A quantitative comparison between electrocoagulation and chemical coagulation for boron removal from boron-containing solution

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Abstract

This paper provides a quantitative comparison of electrocoagulation and chemical coagulation approaches based on boron removal. Electrocoagulation process delivers the coagulant in situ as the sacrificial anode corrodes, due to a fixed current density, while the simultaneous evolution of hydrogen at the cathode allows for pollutant removal by flotation. By comparison, conventional chemical coagulation typically adds a salt of the coagulant, with settling providing the primary pollutant removal path. Chemical coagulation was carried out via jar tests using aluminum chloride. Comparison was done with the same amount of coagulant between electrocoagulation and chemical coagulation processes. Boron removal obtained was higher with electrocoagulation process. In addition, it was seen that chemical coagulation has any effect for boron removal from boron-containing solution. At optimum conditions (e.g. pH 8.0 and aluminum dose of 7.45 g/L), boron removal efficiencies for electrocoagulation and chemical coagulation were 94.0% and 24.0%, respectively.
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Keywords: Electrocoagulation; Chemical coagulation; Boron removal; Aluminum

1. Introduction

Boron has a number of minerals, in nature, mostly calcium and/or sodium borates, such as colemanite, ulexite and tinkal, etc. There is a variety of application including various boron fertilizers, insecticides, corrosion inhibitors in anti-freeze formulations for motor vehicle and other cooling system, buffers in pharmaceutical and dyestuff production, and the use of boron compounds for moderator in nuclear reactor, where anthropogenic water-soluble boron compounds are discharged to aqueous environment. Boron is normally in very low amounts in soil and irrigation waters, but it accumulates very fast in soils irrigated with boron-containing wastewaters because of difficulty of washing it. Boron compounds passing to soil, surface waters and ground waters form many complexes with heavy metals, such as Pb, Cd, Cu, Ni, etc. and these complexes are more toxic than heavy metals forming them. Although little amount of boron is a nutrient for some plants, its excessive amount affects badly the

growth of many agricultural products. The World Health Organization (WHO) has given a recommendation of below 0.3 mg/L boron for the quality of drinking water [1].

In aqueous environments, boron is mainly present as boric acid and partially as borate ions according to the dissociation reaction ($K_a = 6 \times 10^{-10}$, pK_a 9.1) shown in the following equation:

$$B(OH)_{3(aq)} + H_2O \Leftrightarrow B(OH)_{4(aq)}^- + H_{(aq)}^+ \qquad (1)$$
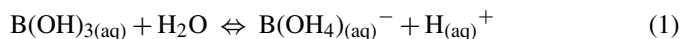
It was estimated that Turkey has about 70% of the known reserves of the world. The known borate reserves in Turkey are located in four main districts, namely Emet, Bigadiç, Kırka and Mustafakemalpaşa [2].

The presence of boron in water has two differentiated origins, one of which is natural origin due to the boron in silts present in the aquifer and the other one is the anthropogenic origin, such as wastewater discharge from boron mines and boric acid plants. In order to prevent the environmental problems of high concentration of boron in waters, their boron contents should be removed by a suitable method [3].

Electrocoagulation is a process consisting of creating metallic hydroxide flocks within the water by electrodissolution the
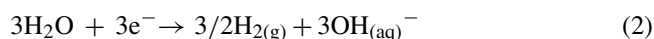
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soluble anodes, usually made of iron or aluminum [4]. The difference between electrocoagulation and chemical coagulation is mainly in the way of which aluminum ions are delivered [5]. In electrocoagulation, coagulation and precipitation are not conducted by delivering chemicals – called coagulants – to the system, but via electrodes in the reactor [6].

Electrocoagulation is based on the fact that the stability of colloids, suspensions and emulsions is influenced by electric charges. Therefore, if additional electrical charges are supplied to the charged particles via appropriate electrodes, the surface charge of particles is neutralized and several particles combine into larger and separable agglomerates [7]. Electrode assembly is the heart of the treatment facility. Therefore, the appropriate selection of its materials is very important. The most common electrode materials for electrocoagulation are aluminum and iron. They are cheap, readily available, and proven effective [8].

