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Boron Removal from Aqueous Solutions by Electrocoagulation at Low Concentrations

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Highlights

- The capability of electrocoagulation was determined in boron removal from aqueous solutions at low concentrations.
- Boron adsorption on produced aluminum corresponds to isotherms of Langmuir and Freundlich and pseudo-second-order model.
- Interfering effect of carbonate ions in boron removal is more than other common anions in water.
- Electrocoagulation can be used for treatment of boron-polluted water at low initial concentrations.

Abstract

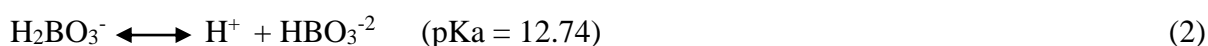
Boron can cause gastrointestinal dysfunction, blood circulation disorder, and reproductive problems. In this research, the effect of some parameters on the efficiency of treatment system is studied in detail. These parameters include pH, distance between electrodes, current density, reaction time, initial concentration of boron, and interference effect of common anions in water including carbonate, sulfate, and chloride. The boron concentration in the samples is determined by standard method. The efficiency of electrocoagulation for boron removal at a concentration of 100 mg/L and under optimal condition (pH: 8, distance between electrodes: 10 mm, reaction time: 60 min, concentration of 100 mg/L and current density: 5.5 mA/Cm²) is good (about 70%). Boron adsorption isotherm on produced aluminum according to Faraday's law corresponds to isotherms of Langmuir ($R^2 = 0.77$) and Freundlich ($R^2 = 0.79$). Boron adsorption kinetics on produced aluminum corresponds to the pseudo-second-order model ($R^2 = 0.99$). Interfering effect of carbonate ions is more than other anions in water such as chloride and sulfate. The addition of two moles of carbonate ions can reduce the efficiency of the process by 15%. Therefore, unlike the chemical coagulation process, using pre-treatment processes for the initial adjustment of pH in water using limewater due to an increase of carbonate in water can decrease the efficiency of electrocoagulation. Electrocoagulation process can be used for treatment of boron-polluted water and wastewaters containing boron at initial concentrations of more than 50 mg/L.

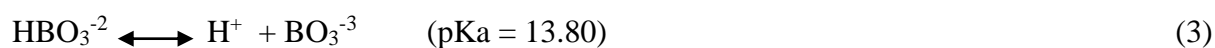
Keywords: Electrocoagulation; Boron; Aqueous Solutions, Aluminum Electrode

Introduction

Boron is an element that in nature, when combined with oxygen and other natural elements, appears in the form of borates and boric acid. This material is mostly used in glass industry, detergents, fire extinguishers, and nuclear industry (Bektaş et al., 2004; Halim et al., 2012a). Although the maximum solubility of boron in water is determined up to 47.2 g/L, its average concentration in surface water resources is 4.5 mg/L, an amount that, according to seasonal effects, is variable up to 7 mg/L (Kluczka et al., 2007). This element enters the environment through the activities of mineral extraction, coal incineration, discharge of wastewater containing detergents produced by borax, boron fertilizers and insecticides, and finally plant and wood incinerators (Demirçivi and Nasun-Saygili, 2008).

The long-term use of water containing boron can cause gastrointestinal dysfunction, blood circulation disorder, and reproductive problems including giving birth to deformed babies (Kloppmann et al., 2005; Kluczka et al., 2007). Furthermore, boron is effective in embryo development in rats and also can bring about change in their sexual organs, ultimately leading to their sterility. Having more than 3 mg/L of boron in agricultural water can cause severe damage to plants and trees, especially to citrus. Leaf blight, stem decay, and abnormality in yield of fruit trees are among the problems of high rate of boron in agricultural water (Halim et al., 2012a; Halim et al., 2012b; Kluczka et al., 2007; Xu and Jiang, 2008). WHO has announced that the maximum allowable concentration of boron in drinking water is 2.4 mg/L; this is while European Union has determined it to be 1 mg/L (Demirçivi and Nasun-Saygili, 2008; Halim et al., 2012a; Isa et al., 2014). Water-soluble boron is available in the form of boric acid (H_3BO_3), borates, and anionic polyborates including $[\text{B}_3\text{O}_3(\text{OH})_4]^-$, $[\text{B}_4\text{O}_5(\text{OH})_4]^{2-}$, $[\text{B}_5\text{O}_6(\text{OH})_4]^-$. Water-soluble boric acid (H_3BO_3) is a little bit acidic and acts as an electron acceptor agent. At low pHs, boron usually appears in the form of molecular boric acid in water (1) whose ionic decomposition reactions have been shown in reactions 1 to 4.





After the reaction with water, boric acid is converted to anion $[\text{B}(\text{OH})_4]^-$:



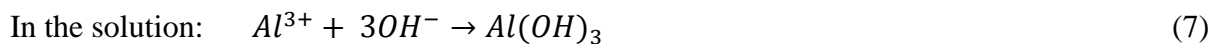
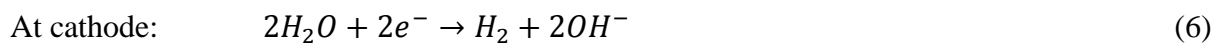
The equilibrium constant for reaction (Demirçivi and Nasun-Saygili, 2008) is not sufficient and the ratio of boric acid at near-neutral pHs in dilute solutions is more than 99 percent. A rise in pH increases the concentration of borates or anion $[\text{B}(\text{OH})_4]^-$ so that it takes the dominant form at pH = 9. Therefore, common treatment methods including coagulation, filtration, precipitation, and common ion exchange especially at low concentrations of boron, cannot remove it properly from water (Organization, 2009; Sasaki et al., 2016; Vasudevan and Lakshmi, 2012). Important processes studied in the field of boron removal from water include reverse osmosis, selective ion exchange, adsorption on zero-valent iron nanoparticles and electrocoagulation (Halim et al., 2012a; Khorsandi et al., 2017). Due to the higher level of boric acid in comparison with the other borates in aquatic environments and because of an ionic dissociation constant of ($\text{pK}_a=9.27$), the efficiency of these processes is low (Bektaş et al., 2004; Khaoula et al., 2013a; Xu and Jiang, 2008).

