



Electrocoagulation: a new approach for the removal of boron containing wastes

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ABSTRACT

Using electrocoagulation (EC) as a new method to treat boron containing effluent/wastes has been studied. With combined mechanisms of adsorption and precipitation, the EC is very effective to remove boron; 82% of boron from model waters ($[B]_0 = 250\text{--}500\text{ mgL}^{-1}$) can be removed at a current density of 62.1 Am^{-2} (equivalent to 3.3 as molar ratio of Al:B). In the treatment of industrial effluents, the EC can effectively remove boron and arsenic simultaneously, the [As] was reduced from 15 to $<0.1\text{ mgL}^{-1}$. A multistage EC configuration was even more effective and the boron removal percentage was $>99.9\%$, i.e., after the fifth stage EC treatment, boron concentration can be decreased from 500 mgL^{-1} to less than 0.5 mgL^{-1} . Chemical adsorption with freshly formed $\text{Al}(\text{OH})_3$ flocs played a dominant role in the removal of boron from the wastes. Pre-pH adjustment was not necessary with the EC in the treating of low pH wastes, whilst all other technologies need to raise the solution pH to neutral status.

Keywords: Adsorption; Boron removal; Electrocoagulation (EC); Precipitation; Wastewater treatment

1. Introduction

Boron is a naturally occurring element that is widely distributed at low concentrations in the environment. Boron always occurs in nature bound to oxygen in the form of borates. Borate deposits are rare, being found in dry regions of the world such as in the USA, Turkey, Argentine, China, Russia and Chile. They are, however, extensively used by industry in the manufacture of glass wool, ceramics, borosilicate glass, flame retardants, detergents, wood preservatives, anti-freeze, micro-nutrient fertilisers etc.

In animals, boric acid and the simple inorganic borates are generally of low toxicity, but high boron doses cause reproductive and developmental effects in several species (rats, mice, rabbits) [1,2]. These equivalent doses would not be encountered, by humans, under any circumstances

due to the physical properties of boric acid and to the limited absorption by non-oral routes, except under conditions of serious abuse by deliberate ingestion. In addition, in humans other toxic effects such as vomiting and diarrhoea are produced which will limit the oral intake by humans. Studies in highly exposed humans indicated that such effects do not occur in humans under the conditions of normal use [3,4].

Based on the animal reproductive effects, various environmental regulation organisations have set up standards or guidelines to regulate the boron concentration in drinking water. In the revised European Community Drinking Water Directive [5], boron concentration should be less than 1.0 mgL^{-1} . The recent updated World Health Organisation (WHO) guidelines [6] for drinking water quality retain the recommended guideline value of boron at 0.5 mgL^{-1} . In 2005, the US Environmental Protection Agency (USEPA) published its second version of the Contaminant Candidate List [7] where boron is included and

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its concentration is recommended not to exceed to 1 mgL^{-1} .

The electrocoagulation (EC) process has been recently reviewed again for water treatment [8,9]. The process involves an electrolytic reactor with aluminium (or iron) electrodes and a separation tank. The water to be treated passes through the reactor and is subject to coagulation/flotation, by Al or Fe ions dissolved from the electrodes, the resulting flocs floating after being captured by hydrogen gas bubbles generated at cathode surfaces. In the process, the metal anode dissolution is accompanied by hydrogen gas evolution at cathodes, the bubbles capturing and floating the suspended solids formed and thus removing contaminants.

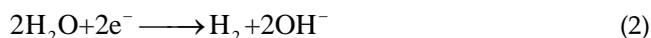
In the EC processes, electrolytic dissolution of anodes (take Al anodes as an example) in water produces aqueous Al(III) species:

- Anodes:

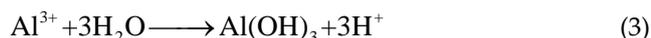


and water decomposition at Al cathodes produces hydrogen bubbles:

- Cathodes:



that float the flocs formed between water contaminants and a range of coagulant species and metal hydroxides formed by hydrolysis:



The amount of Al dissolution can be calculated in terms of Faraday's Law:

$$W = \frac{M_{\text{Al}} I t}{nF} \Phi_e \quad (4)$$

where W is the amount of aluminium to be dissolved from the electrolysis (as gram of Al), M_{Al} ($= 27.0 \text{ gmol}^{-1}$) is the molar mass of aluminium, I is the current used for the electrolysis (ampere), t is the electrolysis time (s), n ($= 3$ for reaction 1) is the charge number of the reaction, F the Faraday constant (96,500 coulombs per equivalent of chemicals produced), and Φ_e the current efficiency for production of dissolved Al(III) species.

