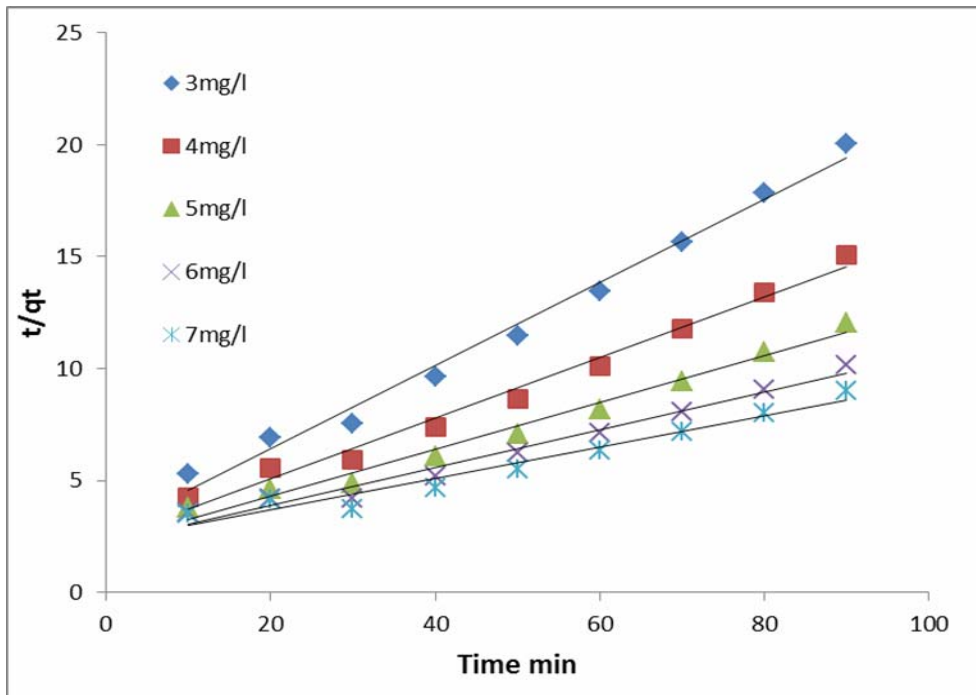
Figure 10: Second order kinetic model plot for different concentration of boron.

Table 2: Comparison between experimental and calculated qe values for different initial boron concentrations in first and second-order adsorption isotherm.

B conc mg/l	qe expt.	first order adsorption			second order adsorption		
		qe calc	K1 1/min	R²	qe calc	k2 * 10⁻ 1/min	R²
3	4.4955	7.7428	0.0859	0.9873	5.379	12.85	0.9884
4	5.976	21.8	0.1064	0.9171	7.3964	7.693	0.9857
5	7.4625	16.1213	0.08198	0.9768	9.5877	4.907	0.9822
6	8.874	15.8635	0.06747	0.9391	11.8906	3.1894	0.9771
7	9.9855	18.16	0.06632	0.9404	14.2247	2.162	0.9561

4. 9 Study of Cost of EC Process

The energy consumption was evaluated for the optimum operating conditions that obtained from the tests shown before. Ec (Kwh/m³) is calculated from the following equation. (Al Gurany, 2009)**.**

$$Ec = \eta IR I \, t / 1000V \qquad (9)$$

Where, ηIR is the potential drop across the solution (V), I is the electrolysis current (A), t is the electrolysis time (h), V is the volume of the solution treated (m³).

For current consumption of 3 mA/cm² (0.72 A in this thesis), voltage of 5.4 V, cell volume of 1.5 l, and 45 min operating time, the Ec is 1.944 Kwh/m³. If electric cost is estimated to be 0.06 $/kWh, then each m³ will cost extra 11 cents for only the electric consumption. There are also other costs, such as sludge movement, magnesium material costs, maintenance, etc.

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The overall expected increase in the operating cost of water treatment using membrane desalination facility (for example) will be about 20%. Water samples were taken from two water treatment plants at Shat Al-Arab then these samples were tested. The results showed that the treated water comes from the treatment plants has high salinity reaches up to thousands of mg/l. Although water salinity is not preferred in drinking water, it enhances the EC process by increasing the solution conductivity which leads to less treatment cost by reducing the operating time. Fig 11 shows how more salty water gives higher removal efficiencies.
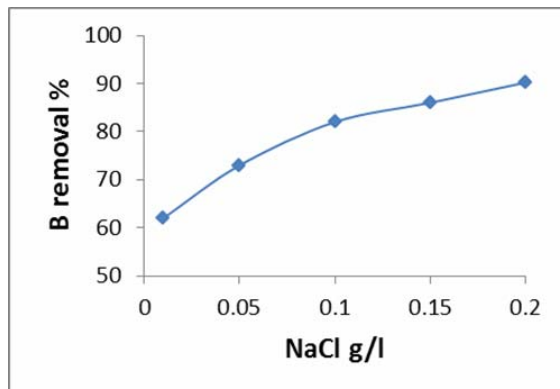

Figure 11: The effect of NaCl concentration of B removal efficiency (30min)

This Figure shows that using a concentration of 200 mg/l of NaCl instead of 100 mg/l gives 90.2% removal efficiency at 30 min. This removal efficiency reduces B level from 5 mg/l to 0.49 mg/l, which is within the drinking water guidelines. So that, using 200 mg/l NaCl reduces the cost of EC process by time reduction. Operating time of 30 min gives Ec equal to 1.296 Kwh/m³. In this case the EC cost will be 7.7 cents instead of 11 cents for only the electric consumption.

5. Conclusions

Experiments have been carried out to determine the best operating conditions which give an acceptable boron removal with the lowest time and cost.

1- The results showed that the optimized boron removal efficiency of 91.5% was achieved at a current of 3mA/cm², pH of 7, NaCl = 0. 1g/l, time = 45min, electrode spacing = 0.5cm, at a stirring speed of 120rpm, and temperature equal to 22°C, using magnesium electrodes as anode and stainless steel electrodes as cathode.

2- Although, aluminum electrodes gave more boron removal efficiency than magnesium electrodes, the use of magnesium electrodes in drinking water treatment is preferred because of the high Al ions residual in the treated water which may lead to some health problems.

3- It was clearly indicated that the boron removal efficiency of 91.5% was achieved from the generation of magnesium hydroxide in the EC cell. The magnesium hydroxide removes boron presents in the water and reduces the boron concentration to less than 0.5 mg/l, and makes it within the acceptable range for drinking water as mentioned by WHO guidelines for boron.

4- The effect of carbonate anions concentration on boron removal also was studied and the result showed a significant effect on the removal efficiency when the carbonate concentration is high, which could be caused by the passivation of the anode causing reduction in anode dissolution.

5- From an experimental study of boron adsorption kinetics, it was found that the boron adsorption process follows Lagerange second-order kinetics rather than first-order kinetics.

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