The process of reverse osmosis is known as an expensive technique with high operating costs and in ordinary conditions, it cannot remove much boron from water. According to the previous studies, the efficiency of this process in boron removal from water in ordinary conditions is about 40 to 60 percent (Xu and Jiang, 2008). Ion exchange process for boron removal from water has been studied using selective exchangers such as Amberlite and IRA743. The high costs of establishment and operation of selective ion exchanger units have questioned its use in practical scale (Simonnot et al., 2000; Vasudevan and Lakshmi, 2012; Xu and Jiang, 2008).

The process of chemical coagulation is also studied for boron removal from water at concentrations of more than 100 mg/L and at pHs of more than 10. The efficiency of this process is reduced by the reduction of boron concentration and initial pH of water. The most important reason for this issue is the dominance of the concentration of boric acid over the other borates at low concentrations and pHs. As according to the studies by Shih et al., the process of chemical precipitation can remove more than 80 percent of borates whereas the removal by boric acid is about 5 percent (Shih et al., 2014).

Therefore, due to the high solubility of boron at common pHs of water, its removal by the processes of reverse osmosis, selective ionic exchanger, and chemical coagulation is quite difficult and it is necessary to increase the pH of water up to more than 10 before the treatment. After the treatment, it also needs to be decreased to the acceptable range of 6.5 to 8.5 (Cengeloglu et al., 2008; Xu and Jiang, 2008). Thus, the establishment and operation costs of refinery are hugely increased.

Unlike the other processes, electrocoagulation is able to remove boric acid directly, possibly because of the production of hydroxide and hydrogen ions in cathode. The main reactions occurring at the aluminum anode and cathode electrodes are:



Hydroxide ion produced in cathode (Eq. 6) and by reaction with boric acid transforms it to negatively charged borates ($[B_3O_3(OH)_4]^{-}$, $[B_4O_5(OH)_4]^{-2}$, $[B_5O_6(OH)_4]^{-}$). By the passage of direct electric current through aluminum electrode, Al^{3+} ions are dissolved in water (Eq. 5) and combined with hydroxyl ions (Eq. 7). First, monovalent metal hydroxides and then positively charged aluminum hydroxides ($Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, $Al_{13}(OH)_{34}^{5+}$) are formed, the latter of which are able to adsorb and remove boron from water due to the positive charge and the ability to create coagulation and flocculation (Xu et al., 2009). However, notwithstanding this, at pHs of more than 9, aluminum hydroxide is soluble in water and lose the ability of adsorption and coagulation. Therefore, the efficiency of this process in boron removal from water at common pHs of water is appropriate and primary and secondary pH adjustments are not necessary in this process (Xu and Jiang, 2008; Xu et al., 2009). Moreover, electrocoagulation is a fairly inexpensive process with high efficiency that produces less sludge and needs smaller physical space (Vasudevan and Lakshmi, 2012).

Several studies have been conducted about boron removal from water using electrocoagulation. These studies have often been conducted on hot mineral waters or waters with concentrations of more than 100 mg/L boron (Bektaş et al., 2004; Sayiner et al., 2008; Xu et al., 2009; Yilmaz et al., 2005). Owing

to their high minerals leading to coprecipitation, their higher electrical conductivity and reduction of required electricity consumption, hot waters have special conditions and are not extensible to waters with low mineral content. As according to the studies by Yoshikawa, in the presence of phosphate and ammonia in wastewater forming $\text{Ca}(\text{OH})_2$ and $(\text{NH}_4)_2\text{HPO}_4$, calcium hydroxide causes boron removal from wastewater by coprecipitation (Yoshikawa et al., 2012). Concentrations of more than 100 mg/L boron in water are within the range of concentrations in industrial wastewater and by increasing the initial concentration of boron, the efficiency of the process increases. However, at concentrations of less than 100 mg/L, there are very few reports of coagulation process efficiency in the removal of boron. Therefore, the aim of this study is to analyze boron removal from water using the process of electrocoagulation at concentrations of less than 100 mg/L. In this regard, the effect of interventional presence of common water anions including bicarbonate, carbonate, sulfate and chloride on the efficiency of boron removal from water is also studied in detail.

Materials and Methods

Electrocoagulation Reactor:

In this experimental study, we used a reactor made of a cube plexiglass with a useful capacity of 250 ml ($L \times W \times h / 10 \times 5 \times 10$ cm) in laboratory scale (Fig. 1). The aluminum anode and cathode plates (15×9 cm with a diameter of 2 mm) were used. The type of DC Power Supply is DAZHENG PS-305D and Power Supply Voltage is up to 30 amps. Calibrated pH meter of Metrohm is used for adjustment of input solution pH. After the preparation of boron stock solution with the initial concentration of 100 mg/L, two mM of NaCl is used for adjustment of its electrical conductivity in the range of municipal water supplies. After sampling and its filtration using a Whatman filter paper with the pore diameter of 0.45 μm , the concentration of boron is determined by a standard method.