There are a range of technologies could be used for removing boron from wastewaters, and these technologies together with their advantages and disadvantages can be seen in Table 1. Selection of these processes should be in terms of the treatment efficiency and operating cost,

which is to be compared in conjunction with the EC reported in this paper; and this can be seen in Section 3.5 later. This paper aims to explore the use of EC as an alternative technique to remove boron from both model boron containing solution and a real industrial effluent and to investigate the corresponding removal mechanisms with EC.

2. Materials and methods

2.1. EC reactor and treatment system

The reactor was configured with four electrodes connected in bipolar series via water, giving three anodes and three cathodes. The materials used for both anodes and cathodes were commercial Al sheets with aluminium content of greater than 99% (Rudgwick Metals, UK). The effective size for one electrode was 175 mm in depth and 46 mm in width, giving the total electrode surface area of 241.5 cm^2 . Aluminium plates were submerging in an acid tank with 3.7% hydrochloric acid for 30 min, and then rinsed by de-ionized water and dried.

The EC treatment system can be seen in Fig. 1, which consists of a reactor, a DC power supply (AP500/7030, HiTek Power, UK), a raw water reservoir, a pump, a discharge receiver, and a multimeter (IDM 93N, ISO-Tech, UK). The connection of the multimeter between the reactor and power supply is to monitor the constant current applied to the experiment.

2.2. Model test solutions and the industrial effluent

A test solution was prepared by dissolving a given amount of boric acid (Aldrich, UK) in 1 L of 0.26 gL^{-1} (4.44 mM) sodium chloride solution (prepared using de-ionised water with GR grade NaCl, Aldrich, UK) and making the boron concentration to be either 250 or 500 mgL^{-1} . The pH of the test solution ranged between 8 and 8.3, and the conductivity of it was between 755 and $767 \text{ }\mu\text{Scm}^{-1}$.

2.3. Operating conditions for various treatment trials

The current applied was 250, 500, 750 and 1000 mA, and the electrolysis time was 20, 40, 60 and 80 min, respectively, in order to maintain a constant molar ratio of Al:B. Solution pH was adjusted before the tests if it was necessary. After the electrolysis, the sample was allowed to stand for 1 h. For the purpose of the comparison, the conventional aluminium coagulation was conducted. A jar test procedure was used, including a 1 min fast mixing at 275 rpm, a 20 min slow mixing at 35 rpm and an 1 h settling. Then the supernatant was withdrawn and filtered using a vacuum pump by a Walkmen G40 filter paper for

Table 1
Advantage and disadvantage of various treatment technologies of removing boron

	Advantage	Disadvantage	References
Chemical lime precipitation, and sedimentation	The lime precipitation could remove boron from greater than 1000 to about 400 mg L ⁻¹	Will produce a voluminous amount of sludge for disposal. Chemical costs could be prohibitively high	[10]
Adsorption on metal hydroxides and activated carbon	Boron adsorption on activated charcoal is more efficient than aluminium hydroxide.	The activated carbon is expensive and also needs to be modified before the use. The adsorption capacity of metal hydroxide is not high	[11,12]
Ion-exchange resin	Amberlite IRA 743 offers very good selected ion exchange efficiency for boron	High capital cost to set up the system and running cost to treat the regeneration effluents	[13–16]
Reverse osmosis and electro dialysis	Effective in removing boron from seawater for drinking water purpose but require multiple stages system or raising influent pH >11 to achieve the effluent [B] <0.5 mg L ⁻¹	Produce brine water waste. Require pressures of 150–400 psi for multiple stages RO and thus high-energy costs. Raising pH >11 results in the membrane scale /fouling problems. Costly maintenance, replacement of membranes and the need of full pre-treatment process contribute significantly to total production cost	[17–22]

