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An empirical model for parameters affecting energy consumption in boron removal from boron-containing wastewaters by electrocoagulation

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Abstract

In this study, it was investigated parameters affecting energy consumption in boron removal from boron containing wastewaters prepared synthetically, via electrocoagulation method. The solution pH, initial boron concentration, dose of supporting electrolyte, current density and temperature of solution were selected as experimental parameters affecting energy consumption. The obtained experimental results showed that boron removal efficiency reached up to 99% under optimum conditions, in which solution pH was 8.0, current density 6.0 mA/cm^2, initial boron concentration 100 mg/L and solution temperature 293 K. The current density was an important parameter affecting energy consumption too. High current density applied to electrocoagulation cell increased energy consumption. Increasing solution temperature caused to decrease energy consumption that high temperature decreased potential applied under constant current density. That increasing initial boron concentration and dose of supporting electrolyte caused to increase specific conductivity of solution decreased energy consumption. As a result, it was seen that energy consumption for boron removal via electrocoagulation method could be minimized at optimum conditions. An empirical model was predicted by statistically. Experimentally obtained values were fitted with values predicted from empirical model being as following;

$$[\mathrm{ECB}] = 7.6 \times 10^6 \times [\mathrm{OH}]^{0.11} \times [\mathrm{CD}]^{0.62} \times [\mathrm{IBC}]^{-0.57} \times [\mathrm{DSE}]^{-0.04} \times [T]^{-2.98} \times [t]$$

Unfortunately, the conditions obtained for optimum boron removal were not the conditions obtained for minimum energy consumption. It was determined that support electrolyte must be used for increase boron removal and decrease electrical energy consumption.

Keywords: Electrocoagulation; Boron removal; Energy consumption; Empirical model; Aluminum electrode

1. Introduction

The elemental boron is widely distributed in nature in low concentrations. Turkey has the largest boron reserve which is approximately 90 million tonnes in the world. It was estimated that Turkey has about 70% of the reserves of the world. The known borate reserves in Turkey are located in four main districts, namely Emet, Bigadiç, Kırka and Mustafakemalpasa [1]. The main sources of boron from wastes, whose presence is detected in surface waters, are urban wastes rich in detergents and cleaning products; industrial wastes, which can come from a wide range of different activities as well as several chemical products used in agriculture [2].

Boron is an essential element for plants. It is present in animal tissue in low concentrations (about 1 mg/L) and is probably an essential micronutrient for the living; however, no essential biochemical function has yet been positively identified to establish its essentiality to animals and humans [3]. The WHO has given a preliminary limit of 0.3 mg/L for drinking water. The EU regulations are suggesting a guideline of 1.0 mg/L. Most of surface and ground waters are below this limit; however, boron contamination of waters is a concern around the world. Boron deficiency in plants may result reduced growth, yield loss, and even death, depending on the severity of deficiency. The tendency of boron to accumulate in vegetable tissues constitutes a

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Nomenclature

ECB	energy consumption per m^3 boron solution ($kW\,h/m^3$)
IBC	initial boron concentration (mg/L)
DSE	dose of supporting electrolyte (mM)
CD	current density (mA/cm^2)
T	temperature (K)
t	reaction time (min)
I	current (A)
V	potential (V)
W	electrical energy consumption ($kW\,h/m^3$)
R	resistance (Ω)
v	volume of solution (m^3)

potential hazard to the health of those consuming food and water with high boron content [4]. In result, although boron is vital as a micronutrient element for plants' growth, it can be detrimental at higher concentrations [5]. For these reason, boron levels in drinking and irrigation waters are endeavor to get under control.

Electrochemical technology contributes in many ways to a cleaner environment and covers a very broad range of technology. Examples include removal of impurities from process liquids, air and soil, recycling of process streams [6]. During the last two decades, a special research field, environmental electrochemistry has been developed. Environmental electrochemistry [7–14] involves electrochemical techniques or methods to remove impurities from gases, liquids and soil to prevent or minimize environmental pollution. In particular, emissions to the atmosphere, discharges of pollutant into waters and disposal of solids to land sites have to be minimized.

Electrocoagulation involves the generation of coagulants in situ by dissolving electrically either aluminum or iron ions from aluminum or iron electrodes, respectively. The metal ion generation takes place at the anode; hydrogen gas is released from the cathode. Also, the hydrogen gas would help to float the flocculated particles out of the water. This process sometimes is called electroflocculation. The electrodes can be arranged in a monopolar or bi-polar mode. The materials can be aluminum or iron in plate form or packed form of scraps such as steel turnings, millings, etc.