The mechanism of electrocoagulation is highly dependent on the chemistry of the aqueous medium, especially conductivity. In addition, other characteristics such as pH, particle size, and chemical constituent concentrations will also influence the electrocoagulation process. When aluminum is used as electrode materials, the reactions are as follows:
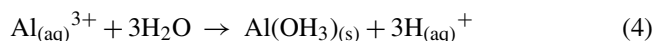
- At the cathode:

$$3H_2O + 3e^- \rightarrow 3/2H_{2(g)} + 3OH_{(aq)}^- \tag{2}$$

- At the anode:

$$Al_{(s)} \rightarrow Al_{(aq)}^{3+} + 3e^- \tag{3}$$

- In the solution:

$$Al_{(aq)}^{3+} + 3H_2O \rightarrow Al(OH)_{3(s)} + 3H_{(aq)}^+ \tag{4}$$

Electrocoagulation has occurred in three steps. In first step, coagulant has formed because of oxidation of anode. In second step, pollutants have stabilized. In last step, stabilized matters have united [9].

Chemical coagulation results from two basic mechanisms; perikinetic or electrokinetic coagulation in which the zeta potential is reduced by ions or colloids of opposite charge to a level below the van der Waals attractive forces and orthokinetic coagulation, in which the micelles aggregate and form clums which agglomerate the colloidal particles.

The addition of high-valence cations depresses the particle charge and the effective distance of the double layer, thereby reducing the zeta potential. As the coagulant dissolves, the cations serve to neutralize the negative charge on the colloids. This occurs before visible flock formation, and mixing rapid is effective in this phase. Microflocks are then formed which retain a positive charge in the acid range because of the adsorption of H^+. These microflocks also serve to neutralize and coat the colloidal particle. Flocculation agglomerates the colloids with a hydrous oxide flock. In this phase, surface adsorption is also active. Colloids not initially adsorbed are removed by enmeshment in the flock [10]. Chemical coagulation separation–treatment technology is generally carried out in the following four phases:

1. Addition of the chemical to the wastewater.
2. Rapid (flash) mixing to distribute the chemical homogeneously throughout the wastewater.
3. Slow mixing to encourage flocculation (formation of the insoluble solid precipitate).
4. Filtration, settling, or decanting to remove the flocculated solid particles.

Chemical coagulation followed by sedimentation is a proven technique for the treatment of high suspended solids wastewater especially those formed by the colloidal matters. Research and practical applications have shown that coagulation will lower the pollution load and could generate an adequate water recovery [11–14]. Coagulation is mainly done with inorganic metal salts, e.g. aluminum and ferric sulphates and chlorides which are widely used as coagulants in water and wastewater treatment and in some other applications. Their mode of action is generally explained in terms of two distinct mechanisms: charge neutralization of negatively charged colloids by cationic hydrolysis products and incorporation of impurities in an amorphous hydroxide precipitate so-called sweep flocculation [15]. The relative importances of these mechanisms depend on factors such as pH and coagulant dosage.

The aim of this paper was to study the comparison of the removal of boron from aqueous solution by electrocoagulation and chemical coagulation. The electrocoagulation process was examined under different values of current density (CD), pH, temperature of solution and initial boron concentrations and the coagulation process was examined under different values of coagulant dosage, pH, temperature of solution and initial boron concentrations in order to determine optimum operating conditions. Obtained experimental data was compared between each treatment process.

2. Experimental

In this study, boron concentration was chosen high because boron concentration from boron industry wastewater was quite high. Wastewater samples used in the experiments were prepared synthetically using $Na_2B_4O_7$ having 99.99% of purity from Merck. The solution with boron concentration of 100 mg/L was prepared by dissolving 459.1 mg borax dried at 105 °C in distilled water and completing with distilled water to 1 L. The same operations were repeated for the solutions with boron concentrations of 500 and 1000 mg/L with different $Na_2B_4O_7$ weights. The parameters chosen in the electrocoagulation and chemical coagulation experiments carried out were pH, initial boron concentration, amount of coagulant and temperature of solution whose ranges were given in Table 1.

2.1. Electrocoagulation experiments

A laboratory-scale batch reactor (16 cm × 8 cm × 8 cm), made of plexiglass was used in electrocoagulation experiments. Two groups of alternating electrodes, being cathodes and anodes (by eight plates of each type) made of aluminum were arranged vertically. The net spacing between the aluminum electrodes

Table 1
Experimental parameters

Parameters	Range
pH	4.0, 6.0, 8.0 and 10.0
Amount of coagulant (g)	1.42, 2.88, 4.37, 5.92 and 7.45
Initial boron concentration (mg/L)	100, 500 and 1000
Temperature of solution (K)	
Electrocoagulation	283, 293, 303 and 313
Chemical Coagulation	293, 313, 333 and 353

was 5 mm. They were connected to terminals of a direct current power supply characterized by the ranges 0–5 A for current and 0–30 V for voltage. At the beginning of each run the solution of boron of the desired concentration fed into the reactor. Each run was timed starting with the dc power supply switching on. Electrical conductivity, pH and temperature were all measured using a WTW multimeter. Electrocoagulation process is demonstrated schematically in Fig. 1.