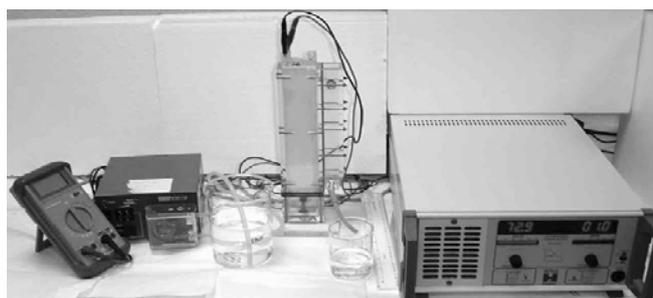


Fig. 1. Electrocoagulation bench test system.

measuring boron concentration and by a 0.45 µm filter paper for measuring Al concentration. The filtrates were collected for the analysis of concentrations of boron and aluminium. Experiments for each condition were repeated. Concentrations and percentage removals of boron were mean values of each experiment.

2.4. Multistage EC study

Model water with boron concentration of 500 mgL⁻¹ was used as test water; 1 M sodium hydroxide was used to adjust the initial pH to 7. Applied current was 500 mA and electrolysis time was 15 min. Treated water was centrifuged by a speed of 3000 rpm for 5 min in order to gain the supernatant for the analysis of boron concentration. Adequate amount of the effluent from the former stage EC treatment was collected and the pH of which was adjusted to 7 for the next stage use. The numbers of EC stages required were determined by whether or not the residual boron concentrations reached to an equilibrium

status. Finally, boron concentration, conductivity and pH value of the treated effluent from each stage were measured and recorded.

2.5. Analytical approach of water quality

An ion chromatography (IC) (Metrohm, Switzerland) equipped with a boron specific anion column (PRP-X100) was used to determine boron concentrations. A solution with 3.2 mM sodium hydroxide (Aldrich, UK) and 0.5 mM sodium carbonate (Aldrich, UK) (pH = 11.55) was used as an eluent. A calibration curve was prepared using standard solutions of boron in the range of 0.1–4 mgL⁻¹. A colorimetric method at a wavelength of 535 nm [23] was used to determine residual Al concentrations. The deviation of boron and Al measurements was less than ±5%.

3. Results and discussion

3.1. Boron removal with EC

A phenomenon that both anode and cathode generate aluminium ions was observed and this is consistent with a previous study [8]. Due to this, the current efficiency, which is defined as the ratio of real Al consumption to that based on the calculation by Faraday Law, ranged between 1.2 and 2.0. Secondly, for the model test solutions, the pH values slightly increased, from 7 to about 8 after electrolysis. Conductivity values were decreased, especially for the conditions of long electrolysis time or larger current applied. Another phenomenon observed was the variation of working potentials in a given experiment, which is

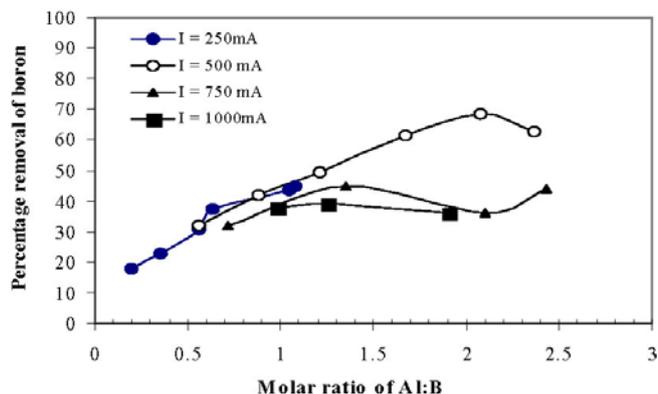


Fig. 2. Effect of molar ratio (Al:B) on the boron removal efficiency. Starting boron concentration = 500 mgL^{-1} , reaction pH = 8.0.

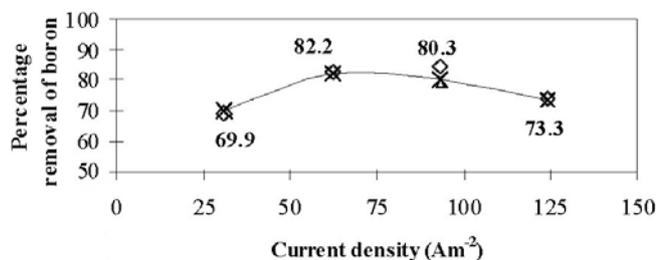


Fig. 3. Effect of the current density on the boron percentage removal. Reaction pH = 8.0–8.4, mole ratio of Al:B = 3.2–3.7. Values shown in the curve were the average percentage boron removal.

related to the properties of test solutions and the electrodes and the electrolysis time.