The Operation of the Electrocoagulation Reactor:

Considering that pH is one of the most important determining factors in chemical processes, the operation of the reactor began with the analysis of pH effect. The efficiency of the process is studied at pHs of 4, 6, 8, and 10; the distances between the electrodes are 5, 10, 15, 20, 25 and 30 mm, the current density is 4, 6, 8, 10, 12 milliamps per square centimeter; the initial concentration of boron is

10, 20, 30, 50, 70, and 100 mg/L, and the time of reaction is 10, 20, 30, 40, 60, 90, and 120 min. To ensure the reliability of the results, each step is repeated twice.

The Method of Analysis:

We used the standard method 3500 and HACH spectrophotometer (DR5000) to measure the concentration of boron. To calculate the amount of aluminum produced in electrocoagulation process, Faraday's law was applied (You and Han, 2016)

$$W = \frac{ITM}{ZF} \quad (8)$$

W: weight of aluminum produced (g), I: intensity of electric current (A), T: reaction time (s)

M: molar mass of the electrode, Z: number of reactor electrodes, F: Faraday constant

(96485/3 coulomb mole⁻¹ = (C/mol)

Results and Discussion:

1. The Effect of Initial pH:

The results of the effect of initial pH on boron removal are shown in Fig.2. Accordingly, the maximum efficiency of boron removal is at the initial pH of 8 and then we had the most removal rate at pHs of 6, 10, and 4 respectively.

The higher efficiency of the system in boron removal from water at pH=8 can be related to the change of the electric charge of boron and aluminum compounds at different pHs. At lower pHs, boron often appears in the form of boric acid, and at higher pHs, it is usually in the form of anionic borates. On the other hand, at pHs of less than 9, aluminum is often in the form of Al(OH)₃ and thus can play the role of a coagulant. At pHs of more than 9, a part of the aluminum compounds turns into an anion [Al(OH)₄⁻] and apart from sharing the same electric charge with borates, it is soluble in water and cannot coagulate or remove borates. Hence, at pH=8, the maximum rate of boron removal happens due to the formation of borates (anion) and Al⁺³ (cation). In order to remove boron from water using electrocoagulation, Yilmaz et al. also reported the optimum pH to be 8 (Yilmaz et al., 2010).

2. The effect of electrolyte (NaCl) concentration:

Due to the direct connection between concentration of electrolyte (NaCl) in water and electrical conductivity of water, the efficiency of electrocoagulation was studied at a constant AC voltage of 18 V and in 0,20, 40, 120, 200 and 280 mg/L concentrations of NaCl.

According to the results shown in Fig.3, the efficiency of electrocoagulation, due to the fixation of AC voltage, is improved by increasing concentration of dissolved minerals in water, and it reaches its maximum value in mineral concentration of 120 mg/L of NaCl. With increasing the concentration of NaCl to more than 120 mg/L, the efficiency of electrocoagulation was relatively reduced due to rise of the conversion of electrical current to heat and temperature (Khaoula et al., 2013b).

3. The Effect of Distance between Electrodes:

The results of the effect of distance between electrodes are shown in Fig. 4. Accordingly, with a decrease in distance between the electrodes, the efficiency of the reactor increases, so that it reaches the maximum amount of boron removal at a distance of 10 mm.

Due to better transmission of electric current, with a decrease in distance between the electrodes, the efficiency of the system in boron removal increases, so that it reaches the maximum rate at a distance of 10 mm. Nevertheless, the reduction of the efficiency of the system at a distance of 5 mm can be related to the reduction of the distance, hydrogen gas bubbles trapped between anode and cathode, and the resultant resistance against the transmission of electric current between the electrodes. This results are consistent with the results of previous studies (Luqman Chuah and Danial, 2015). As a result, with the reduction of the electrical conductivity, the efficiency of the system reduces too. Therefore, in this study, the optimum distance between the electrodes is set to be 10 mm. An analysis of the literature shows that the effect of distance on boron removal from water by electrocoagulation has not been reported in previous studies. Moreover, different distances ranging from a few millimeters to a few

centimeters have been used in their studies (Bektaş et al., 2004; Isa et al., 2014; Khaoula et al., 2013a).

4. The Effect of Current density:

The effect of current density on the process of boron removal at pH and the optimum distance between the electrodes is analyzed in detail and the results are shown in Fig. 5.

As according to Fig. 5, the efficiency of the reactor in boron removal is risen with an increase in current density, and the maximum amount of removal is at the current of 5.5 mA/cm². With an increase in electric current density, the amount of aluminum produced in anode (mmol) increases according to Faraday's law of induction (Xu et al., 2009), and thus it leads to further production of Al(OH)₃ as a coagulant. Therefore, the production of aluminum can be a possible reason for improvement in the efficiency of the system with an increase in electric current density. An analysis of the proportion of boron removal to aluminum (mmol/mmol) shows that with an increase in electric current density, the proportion of boron removal to aluminum increases too. In this regard, studies by Yilmaz suggest that optimal flow rate is 6 mA/cm², an amount that corresponds to the results of the present plan (Yilmaz et al., 2005).