2.2. Chemical coagulation experiments

Chemical coagulation was evaluated using an adaptation of the standard jar testing technique, with AR grade (99.8% pure) aluminum chloride as the chemical coagulant. NaOH and HCl solutions were added for any subsequent pH adjustment. Jar tests were performed using a Phipps and Bird six-paddle stirrer. Desired aluminum was added to boron-containing solution. The procedures consisted of a 2 min rapid mix (120 rpm), 25 min slow mix (25 rpm), and a 60 min settling period. After settling for 60 min, supernatants were collected to measure residual boron potentiometrically by means of mannitol and measured pH values with a pH-meter (WTW). All reagents used were of analytical grade. Chemical coagulation process was demonstrated schematically in Fig. 2.

The analytical determination of boron was done potentiometrically by means of mannitol, which forms a complex compound with boric acid. For both processes compared, boron analyses were carried out following: Solution pH was adjusted to 7.60 after sample was filtered. Then, 5 g mannitol was added to solution. The solution was titrated with 0.5N KOH until solution pH became 7.60. Boron amount was calculated from KOH consumption. 1 ml 0.5N KOH is equal to 17.41 mg B_2O_3 [16]. This method was preferred in order to prevent aluminum inter-

Fig. 1. Schematic view of the experimental system of electrocoagulation process (1: dc power supply, 2: electrocoagulation cell, 3: magnetic stirrer, 4: pump, 5: constant temperature circulator, 6: pH and conductivity meter, 7: ampermeter, 8: voltmeter, 9: pH control unit and, 10: computer).
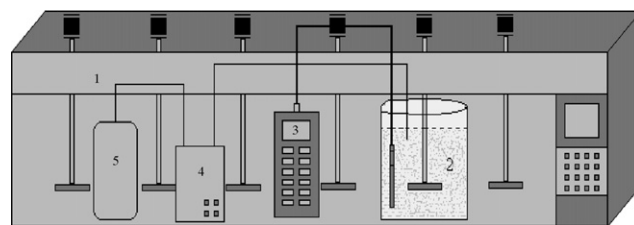

Fig. 2. Schematic view of the experimental system of chemical coagulation process (1: jar tester, 2: chemical coagulation cell, 3: pH meter, 4: pump, 5: coagulant reservoir).

ference in boron detection done by spectrophotometric methods such as carmine, azomethine-H and curcumin methods [17].
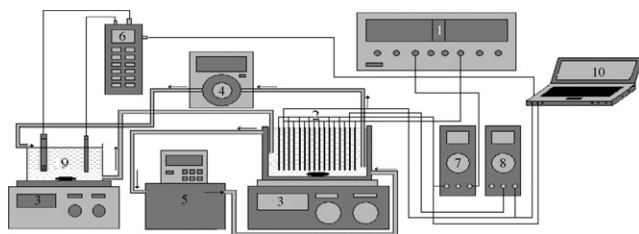
3. Results and discussion

3.1. The effects of parameters

In the runs, it has been compared the effects of parameters such as pH, initial boron concentration, amount of coagulant and temperature of solution under the conditions which the reaction time hold in constant for both electrocoagulation and chemical coagulation processes. Obtained experimental data's are following.

3.2. The effect of dissolved aluminum

The authors [18,19] working with aluminum electrodes have observed that aluminum cathodes are dissolved and this phenomenon cannot be explained by the electrochemical process. Generally, the cathodes are supposed to be attacked by hydroxyl ions generated during water reduction. The reaction written as follows:

$$2Al + 6H_2O + 2OH^- \rightarrow 2Al(OH)_{(4)}^- + 3H_{(2)} \qquad (5)$$
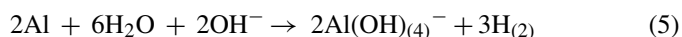
This reaction is the most probable, but it has not been strictly proved that it occurs in the electrocoagulation process. We have compared the experimental amount of aluminum created at the cathode with the theoretical amount calculated using the second Faraday's law. This law is used to relate the current flow (I for time t) to the amounts (m) of aluminum and hydroxide ions generated within the reactor, and hence provide a theoretical amount for total aluminum