The most widely used electrode materials in electrocoagulation process are aluminum and iron. In the case of aluminum, main reactions are as

$$\text{Anode}: Al_{(s)} \rightarrow Al_{(aq)}{}^{3+} + 3e^- \quad \text{(I)}$$

$$\text{Cathode}: 3H_2O + 3e^- \rightarrow \tfrac{3}{2}2H_{2(g)} + 3OH_{(aq)}{}^- \quad \text{(II)}$$

On the other hand, at high pH values, both cathode and anode may be chemically attacked by OH^- ions [15]:

$$2Al_{(s)} + 6H_2O + 2OH_{(aq)}{}^- \rightarrow 2Al(OH)_{4(aq)}{}^- + 3H_{2(g)} \quad \text{(III)}$$

Al^{3+} and OH^- ions generated by electrode reactions (I) and (II) react to form various monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2{}^+$, $Al_2(OH)_2{}^{4+}$, $Al(OH)_4{}^-$, and polymeric species such as $Al_6(OH)_{15}{}^{3+}$, $Al_7(OH)_{17}{}^{4+}$, $Al_8(OH)_{20}{}^{4+}$, $Al_{13}O_4(OH)_{24}{}^{7+}$, $Al_{13}(OH)_{34}{}^{5+}$, which transform finally into $Al(OH)_{3(s)}$ according to complex precipitation kinetics[16–18].

$$Al_{(aq)}{}^{3+} + 3H_2O \rightarrow Al(OH)_{3(s)} + 3H_{(aq)}{}^+ \quad \text{(IV)}$$

Freshly formed amorphous $Al(OH)_{3(s)}$ "sweep flocks" have large surface areas which is beneficial for a rapid adsorption of soluble organic compounds and trapping of colloidal particles. Finally, these flocks are removed easily from aqueous medium by sedimentation or H_2 flotation.

The economic aspect of the electrocoagulation process is not well investigated, except a few researches. In this respect, the effects of the electrode material type as well as process variables on process economics need to be studied in detail [19].

The aim of this paper is to study parameters affecting energy consumption in the removal of boron from synthetic wastewaters by electrocoagulation, which is a new processes applied to boron containing wastewater. The process was examined under different values of current density (CD), pH, initial boron concentrations (IBC), and dose of supporting electrolyte (DSE), in order to determine optimum operating conditions.

Table 1
Experimental parameters

Parameters	Range
pH	4.0, 5.0, 6.0, 7.0, 8.0 and 9.0
Current density (mA/cm^2)	1.2, 2.4, 3.6, 4.8 and 6.0
Boron concentration (mg/L)	100, 250, 500 and 1000
Temperature (K)	283, 293, 303 and 313
Dose of supporting electrolyte (mM) $CaCl_2$	5, 10 and 15

2. Experimental

In this study, high boron concentrations were chosen because boron concentration in boron industry wastewater was quite high. Boron industries wastewater containing large amount impurities was not used in experiment because the impurities could have effected to boron removal mechanism by electrocoagulation method. Wastewater samples used in the experiments were prepared synthetically using $Na_2B_4O_7$ having 99.99% of purity from Merck. The parameters chosen in the experiments carried out were pH, boron concentration, current density, temperature and dose of supporting electrolyte, whose ranges were given in Table 1. pHs of the original boron solutions were about 9.0. To adjust these pHs to studied pH in investigating the effect of pH, appropriate amounts of HNO_3 were added to original solution.

A laboratory-scale reactor (16 cm × 8 cm × 8 cm), made of plexiglass, was used in all experiments (Fig. 1). Aluminum electrodes were used as cathodes and anodes. The net spacing between the aluminum electrodes was 0.5 cm. Electrodes were connected to a digital dc power supply characterized by the ranges 1.2–6.0 mA/cm^2 for current and 0–30 V for voltage in monopolar mode. Two digital multimeters as ampermeter and voltmeter were used to measure the current passing through the circuit and the applied potential, respectively. The electrocoag-