Fig. 2 presents the results of boron removal from the model test solution ($[\text{B}]_0 = 500 \text{ mgL}^{-1}$) with EC at operating currents of 250, 500, 750 and 1000 mA, respectively. It can be seen that high boron percentage removals were achieved at a current of 500 mA (i.e., current density = 62.1 Am^{-2}), and a boron removal percentage (68.3%) was achieved at the molar ratio of Al:B of 2.1.

Fig. 3 shows the boron removal with Al-EC for treating an industry wastewater ($[\text{B}]_0 = 250 \text{ mgL}^{-1}$). It can be seen that for a given Al:B molar ratio (3.4:1), the greatest boron removal (82.2%) was achieved at an optimum current density, i.e., 62.5 Am^{-2} . High current density did not improve the remove efficiency but resulted in high working potential and therefore, relative high energy consumption.

Table 2 shows that there were various ions existing in the industrial wastewaters, especially the presence of arsenic (As) ions raises the significant hazardous effect on the environment. It is interesting to note that with Al-EC, the removal of boron did not deteriorate the removal of As and all other ions presenting in the industrial wastewater. [As] was reduced significantly, from 15 mg L^{-1} to

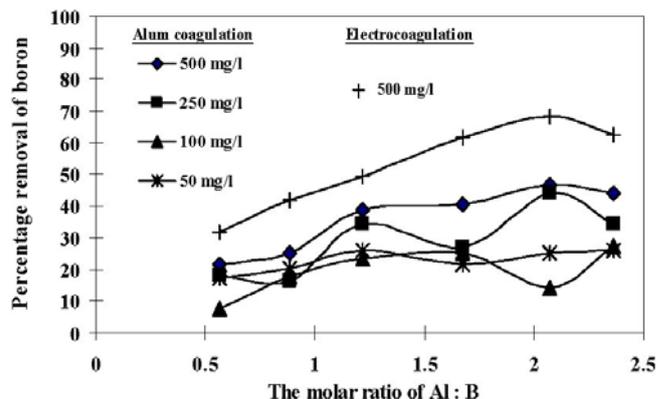


Fig. 4. Boron removal % vs. the dose ratio of Al:B with electrocoagulation and alum coagulation. Coagulation pH = 8. Electrocoagulation was operated under current density of 62.11 Am^{-2} and pH 8.

$<0.1 \text{ mgL}^{-1}$, as well as most other ions, especially of Ca^{2+} , Mg^{2+} , SO_4^{2-} and PO_4^{3-} (Table 2).

Conventional coagulation aims to destabilise the colloidal particles and to promote inter-particles attracting force which results in the formation of large and denser flocs. Then pollutants in the water could be either co-precipitated or adsorbed onto the surface of such flocs and are separated from the water. Whilst the EC process possesses the coagulation function by releasing Al or Fe ions into water from the electrolysis, it has additional two functions, i.e., flotation and oxidation. Hydrogen gas bubbles generated from the cathodes [Eq. (2)] have a bubble size range (30–50 μm), which is suitable for the flotation of flocs and then to help separating flocs from water [8,9]. Also, anodic oxidation during the electrocoagulation was observed when a study was carried out to investigate the As removal with the EC [24], where, As(III) was oxidised to As(V) and then the overall As removal efficiency with the EC was greater than that with coagulation. Therefore, it is not unusually that the EC can remove boron as well as other presenting ions in the wastewater via coagulation/precipitation and adsorption, and this will be discussed in detail later.

The comparative boron removal performance with EC and alum coagulation can be seen in Fig. 4. For a given coagulation pH 8 and a similar dose compared over dose range between 0.5 and 2.5 (as molar ratio of Al:B), the EC can remove between 15 and 20% more boron than alum coagulation. This is very encouraging, not only based on the superior treatment performance but also in terms of the operating cost. A study presented elsewhere [25] demonstrated that the running cost of EC is estimated 6.2 times cheaper than that of alum coagulation for achieving the same boron removal target; the economic advantage of EC in the treatment of wastewaters containing boron is thus significantly.