5. The Effect of Initial Concentration of Boron:

The effect of initial concentration of boron is examined while the solutions contained the initial concentrations of 10, 20, 40, 50, 70, and 100 mg/L, distance between electrodes is 10 mm, the current density is 5.5 mA/cm², and the initial pH is 8.

As according to Fig. 6, by reducing the initial concentration of boron in solution, the efficiency of reactor in boron removal declines. This could be related to the function of adsorption processes in boron removal. By reducing the initial concentration of boron in the adsorption process, the equilibrium concentration drops, resulting in reduced absorption of boron compared to the adsorbent. Thereby, lower initial concentration of boron in the water reduces the absorption of boron on produced aluminum. A review of the related literature shows that these

results correspond to the results of the studies by Sayiner et al. and Bektas et al. (Bektaş et al., 2004; Sayiner et al., 2008). This is while the study by Yilmaz et al. is on natural hot waters, and thus its results are different from the others.

6. The Effect of Reaction Time:

In this study, the effect of reaction time on boron removal, aluminum production rate and the proportion of removed boron to produced aluminum (mmol) is analyzed within the time range of 10 to 120 minutes at the concentration of 100 mg/L, and the results are shown in Fig. 7.

As shown in Fig. 7, the efficiency of the process of electrocoagulation increases with a rise in reaction time. This rising trend has a steep slope by the reaction time of 40 minutes and then, after that, has a linear trend with a low slope. The rising trend of efficiency over the first forty minutes can be related to the high concentration of boron in water. As already mentioned in the fourth part (the effect of concentration), with a decrease in the concentration of boron, due to a decrease in transmission of electric current, its removal from water will also be more difficult. Moreover, an analysis of produced aluminum indicates that with an increase in reaction time, its production is linear. However, due to a decrease in boron concentration in water and a decrease in its equilibrium concentration, the amount of boron adsorbed by aluminum (mmol) has a decreasing trend, as a result of which the amount of boron removal from water in higher reaction times decreases. Therefore, in reaction time of less than 40 minutes, the rising trend of efficiency is higher than its following reactions. This result corresponds to the findings of Bektas et al. and Sayiner et al. (1, 16).

7. Determination of Isotherm of Boron Adsorption:

Given that boron removal from water by electrocoagulation is a variable of different processes including electrochemical processes, coagulation and flotation. One of the important processes involved in elimination of boron from water is the absorption process (Holt et al., 2005; Nidheesh and Singh, 2017). In order to determine the isotherm of boron adsorption on produced aluminum, Langmuir and Freundlich adsorption models are used in the process and their results are shown in Fig. 8 and 9. The isotherm of boron

adsorption on produced aluminum, according to Faraday's law of induction, corresponds to Langmuir ($R^2= 0.77$) and Freundlich ($R^2= 0.79$) isotherms at 20°C.

8. Determination of Kinetics of Boron Adsorption:

An electrocoagulation process always consists of two steps: (1) production of coagulant, (2) pollutant removal by coagulant. Therefore, the kinetics of boron adsorption were determined separately in the first and second steps (Fig. 10). As seen in Fig. 10, the boron absorption rate in the reaction time of 10 min to 40 min (the step 1 or production of coagulant), is lower than the reaction time of 40 min to 120 min (stage 2 or pollutant removal by coagulant). As according to the Fig. 10, the kinetics of boron adsorption in the first and second steps corresponds to the pseudo-second-order model ($R^2= 0.99$), and they can be used to determine the amount of boron adsorption (Henry Ezechi et al., 2015).

8. Interference Effect of common anions:

In order to analyze the effect of common and interfering anions on the efficiency of boron removal, including bicarbonate, carbonate, sulfate and chloride instead of sodium 2 moles of mineral salts (NaHCO_3 , Na_2CO_3 and NaSO_4) are added to distilled water containing boron, and the amounts of boron removal are compared in their presence. A comparison of the amount of boron removal in the presence of bicarbonate, carbonate, sulfate and chloride is shown in Fig. 11, according to which the role of carbonate in interference and reduction of boron removal is 15 %, which is more than the others. This is while sulfate does not reduce the efficiency of boron removal; on the contrary it sufficiently increases the efficiency of boron removal from water.

As previously described, one of the important advantages of electrocoagulation is the production of hydrogen and hydroxide in cathode, hence transformation of boric acid to negatively charged borates. These compounds can be easily adsorbed on the positively charged compounds of aluminum (Xu et al., 2009).

Carbonate is known as a reactive, hydroxide-consuming material. By consuming hydroxide ion produced in cathode, the amount of transformation from boric acid to borates decreases. Therefore,

the amount of boron removal from water is reduced by 15%. This is while chlorine and sulfate anions are not reactive with hydroxide and its consumer, nor does it affect the efficiency of system in boron removal from water. Therefore, the efficiency of electrocoagulation in boron treatment from water and wastewater containing high carbonate or from limewater as a pH regulator can be under the influence of carbonate ion.

Conclusion

The results of this study show that the efficiency of electrocoagulation has a reverse relationship with the initial concentration of boron in water. Nevertheless, the efficiency of this process in boron removal from water at initial concentration of 100 mg/L under optimal conditions (pH= 8, electrode distance= 10 mm, reaction time= 60 min., current density = 5.5 mA/Cm²) is appropriate (70%) and can be used as a treatment process. The isotherm of boron adsorption on produced aluminum, according to Faraday's law of induction, corresponds to Langmuir ($R^2= 0.77$) and Freundlich ($R^2= 0.79$) isotherms and kinetics of boron adsorption on aluminum with the pseudo-second-order model ($R^2= 0.79$). As a consumer of hydroxide, carbonate, which is the conversion factor in transformation of boric acid to negatively charged borates, has more interfering effect than common anion in water, including chloride and sulfate. Moreover, 2 moles of carbonate in water can reduce the efficiency of electrocoagulation by 15%. The process of electrocoagulation can be used for treatment of boron-infected water and wastewaters containing boron at initial concentrations of more than 50 mg/L.