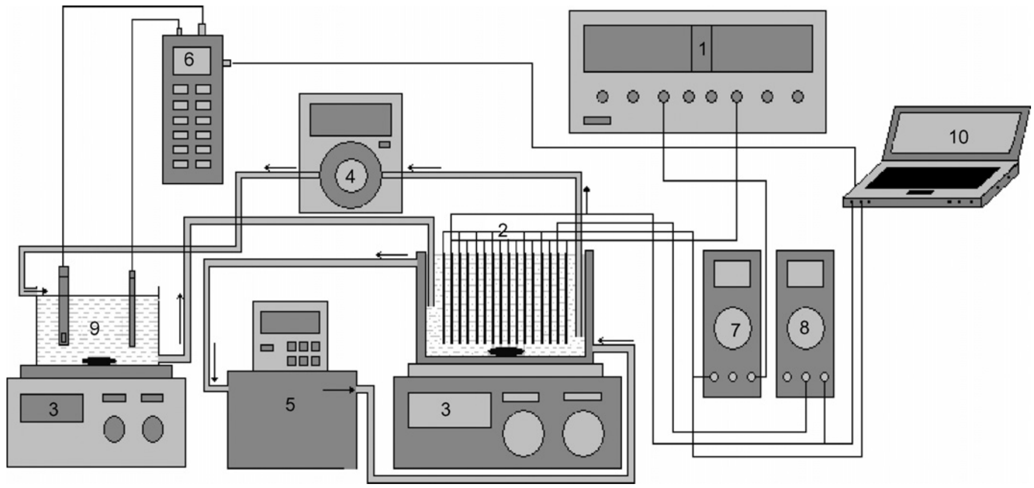

Fig. 1. Schematic view of the experimental system (1: dc power supply, 2: electrocoagulation cell, 3: magnetic stirrer, 4: pump, 5: constant temperature circulator, 6: pH and conductivity meter, 7: ampermeter, 8: voltmeter, 9: pH control unit and 10: computer).

ulation unit has been stirred at 150 rpm by a magnetic stirrer. During the experiments, temperature, conductivity and pH of the wastewaters were measured by a multi-parameter. The reactor fed with solution of boron of the desired concentration at the beginning of each run. Each run was timed starting with the dc power supply switching on.

The analytical determination of boron was done potentiometrically by means of mannitol, which forms a complex compound with boric acid. For this purpose, boron analysis was carried out as follows: 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. One millilitre 0.5N KOH is equivalent to 17.41 mg B_2O_3 [20].

In order to simulate the processes in the electrocoagulation tank a computer programme was used. The electrical energy consumption in the electrocoagulation tank was modeled according to nonlinear estimation method and Statistica 6.0 was chosen as simulation software. Empirical model is an important step in any modeling effort. In calibration, values were assigned to the parameters used in the model such that the difference between model predictions and observations is at minimum.

3. Results and discussion

3.1. The effects of parameters

The efficiency of boron removal from wastewaters by electrochemical process depends on several operating parameters: initial pH, current density, initial boron concentration, dose of supporting electrolyte and temperature.

3.1.1. The effect of pH

The influent pH is one of the important factors affecting electrical energy consumption in electrochemical process. To investigate this effect, a series of experiments were performed using solution containing boron of 500 mg/L. The effect of pH on the boron removal was examined at 4.0, 5.0, 6.0, 7.0, 8.0, and 9.0 pHs. Current density of 3.0 mA/cm^2 and stirring speed of 150 rpm was kept constant in the experiments. The results obtained are shown graphically in Fig. 2 for 500 mg/L boron concentration. As seen in Fig. 2, the lowest energy consumption curve was obtained in the experiments carried out with 4.0 of solution pH because it had the highest conductivity. When specific conductivity of solution with pH 4.0 reached to 8.80 mS/cm, specific conductivity of solution with pH 9.0 remained at 4.61 mS/cm. The solutions which are given conductivity values had 500 mg/L initial boron concentrations. The effect of conductivity on electrical energy consumption could be explained with following equations:

$$W = \frac{VIt}{v} \tag{1}$$

where W is the electrical energy consumption (kW h/m^3), V the potential (V), I the current (A), t the time (h) and v is the volume of solution (m^3). Applied potential could be explained with the