Table 2
Industry wastewater treated by EC (Al electrodes)

Parameter	B	As ³⁺	Ca ²⁺	Mg ²⁺	Na ⁺	SO ₄ ²⁻	Cl ⁻	PO ₄ ³⁻
Raw industry waste water (mg L ⁻¹)	250.0	15.0	600.0	90.0	132.0	2055.0	35.2	8.5
EC effluent (mg L ⁻¹)	44.5	< 0.1	273.0	48.5	115.2	855.3	21.8	1.1

Table 3
 K_{sp} for Al(OH)₃·nH₂O at 20°C

[B] ₀ (mg/L)	Al/B	[Al ³⁺] (M)	[BO ₂] (M)	[OH ⁻] (M)	K_{sp}
250	3.5	1.61×10 ⁻²⁶	0.058	5.45×10 ⁻⁷	2.76×10 ⁻⁴⁰
50	12	1.39×10 ⁻²⁶	0.273	2.71×10 ⁻⁷	2.78×10 ⁻⁴⁰
10	32	1.48×10 ⁻²⁶	0.115	3.57×10 ⁻⁷	2.18×10 ⁻⁴⁰
10	62	1.42×10 ⁻²⁶	0.215	2.98×10 ⁻⁷	2.71×10 ⁻⁴⁰
Mean					2.61×10 ⁻⁴⁰

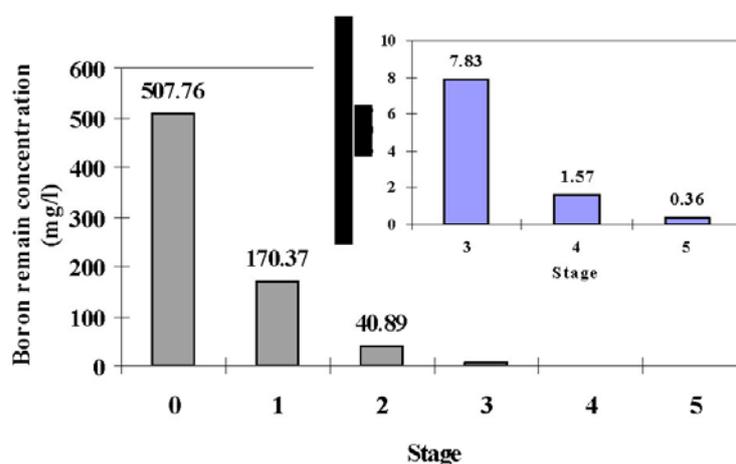


Fig. 5. Boron removal by multistage EC treatment. Al electrodes; current = 500 mA; $t = 15$ min for each stage; $[B]_0 = 500$ mg/L; $pH_0 = 7$; $T = 20^\circ\text{C}$.

3.2. Multistage-EC for treating boron model water

Fig. 5 shows study results of multistage EC in the treatment of a model water with $[B]_0$ of 500 mg/L. The results demonstrate that after two stages treatment, residual boron concentration was reduced to 4089 mg/L and after the fifth stage treatment, boron concentration can be decreased to 0.36 mg/L which was less than the concentration of the WHO recommended for drinking water.

3.3. Mechanisms of boron removal with EC

Al EC has been confirmed to involve dissolution of Al(III) ions from both the anode and cathode with the simultaneous formation of hydrogen gas bubbles at the cathode. In the bulk solutions, the Al(III) ions will react with hydroxide ions to form Al(OH)₃ flocs which have been confirmed to possess positive charge in this study.

The resulting Al(OH)₃ flocs act as an adsorbent as well as a precipitant to adsorb and precipitate borate and therefore, remove boron from water.

Adsorption experiments were performed in order to confirm these assumptions. The freshly formed Al(OH)₃ flocs produced by EC were mixed with the above model water for a given ratio of Al:B and a temperature and for 4 h which was the confirmed equilibrium time of the adsorption. Then the mixture was centrifuged (3000 rpm for 5 min) and the resulting supernatant was withdrawn and the boron concentrations was analysed. The data were used to establish the adsorption isotherms.

Fig. 6 shows the overall boron removal by the EC was 10–20% greater than that by the adsorption with freshly formed Al(OH)₃ flocs. This can be attributed to that in the EC, the precipitation process also occurs, which enhance the overall removal performance. The solubility product constant for the boron-Al precipitants was derived and can be seen in Table 3.