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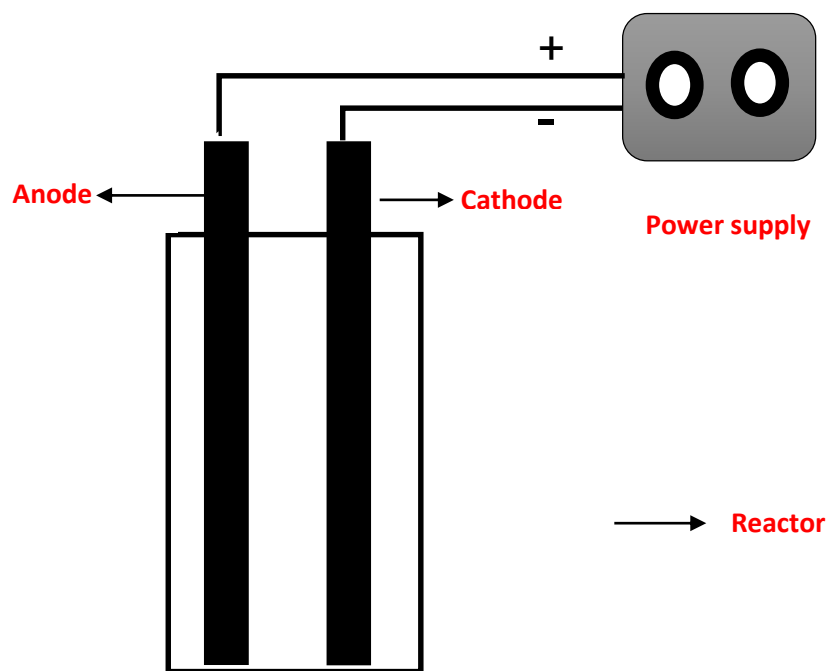


Fig. 1. Schematic of electrocoagulation reactor

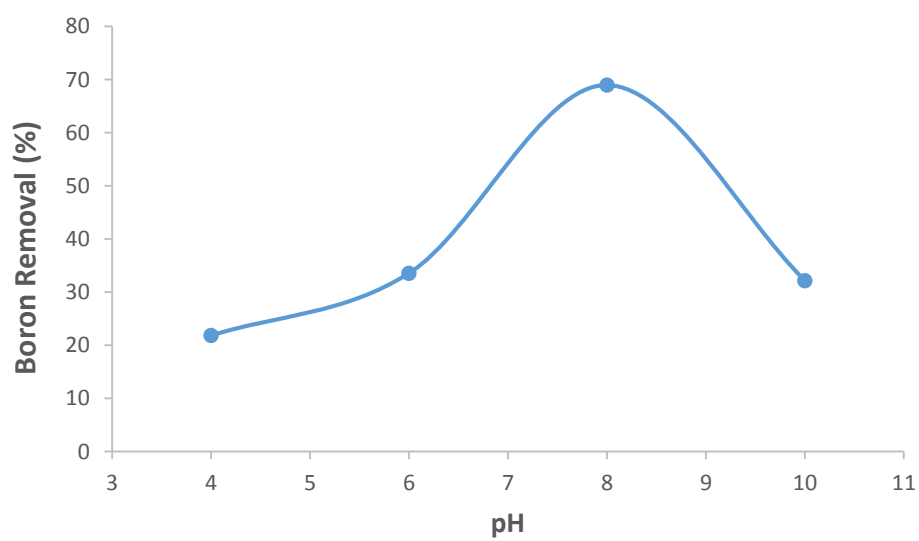


Fig.2. The effect of initial pH

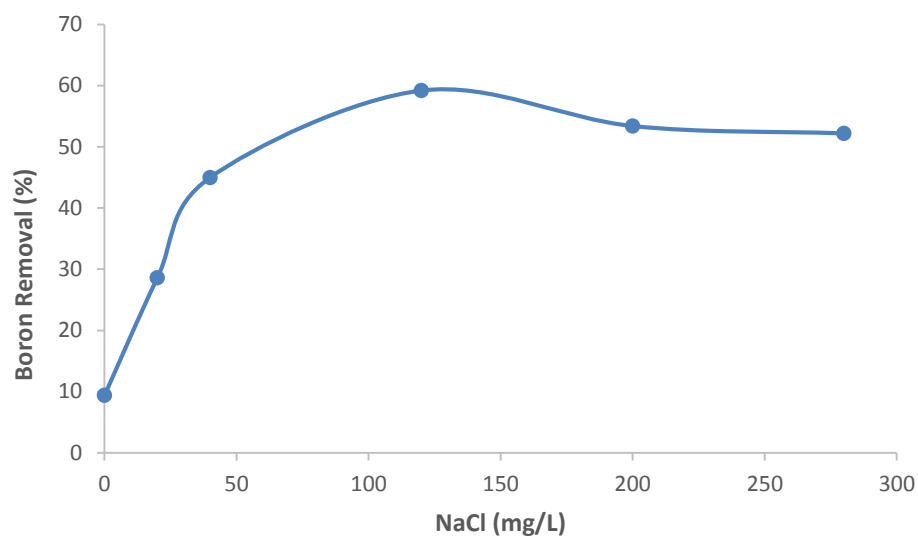


Fig. 3. The effect of the concentration of electrolyte (NaCl)