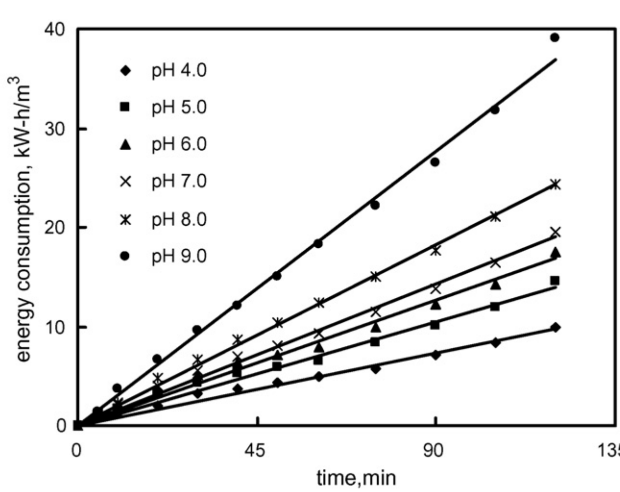

Fig. 2. The effect of pH on energy consumption (boron concentration: 500 mg/L, current density: 3.0 mA/cm^2, solution temperature: 293 K, and stirring speed: 150 rpm).

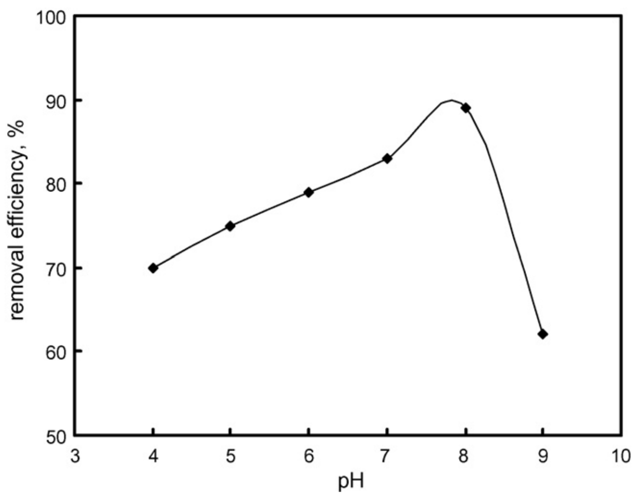

Fig. 3. The effect of pH on boron removal (boron concentration: 500 mg/L, current density: 3.0 mA/cm^2, solution temperature: 293 K and stirring speed: 150 rpm).

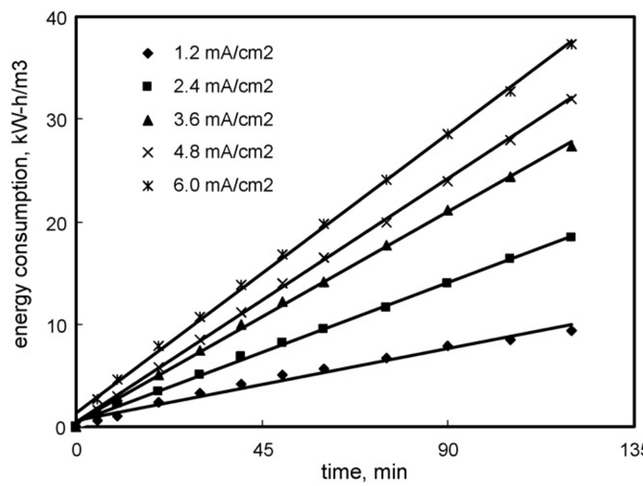

Fig. 4. The effect of current density (1.2–6.0 mA/cm^2) on energy consumption (boron concentration: 500 mg/L, solution pH: 8.0, solution temperature: 293 K and stirring speed: 150 rpm).

equation:

$$V = IR \quad (2)$$

where R is the resistance (Ω). From Eqs. (1) and (2), following equation could be obtained:

$$W = \frac{I^2 Rt}{v} \quad (3)$$

Electrical conductivity is a measure of how well a material accommodates the transport of electric charge. Electrical conduction is an electrical phenomenon in which a material (solid or otherwise) contains movable particles with electric charge, which can carry electricity. When a difference of electrical potential is applied to a conductor, an electric current appears. Conductivity stated as the inverse of electrical resistivity, is defined as the ratio of the current density to the electric field strength and has the SI units of Siemens per meter (S/m).

Electrical conductivity caused to decrease energy consumption because there was a relationship between electrical conductivity and resistance. The pH of $Na_2B_4O_7$ solution containing 500 mg/L boron was about 9.0 and its conductivity was about 2.70 mS/cm. The decreasing pH of solution caused to rise of electrical conductivity. Thus, high conductivity values of solution caused to low resistance values and low energy consumption. According to these data, obtained energy consumption curves and boron removal efficiencies were demonstrated in Figs. 2 and 3, respectively.