$$m = \frac{ItM}{nF} \qquad (6)$$

n is the number of electrons transferred in the reaction at the electrode, M the molecular weight (g/mol), I is current flow and F is Faraday's constant (96,486 C/mol). In experiments at constant current intensity (1–5 A), the cell voltage was recorded over electrolysis times 3 h. Samples of electrolyte were taken and the dissolved aluminum concentrations determined by spectrophotometer. Total dissolved Al + 3 analyses were carried out at 525 nm wave lengths according to Eriochrome cyanine R method [20]. Obtained experimental and theoretical data were shown in Fig. 3. It was shown that current efficiency was approximately 142–149% from experimental results. The
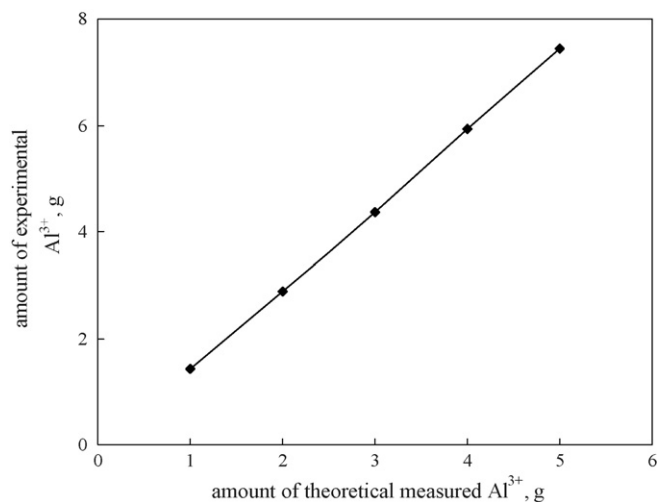
Fig. 3. Amount of theoretical measured aluminum vs. amount of experimental measured aluminum.

Fig. 4. Activity–pH diagram for Al(III) species in equilibrium with Al(OH)$_3$ (amorphous).

amount of Al^{3+} used chemical coagulation experiments was equal to amount of total dissolved Al^{3+} in electrocoagulation experiments. The amount of Al^{3+} used all experiments for both processes were shown in Table 2.

3.3. The effect of pH

Solution pH is one of the important factors that affect the performance of electrochemical and chemical processes. Hence experiments were conducted to study the effect of pH on the boron removal. To evaluate this effect for each processes, a series of experiments were performed, using solution containing boron of 1000 mg/L. The effect of pH on the boron removal was examined at 4.0, 6.0, 8.0, and 10.0 pH's. For electrocoagulation, current intensity of 5.0 A (equal to 7.45 g total dissolved Al^{3+}) and stirring speed of 150 rpm were kept constant in the experiments. For chemical coagulation, amount of coagulant of 7.45 g Al^{3+} and 2 min rapid mix (120 rpm), 25 min slow mix (25 rpm) were kept constant in the experiments. Boron removal increased with increasing the pH up to 8.0, and then decreased for each treatment processes. When boron removal efficiency reached up to 94% by electrocoagulation, this value reached to 24% by chemical coagulation. The extent of hydrolysis depends upon total metal concentration and pH, as well as the amount of other species present in solution. Fig. 4 is the solubility diagram for aluminum hydroxide, Al(OH)$_3$(s), assuming only mononuclear species. The solubility boundary denotes the thermodynamic
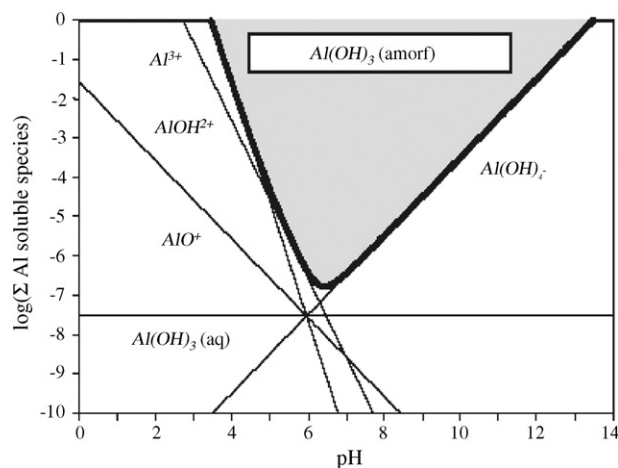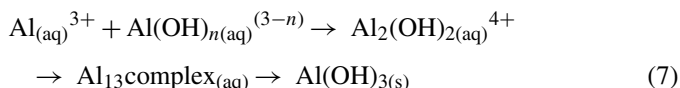
equilibrium that exists between the dominant aluminum species at a given pH and solid aluminum hydroxide. The minimum solubility occurs at approximately pH 6.5, with solubility increasing as the solution becomes more acidic or alkaline. However, as the aluminum concentration increases, polynuclear aluminum complexes are formed and aluminum hydroxide precipitates, as shown below [13]