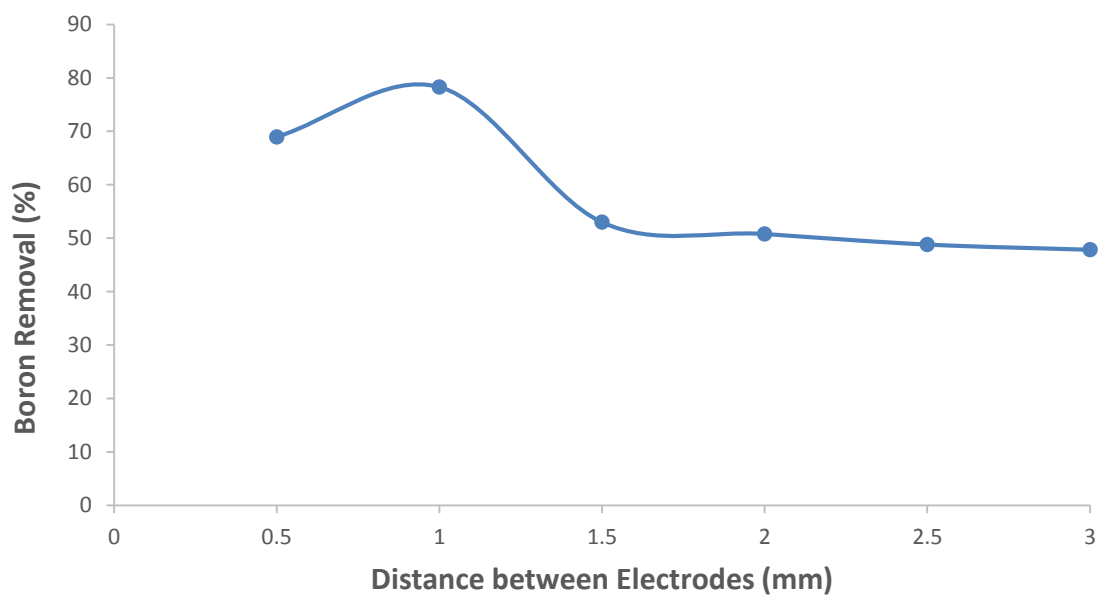


Fig. 4. The effect of distance between electrodes

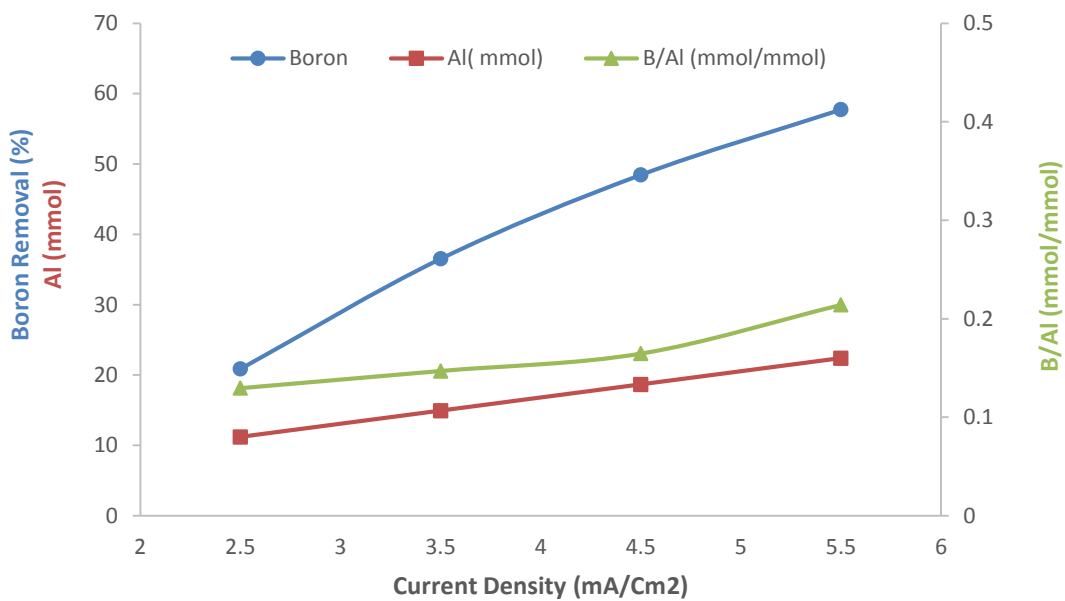


Fig. 5. The effect of the current density

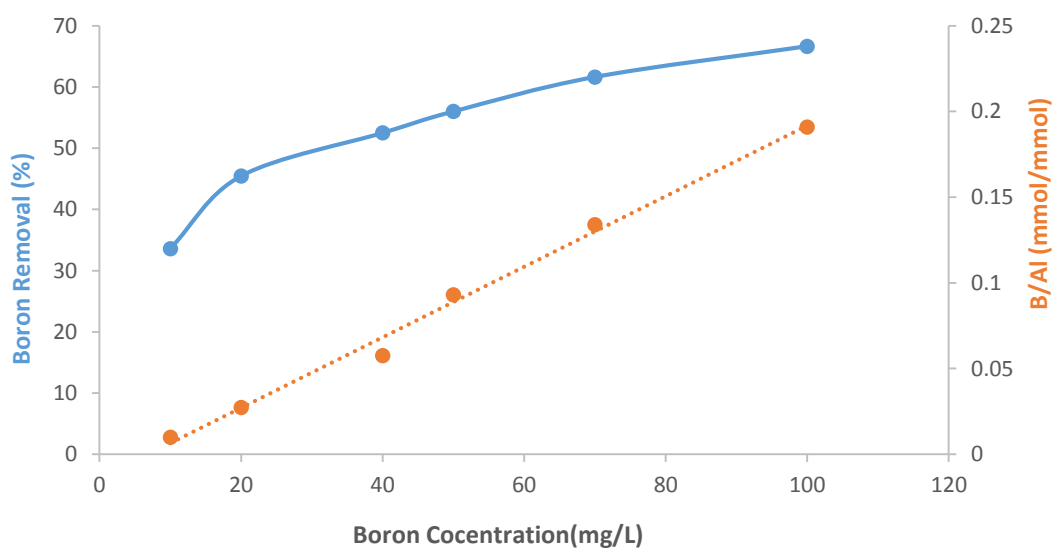