3.1.2. The effect of current density

The current density determines the coagulant dosage rate. Thus, this parameter should have a significant impact on removal efficiencies of pollutants. To investigate the effect of current density on the energy consumption, a series of experiments were carried out by solutions containing a constant pollutants loading with current density being varied from 1.2 to 6.0 mA/cm^2. Solution pH of 8.0 and stirring speed of 150 rpm were kept constant and boron concentration of 500 mg/L was taken in experiments. The results obtained are shown graphically in Fig. 4. Although that current density was increased from 1.2 to 6.0 mA/cm^2 increased from 57.4 to 93.0% of boron removal efficiency, energy consumption reached from 7.7 to 38.48 kW h/m^3. The obtained results for boron removal were demonstrated in Fig. 5. High electrical energy consumption with increasing current density was an expected result because energy consumption impressed linearly current density as seen in Eq. (1). Although higher current density caused to solve more electrode material and remove more pollutant, this state was not desired for electrical energy consumption.

3.1.3. The effect of initial boron concentration

The effect of initial boron concentration on electrical energy consumption was examined with solutions including boron of 100, 250, 500 and 1000 mg/L. Current density of 3.0 mA/cm^2, optimum pH of 8.0 and stirring speed of 150 rpm were kept constant in the experiments. Increasing initial boron concentration

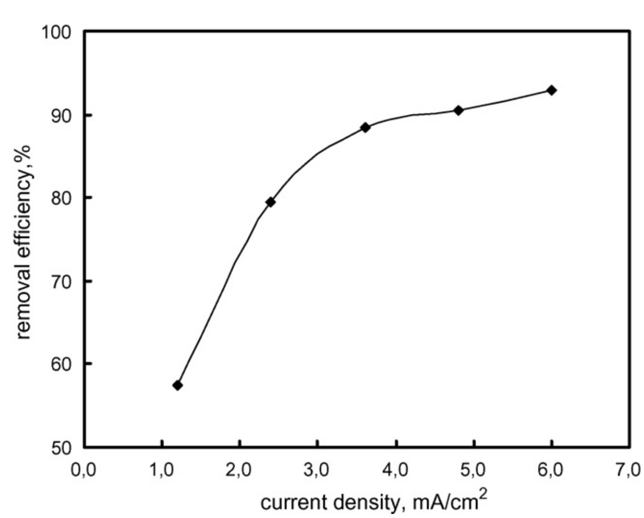

Fig. 5. The effect of current density on boron removal (boron concentration: 500 mg/L, pH: 8.0, stirring speed: 150 rpm).

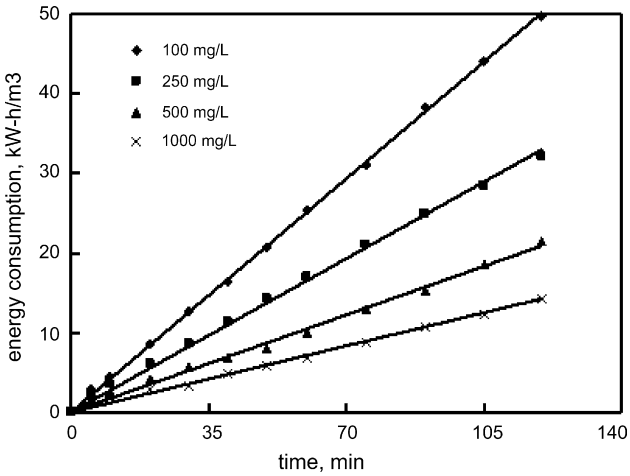

Fig. 6. The effect of initial boron concentration on energy consumption (current density: 3.0 mA/cm^2, pH: 8.0, solution temperature: 293 K and stirring speed: 150 rpm).

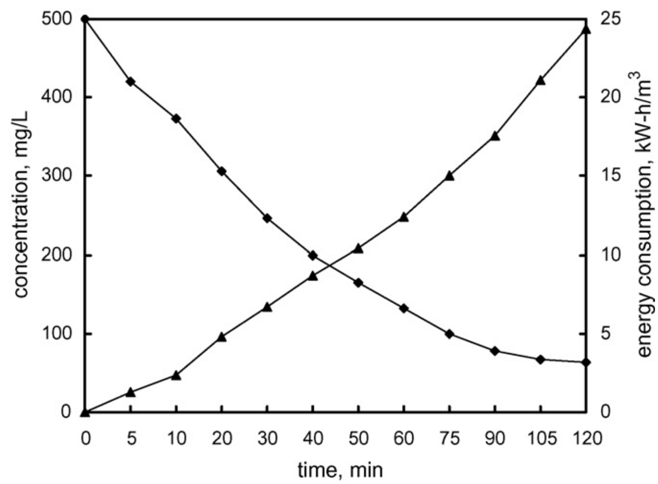

Fig. 8. Change in boron removal and energy consumption as a function of reaction time.