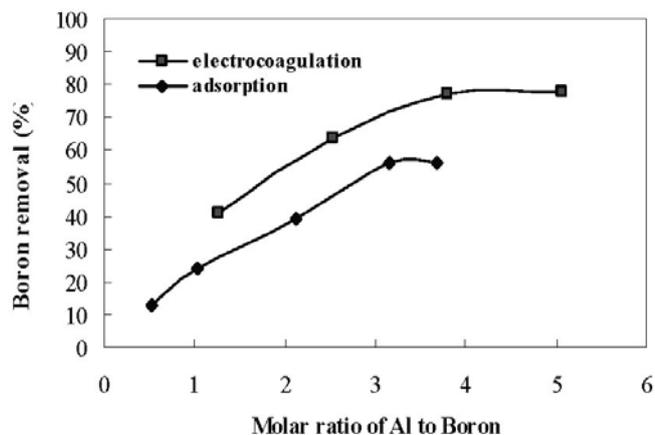


Fig. 6. B removal with electrocoagulation and adsorption. $[B]_0 = 250 \text{ mgL}^{-1}$.

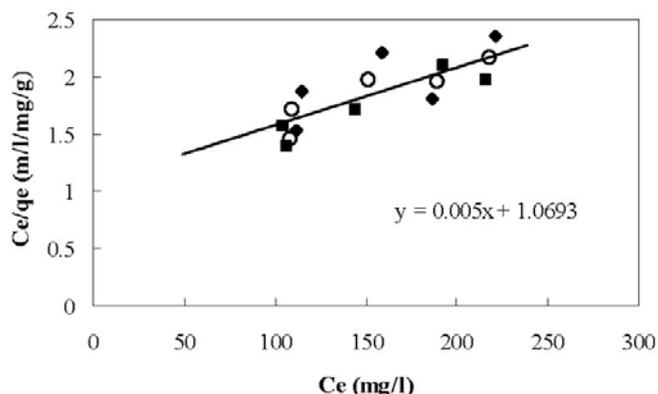


Fig. 7. Langmuir isotherm for boron adsorption. $[B]_0 = 250 \text{ mgL}^{-1}$, $T = 20^\circ\text{C}$.

Table 4
Treatment cost of boron removal

Technology	USD/g boron removed	USD/m ³ water treated	References/note
Chemical lime precipitation and sedimentation	0.285	195 (Raw water with 1000 mg B L^{-1} and was reduced to 315 mg B L^{-1})	[10] Lime unit cost is updated
Adsorption on activated carbon	2.35	169.2 (Raw water with 100 mg B L^{-1} and was reduced to 28 mg B L^{-1})	[11,12] Without including the cost of reactivation
Ion-exchange resin	0.03	0.04–0.06 (Feed water was RO permeate with boron concentration of 1.8 mg L^{-1} which was reduced to 0.3 mg L^{-1})	[15] Capital and operating cost of RO was not included
Reverse osmosis	0.155	$0.7+0.06 = 0.76$ (Two/three stages RO, seawater as feed with boron concentration of 5.3 mg L^{-1} which was reduced to 0.4 mg L^{-1})	[21,26]
Electrocoagulation	0.2	30 (Raw water with 200 mg B L^{-1} and was reduced to 50 mg B L^{-1})	[25]

Fig. 7 shows a Langmuir isotherm [Eq. (5)] for the boron adsorption by the freshly formed $\text{Al}(\text{OH})_3$ flocs produced by the EC at a constant temperature of 20°C .

$$q_e = \frac{C_e}{1.0693 + 0.005C_e} \quad (5)$$

where q_e is the adsorption capacity and C_e is the equilibrium concentration of boron. From Fig. 7, it can be estimated that the maximum adsorption capacity of boron with $\text{Al}(\text{OH})_3$ flocs was $200 \text{ mg B per g of Al}$ for the study conditions. The evidence of that the EC involves both the adsorption and precipitation has been confirmed in the study.