$$Al_{(aq)}^{3+} + Al(OH)_{n(aq)}^{(3-n)} \rightarrow Al_2(OH)_{2(aq)}^{4+}$$
$$\rightarrow Al_{13}complex_{(aq)} \rightarrow Al(OH)_{3(s)} \qquad (7)$$

If activity–pH diagram for Al(III) species in equilibrium with Al(OH)$_{3(s)}$ is investigated, it will be seen that dominant Al(III) species is in the form of Al(OH)$_{3(s)}$ at pH 5.0–8.5 interval. Al(OH)$_{4(aq)}^-$ forms at the higher pH and Al(OH)$_{4(aq)}^-$ is a dissolving form and does not form flocks [21]. Experiments were carried at pH 4–10 because Al(OH)$_3$ formed at pH 5.0–8.5 interval, according to activity-pH diagram for Al(III) species shown in Fig. 4. When effect of pH on boron removal was investigated, borate ions species presented in solution must be known. For this purpose, diagram showing borate ions species at varied pH intervals was demonstrated in Fig. 5. As seen Fig. 5, when solution pH was higher than pH 9.0, borate ions in solution was dominantly B(OH)$_4^-$ form. When solution pH was lower than pH 9.0, borate ions in solution was dominantly B(OH)$_3$ form. The highest boron removal efficiency was obtained at pH 8.0 because boron was at B(OH)$_3$ form and the formation Al(OH)$_{3(s)}$ was a quite high at this pH.

Table 2
The amount of used Al^{3+} for both processes

Current intensity (A)	Electrocoagulation process			Chemical coagulation process
	Amount of theoretical Al^{3+} (g)	Amount of theoretical Al^{3+} (g)	Current efficiency (%)	Amount of Al^{3+} added to solution
1	1	1.42	142.17	1.42
2	2	2.88	144.12	2.88
3	3	4.37	145.67	4.37
4	4	5.92	146.85	5.92
5	5	7.45	148.95	7.45
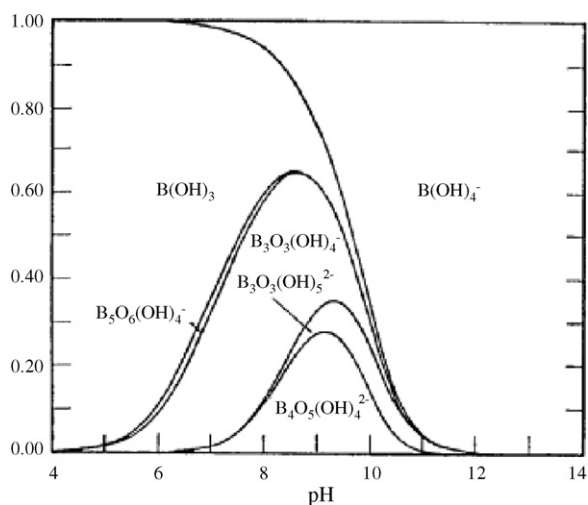
Fig. 5. Changing of borate ions species depending pH in aqueous media.

Fig. 7. The effect of initial boron concentration on boron removal by electrocoagulation and chemical coagulation, pH 8.0, 7.45 g amount of coagulant.

When boron removal by electrocoagulation process was investigated, the pH was kept nearly constant throughout the experiments by adding (w/w, 63%) concentrated HNO_3. Maximum 3–4 mL concentrated HNO_3 was added to solution for constant pH during experiment period. According to following reaction, HNO_3 kept nearly constant pH via neutralization

$$OH_{(aq)}^- + HNO_{3(aq)} \rightarrow H_2O + NO_{3(aq)}^- \qquad (8)$$
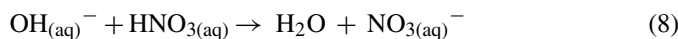
Under these conditions, nitrate ion was not expected to be effect on boron removal. As a result of obtained data, optimum boron removal was reached at pH 8.0. This is probably resulting from borate and Al(III) species which form in solution at this pH. The results obtained for each treatment processes are shown graphically in Fig. 6 for 1000 mg/L boron concentration.