increased amount of ions in solution. As a result of this case, the solution conductivity increased with increasing boron concentration. Increasing of amount of ionized species in solution obtained more transmission for electric applied under constant current density. In order word, this situation caused to decrease total resistance in electrocoagulation cell. Because of increasing boron concentration, potential applied to solution and energy consumption decreased. In Fig. 8, the change of boron removal and energy consumption for 500 mg/L was demonstrated as a function of reaction time. As seen in Fig. 8, energy consumption increased constantly with increasing boron removal during reaction time. Other initial boron concentrations had the same tendency. The results obtained were shown graphically in Figs. 6–8, respectively.

3.1.4. The effect of solution temperature

The effect of temperature on the boron removal was examined with 283, 293, 303 and 313 K. Current density of 3.0 mA/cm^2, initial boron concentration of 500 mg/L, stirring speed of 150 rpm and optimum pH of 8.0 was kept constant in the experiments. Higher temperature gives higher conductivity, hence lower energy consumption [21]. The effect of temperature on energy consumption could be explained as follows; the higher temperature was the lower viscosity of solution. Thus, the fluidity of solution increased and the ions moved more easily in solution. On the other hand, increase in the temperature increased kinetic energy of particles in the solution and the particles moved more rapidly. As a result, conductivity increased and energy consumption decreased. Increasing temperature of solution increased boron removal efficiency, too. The results were shown graphically in Figs. 9 and 10, respectively.

3.1.5. The effect of dose of supporting electrolyte

The effect of dose of supporting electrolyte on the boron removal was examined with 5, 10 and 15 mM $CaCl_2$. Current density of 3.0 mA/cm^2, optimum pH of 8.0 and stirring speed of 150 rpm were kept constant in the experiments. All doses of

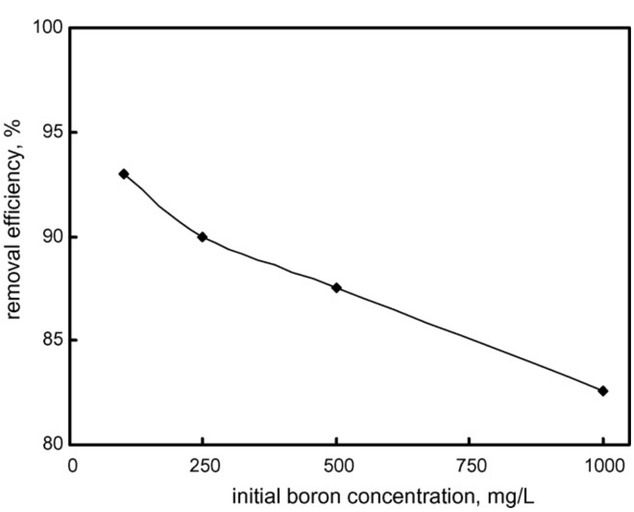

Fig. 7. The effect of initial boron concentration on boron removal (current density: 3.0 mA/cm^2, pH: 8.0, solution temperature: 293 K and stirring speed: 150 rpm).

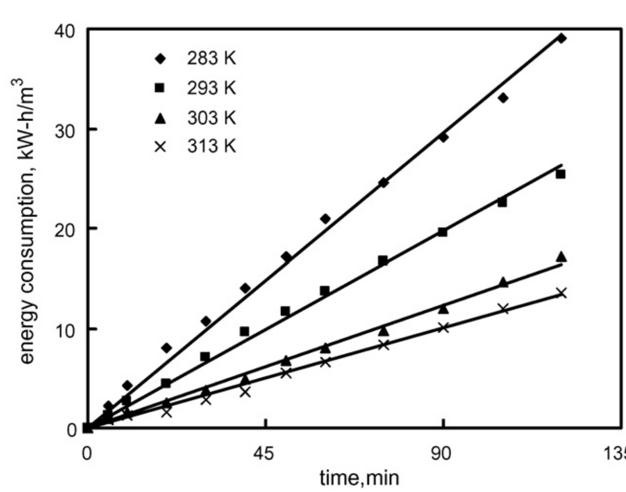

Fig. 9. The effect of temperature on energy consumption (current density: 3.0 mA/cm^2, pH: 8.0, initial boron concentration: 500 mg/L and stirring speed: 150 rpm).