Fig. 6. The effect of Initial concentration

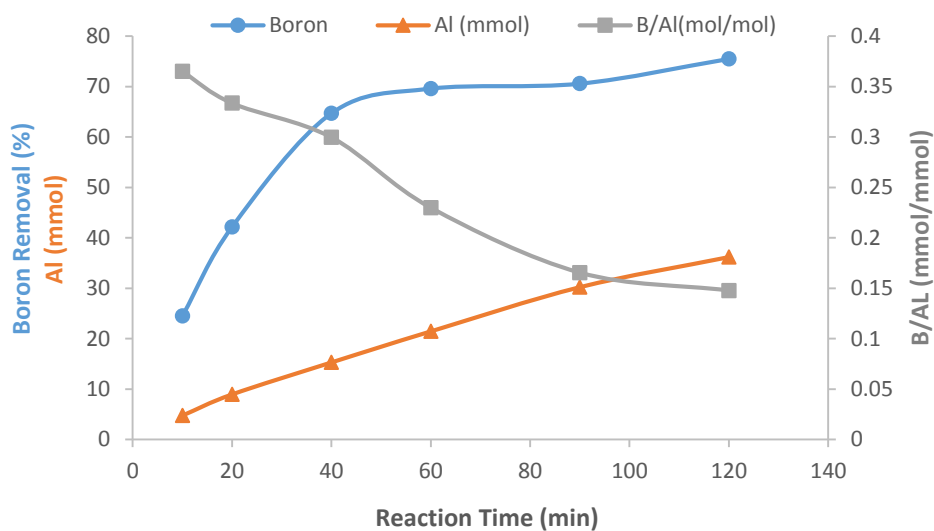


Fig. 7. The effect of reaction time

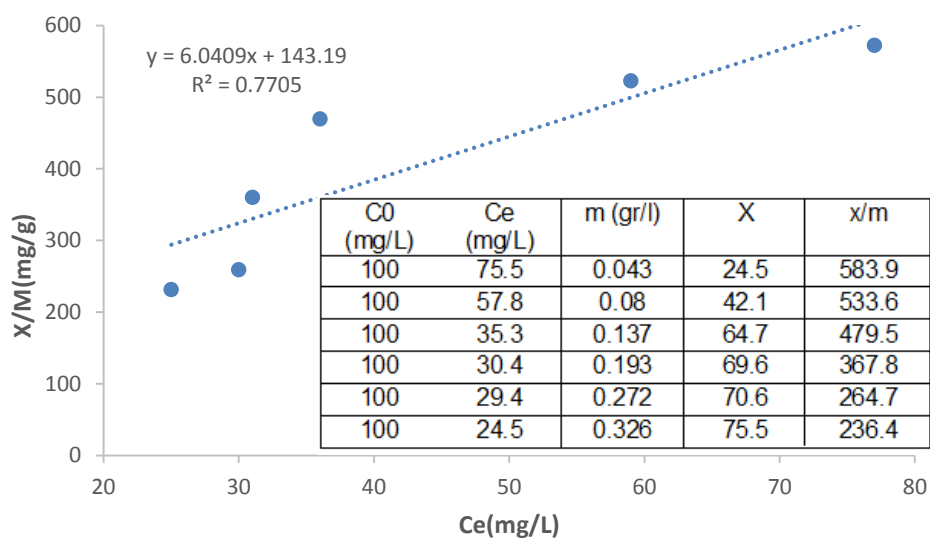


Fig.8. The Langmuir isotherm

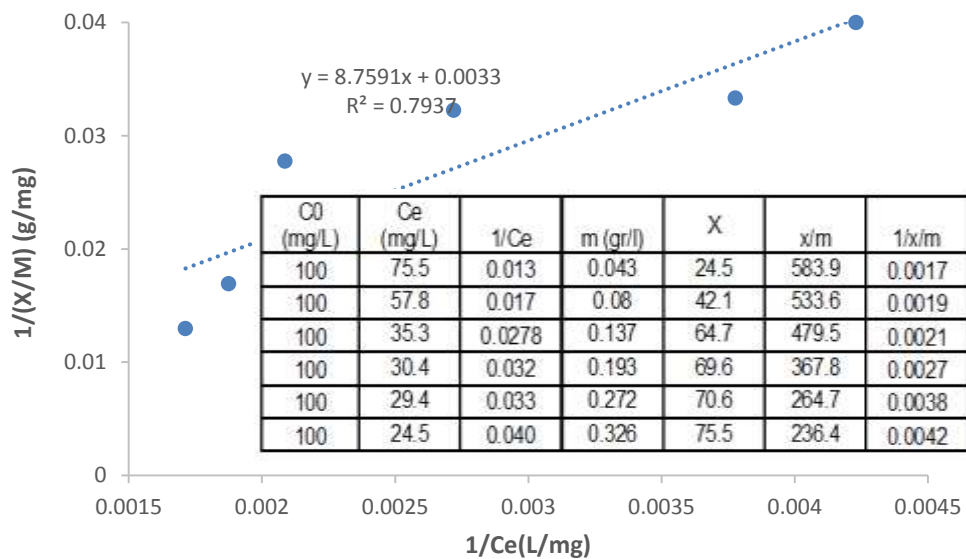