3.4. The effect of initial boron concentration

The effect of initial boron concentration on the boron removal was examined with solutions including boron of 100, 500 and
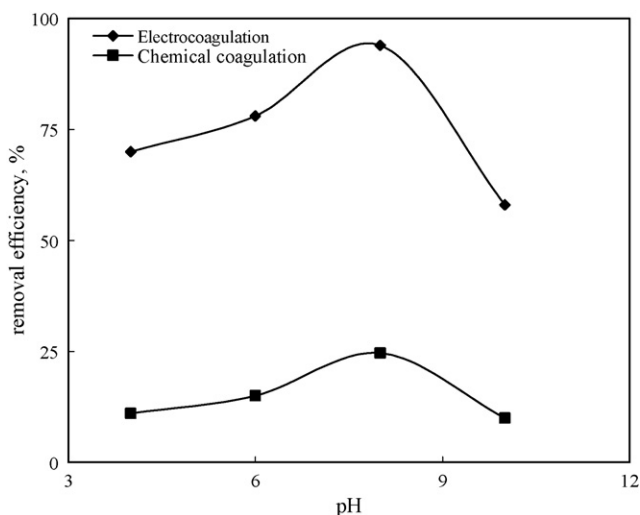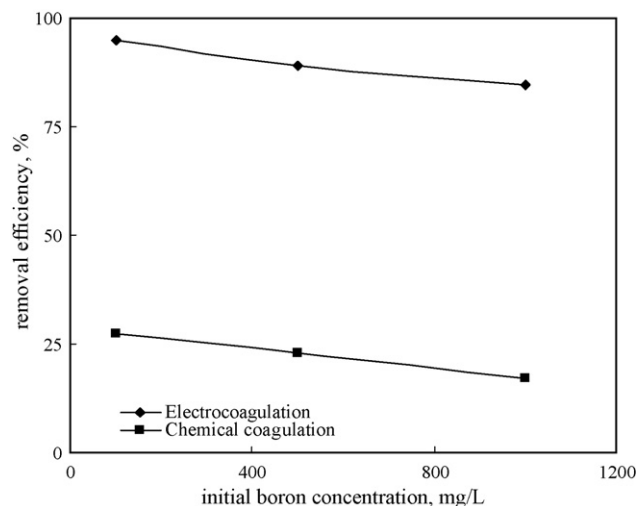

Fig. 6. The effect of pH on boron removal by electrocoagulation and chemical coagulation, 1000 mg/L initial boron concentration, 7.45 g amount of coagulant.

1000 mg/L for each process. Current intensity of 5.0 A (equal to 7.45 g total dissolved Al^{3+}), optimum pH of 8.0 and stirring speed of 150 rpm were kept constant in the experiments for electrocoagulation processes. For chemical coagulation, amount of coagulant of 7.45 g Al^{3+}, pH 8.0 and 2 min rapid mix (120 rpm), 25 min slow mix (25 rpm) were kept constant in the experiments. As a result of experimental data, boron removal efficiency decreased with increasing boron concentration for each treatment processes. This can be explained as following: although the same amount Al^{3+} passed to solution at the same current density and the same amount of coagulant for all boron concentration, Al^{3+} was insufficient for solutions including higher boron concentration. In addition to above-mentioned state, boron removal efficiency of electrocoagulation process was very higher than that of chemical coagulation process. The results obtained were shown graphically in Fig. 7.