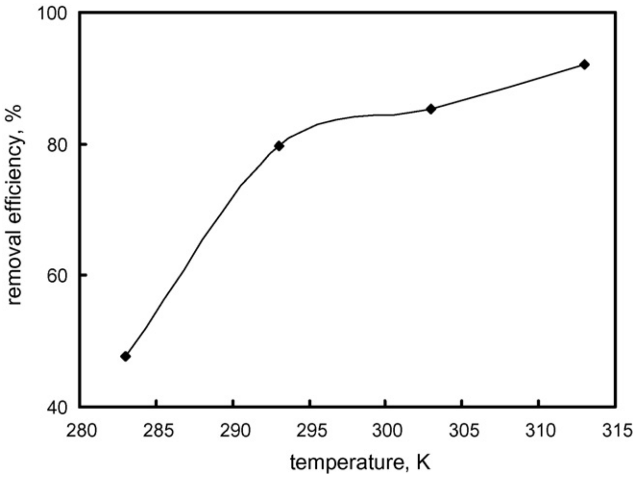

Fig. 10. The effect of temperature on boron removal (current density: 3.0 mA/cm^2, pH: 8.0, initial boron concentration: 500 mg/L and stirring speed: 150 rpm).

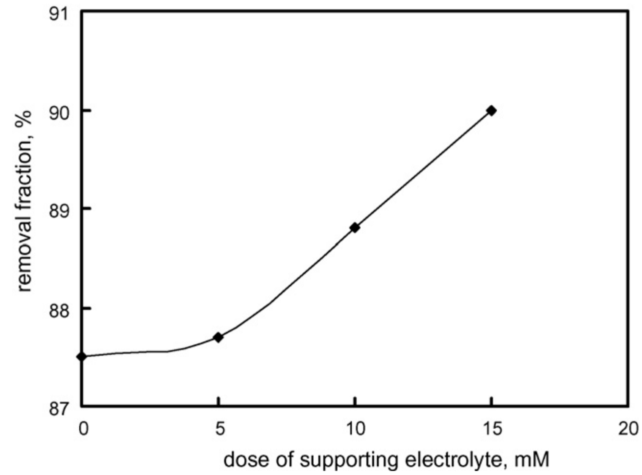

Fig. 12. The effect of dose of supporting electrolyte on boron removal (current density: 3.0 mA/cm^2, pH: 8.0, initial boron concentration: 500 mg/L and stirring speed: 150 rpm).

supporting electrolyte were examined for boron concentration of 500 mg/L. The energy consumption decreased with increasing dose of supporting electrolyte because potential decreased under constant current density. Increasing supporting electrolyte dose increased amounts of ions in solution and the conductivity of solution. In order word, this situation caused to decrease total resistance in electrocoagulation cell. The results showed that boron removal increased with increasing dose of supporting electrolyte as seen in Figs. 11 and 12, respectively.

3.2. Empirical model

An empirical model is developed to relate the critical parameters such as pH, current density, initial boron concentration, solution temperature and dose of supporting electrolyte for boron removal using electrocoagulation process. Based on the operational parameters, an empirical equation is given to calculate the optimal boron removal rate. The results show good agreement between the experimental data and the predictive equation. In this study, a manual approach was used for model calibration. Default values of parameters related to electrochemical processes were employed initially. Then, differences between predicted and observed values noted and adjustments made in parameter values until an efficient match between observed and calculated values of desired variables were reached. Change in the default values of six model parameters gave a reasonable match for the investigated variables. Experimentally obtained values including pH, current density, initial boron concentration, temperature of solution and dose of supporting electrolyte were easily transferred on the Statistica 6.0 programme as first step using user friendly graphical icons. Some operational parameters of the electrocoagulation which were entered to the Statistica 6.0 programme as operational data are given at Table 1. It is concluded that the electrocoagulation technology using aluminium