3.4. Effect of solution pH on the boron removal performance with EC

It is well acknowledged that the boron compounds speciation depends on solution pH. At pH less than 4, the dominant species is boric acid which is difficult dissociated to form borate ions and then little removed from water using all available technologies. However, in this study, we observed that EC can effectively remove boric acid while solution pH 3 without a pre-pH adjustment; the percentage removal of boron under such initial solution pH was as high as 70% for Al-EC (Fig. 8a) and 50% for the Fe-EC (Fig. 8b). This is an extra advantage of using the EC for the remediation of boron containing wastes.

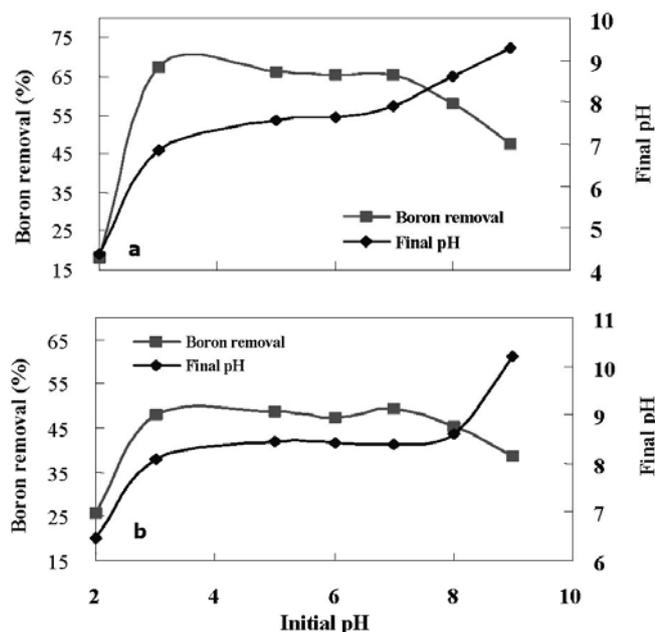


Fig. 8. Effect of initial solution pH on (a) boron removal with Al-electrocoagulation and (b) boron removal with Fe-electrocoagulation. Current = 500 mA; $[B]_0 = 250 \text{ mg/L}$; $t = 10 \text{ min}$, $T = 20^\circ\text{C}$.

3.5. Comparative cost of various technologies in the treatment of boron containing waters

As shown in Table 1, there are a range of technologies that could be used to remove boron from water and wastewaters based on the quality of water to be treated and the targeted residual concentrations in the effluent. The cost comparison of these technologies is complex and cannot be made in a straightforward way. From Table 4, it can be seen that some technologies (e.g., chemical precipitation and electrocoagulation (EC)) could be used to treat wastewaters with high boron concentrations (up to several thousands mg L^{-1}), but some others (e.g., activated carbon,) could only be used to treat waters with low boron concentrations (several tens of mg L^{-1}). Especially, the ion exchange and reverse osmosis (RO) are used in most cases for removing boron from the RO permeate with boron concentrations of either less than or around 5 mg L^{-1} and mainly for the production of drinking water but not for the wastewater treatment. The EC technology reported in this paper, however, can be used to treat either high or low boron concentrations ($10\text{--}1000 \text{ mg L}^{-1}$) and thus it is a versatile technique.

4. Conclusions

This study demonstrates that in the treatment of boron containing wastes with boron concentration of 250--

500 mgL^{-1} , the EC can achieve the maximum boron removal of 82% at a dose of 3.1–3.3 (as molar ratio of Al:B, and for current density of 62.1 Am^{-2}), and can reduce the arsenic concentration from 15 to $<0.1 \text{ mgL}^{-1}$ and other presenting ions (e.g., Ca, Mg and sulphate) in the treatment of an industrial effluent sample. Moreover, multistage EC can reduce boron concentration from 500 mgL^{-1} to less than 0.5 mgL^{-1} . The combined reaction mechanisms of EC in the treatment of boron containing wastewaters are proposed to be adsorption and precipitation (Figs. 6 and 7). In the process, both hydrogen gas bubbles and hydroxide ions are generated [Eq. (2)] and this changes the boric acid to borates which are readily to be adsorbed and precipitated (Fig. 7 and Table 3). Thus, if wastewater pH is low and boric acid is presented, no pre-pH adjustment is required in order to achieve a high boron removal efficiency with the EC, whilst all other technologies are unable to remove boric acid directly.

The advantages of using the EC for the treating of boron containing effluent/wastes have been confirmed by this study but this needs a validation through a pilot- or full-scale trial.

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