3.5. The effect of amount of coagulant

The current density determines the coagulant dosage rate at electrocoagulation process. Thus, this parameter should have a significant impact on removal efficiencies of pollutants. To investigate the effect of current density on the boron removal, a series of experiments were carried out by solutions containing a constant pollutants loading with current intensity being varied from 1.0 to 5.0 A (equal to 1.42–7.45 g total dissolved Al^{3+}). 8.0 of solution of pH and stirring speed of 150 rpm were kept constant and boron concentration was taken 1000 mg/L in experiments. Amount of coagulant is an important parameter for chemical coagulation which more pollutant reacts with increasing amount of coagulant. Consequently, increasing amount of coagulant must increase removal efficiency. To investigate the effect of amount of coagulant on the boron removal, a series of experiments were carried out by solutions containing a constant pollutants loading and coagulant amount being varied from 1.42 to 7.45 g Al^{3+}. In experiments, 8.0 of solution pH and 2 min rapid mix (120 rpm), 25 min slow mix (25 rpm) were kept
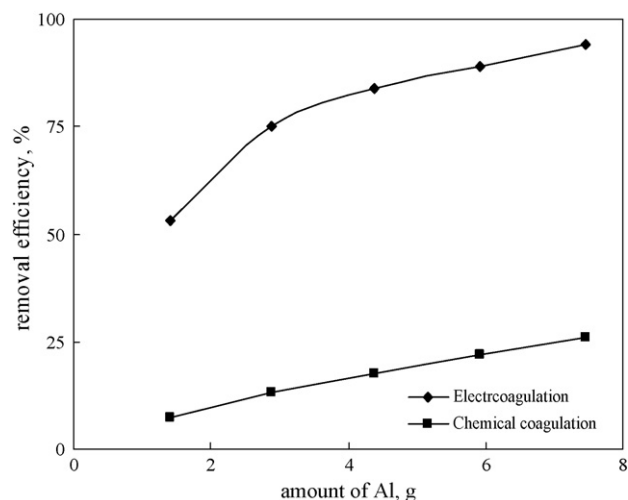
Fig. 8. The effect of amount of coagulant on boron removal by electrocoagulation and chemical coagulation, 1000 mg/L initial boron concentration, pH 8.0.
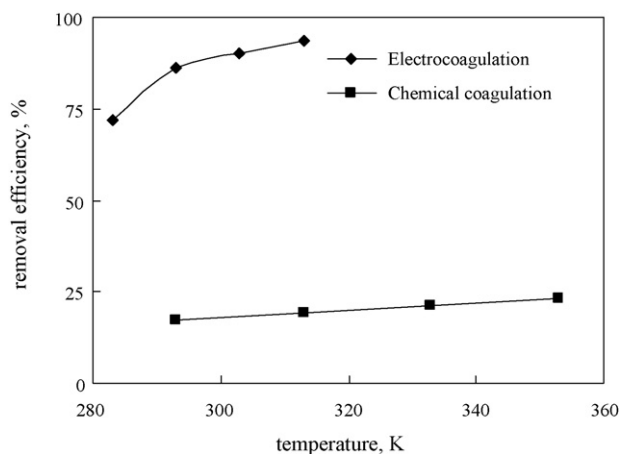

Fig. 9. The effect of temperature of solution on boron removal by electrocoagulation and chemical coagulation, 1000 mg/L initial boron concentration, pH 8.0, 7.45 g amount of coagulant.

constant and boron concentration was taken 1000 mg/L. When the effect of amount of coagulant on boron removal was investigated, amounts of coagulant passing to solution and added to the solution were equal for both processes. Dissolving rate of Al electrode increased with increasing current intensity in electrocoagulation due to equation (5). Consequently, boron removal increased with increasing current density and amount of coagulant because more Al^{3+} passed to solution at these state and formation rate of $Al(OH)_3$ increased. On the other hand, boron removal efficiency for electrocoagulation was very higher than those of chemical coagulation. It was demonstrated that aluminum was used more effectively during electrocoagulation compared with chemical coagulation as performed in this study. The results obtained were shown graphically in Fig. 8 for 1000 mg/L boron concentration.

3.6. The effect of temperature of solution

The effect of temperature on the boron removal was examined with 283, 293, 303 and 313 K for electrocoagulation and 293, 313, 333 and 353 K for chemical coagulation. Current intensity of 5.0 A (equal to 7.45 g total dissolved Al^{3+}), initial boron concentration of 1000 mg/L, stirring speed of 150 rpm and optimum pH of 8.0 were kept constant for electrocoagulation in the experiments. Amount of coagulant of 7.45 g Al^{3+}, initial boron concentration of 1000 mg/L, 2 min rapid mix (120 rpm), 25 min slow mix (25 rpm) and optimum pH of 8.0 were kept constant for chemical coagulation in the experiments. Increasing temperature of solution increased boron removal efficiency for each treatment processes. Temperature had an important effect on boron removal and boron removal efficiency was more effective at higher temperatures. It was not investigated high temperature intervals because boron removal efficiency was a quite high at electrocoagulation process. Contrary to above statement, very high temperature intervals were investigated to increase boron removal efficiency at chemical coagulation process. However, it was not reached to desired boron removal efficiency. Boron

removal efficiency was reached from 72% to 96.6% with increasing temperature in electrocoagulation process; however boron removal efficiency reached from 17% to 25% with increasing temperature in chemical coagulation process. Formed $Al(OH)_3$ flocks reacted faster and more to constant pollutant because mobility and collisions of ions with hydroxide polymer could be increase. As a result, boron removal efficiency increased with increasing temperature for both processes. The results obtained were shown graphically in Fig. 9 for 1000 mg/L boron concentration.