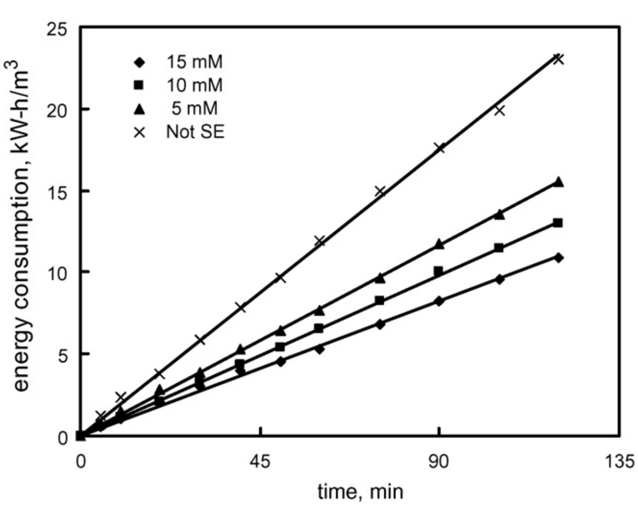

Fig. 11. The effect of dose of supporting electrolyte on energy consumption (current density: 3.0 mA/cm^2, pH: 8.0, initial boron concentration: 500 mg/L and stirring speed: 150 rpm).

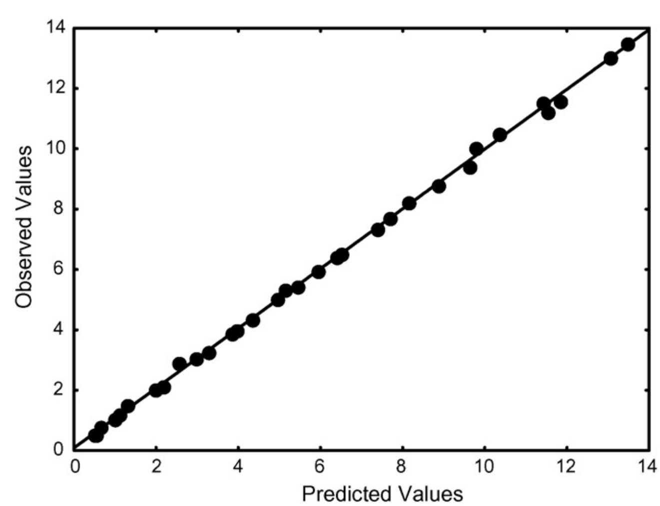

Fig. 13. Plot of predicted energy consumption per amount of removal boron vs. observed energy consumption per amount of removal boron.

electrodes is a viable process for boron removal of industrial effluent that contains excess boron. Empirical model obtained via Statistica 6.0 programme was found as follows, obtained and predicted values were fitted as seen Fig. 13.

$$[\mathrm{ECB}] = 7.6 \times 10^6 \times [\mathrm{OH}]^{0.11} \times [\mathrm{CD}]^{0.62} \times [\mathrm{IBC}]^{-0.57} \times [\mathrm{DSE}]^{-0.04} \times [T]^{-2.98} \times [t]$$

4. Conclusions

In this study, it was investigated parameters affecting energy consumption in boron removal from boron containing wastewaters prepared synthetically, via electrocoagulation method.

The effect of pH on energy consumption was based on specific conductivity of solution. The lower the pH of solution had the higher the conductivity and the lower the energy consumption. When the effect of current density on energy consumption was investigated, it was seen that current density had an important parameter on energy consumption. The energy consumption increased with increasing current density because high current density caused to increase total resistance formed in electrocoagulation cell under constant specific conductivity of solution. Thus, applied potential to system and energy consumption increased. However, obtained results shown that high current density caused to increase boron removal. When effect of initial boron concentration on energy consumption was investigated, the obtained results shown that increasing boron concentration increased conductivity of solution. Thus, solution with higher boron concentration had more ions at the same volume. The higher conductivity values decreased energy consumption. The effect of temperature on energy consumption could be explained as following; because of decreasing viscosity of solution by increasing the temperature, the fluidity of solution increased. So, ions in solution moved more easily, conductivity increased and energy consumption decreased. On the other hand, increasing kinetic energies of particles in the solution increased conductivity and decreased energy consumption. The constituted experiments to investigate effects of dose of supporting electrolyte on energy consumption shown that the effects of dose of supporting electrolyte were the same tendency with the effect of initial boron concentration on energy consumption. As a result, using supporting electrolyte decreased energy consumption. In the light of above conclusion, in order to obtain to optimum energy consumption per unit pollutant removal at optimum pH, process must have operated under the conditions of the lower current density, the higher temperature, the higher boron concentration and the higher supporting electrolyte concentration.

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