Fig. 9: The freundlich isotherm

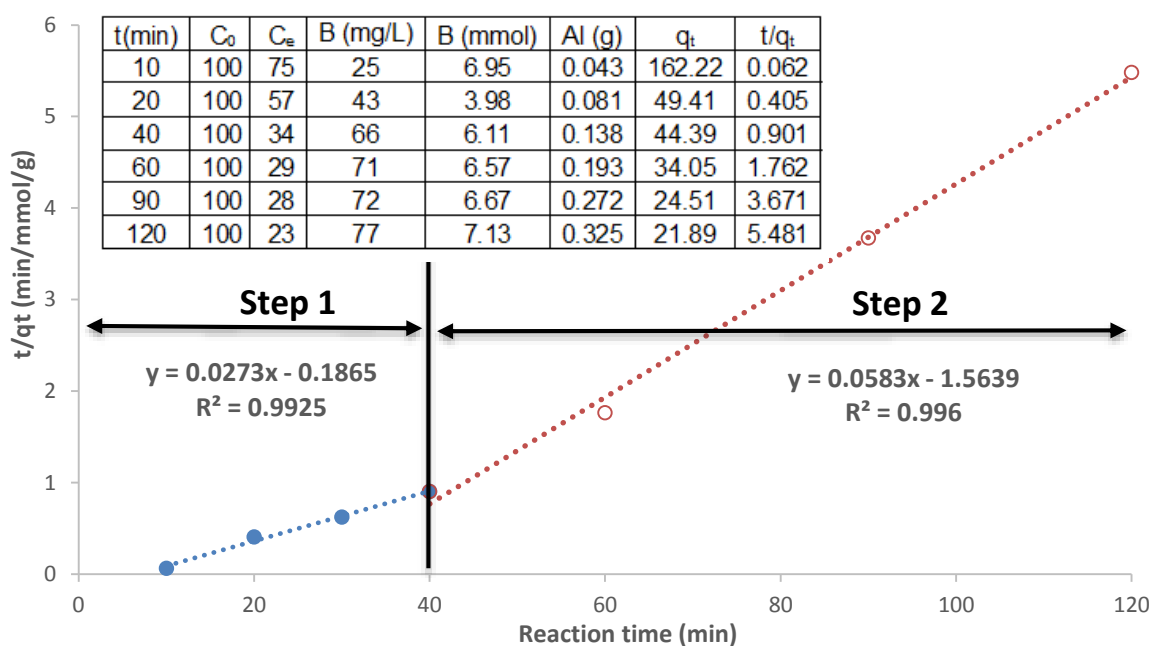


Fig. 10. Kinetics of boron adsorption on produced aluminum

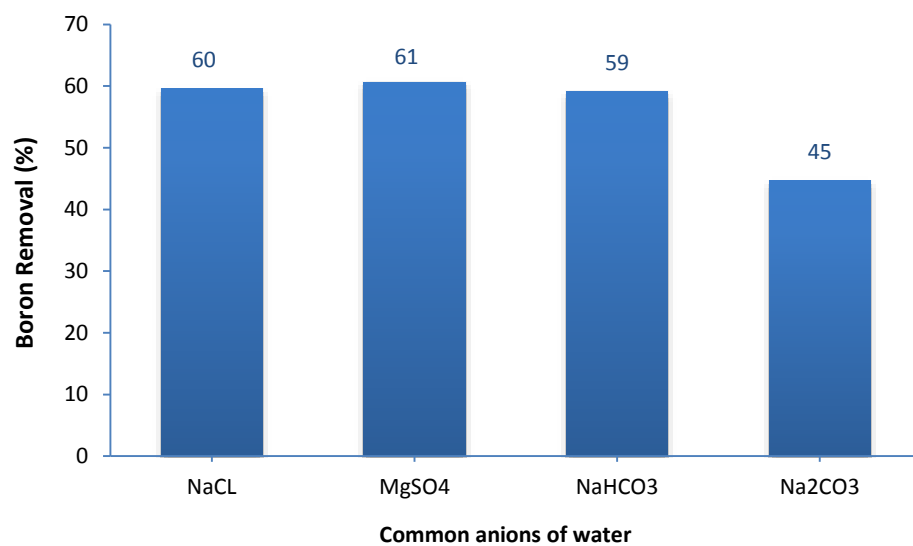


Fig. 11. Interference effect of common anions