3.7. Economic evaluation

The economic study included chemicals consumption, metallic sludge disposal and energy consumption. The chemical costs were determined by using $AlCl_3$ cost of 0.8 \$/kg and using other chemicals (e.g. HNO_3, NaOH for pH adjustment) cost of 0.03 \$/m^3 for chemical coagulation. The electrochemical costs were determined by using Al electrode cost of 1.8 \$/kg for electrochemical coagulation. Sludge transportation and disposal was evaluated at 0.015 \$/kg for each process. The total cost for each process tested was evaluated in term of money spent per kg of dry soil treated. The energy consumed was estimated at a cost of 0.06 \$/kWh. The electrical energy consumption at electrocoagulation process could be explained with following equations:

$$W = \frac{VIt}{v} \tag{9}$$

where W is the electrical energy consumption (kW h/m^3), V the potential (V), I the current (A), t the time (h), v is the volume of solution (m^3). When effect of current intensity on boron removal was investigated at electrocoagulation process, values of current intensity were varied form 1.0 to 5.0 A. The energy consumption was calculated for these values. Obtained data was graphically shown Fig. 10. Values of the total operating cost are calculated based on economic data obtained from Turkish market in 2006.
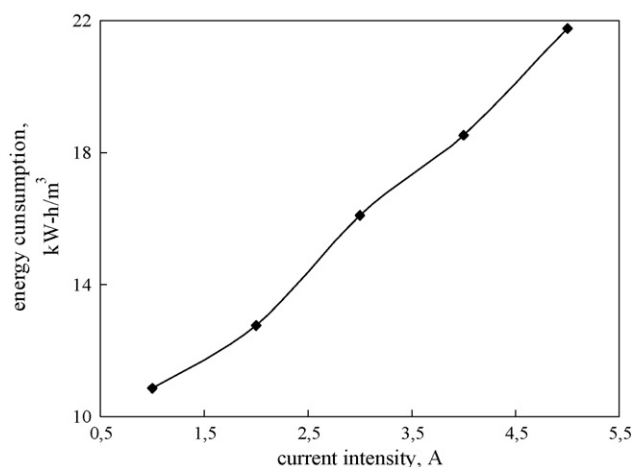
Fig. 10. The effect of current intensity on total operating cost for Al electrodes.

4. Conclusion

This study has shown the possibility to use electrochemical technique to remove efficiently boron compared to traditional chemical coagulation using aluminum chloride. The comparison of electrocoagulation and chemical coagulation processes used for boron treatment, demonstrated to have the practical advantage of electrochemical treatment in terms of effectiveness. The use of aluminum as sacrificial electrode material in the treatment of boron wastewater by electrocoagulation was found to be pH dependent. The most effective removal capacity was achieved at the pH 8. Increasing boron concentration decreased boron removal efficiency. The treatment rate was seen to increase with increasing the current density. The highest current density gave the quickest treatment for boron removal from synthetically prepared waters containing boron equivalent industrial wastewaters'. Although all operational parameters were same in electrocoagulation and chemical coagulation, boron removal efficiency between each treatment processes was a quite different. Therefore, chemical coagulation process was not appropriate for boron removal efficiency. The fundamental physical separation processes are different for chemical coagulation and electrocoagulation, with settling being the key removal path in the former case. In electrocoagulation, however, the dominant removal path is determined by the applied current. The mechanism of coagulant delivery is a key difference between chemical coagulation and electrocoagulation performance. Explanation of the relationship between thermodynamic equilibrium and reaction kinetics in terms of pollutant removal are clearly significant for a deeper understanding of electrocoagulation. In a comparison of the processes, electrocoagulation significantly outperformed chemical coagulation with aluminum chloride as a treatment for boron. For example, at pH 8.0 and aluminum dose of 7.45 g/L, boron removal efficiency was 94.0% and 24.0% for electrocoagulation and chemical coagulation, respectively.

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