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# Pre-treatment of high boron containing wastewater by electro-coagulation

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**ABSTRACT**

The aim of this study is to evaluate the ability of the electro-coagulation (EC) process with aluminum electrodes for removing boron from process and ground waters prior to the reverse osmosis (RO) system. The effect of the operating parameters, such as pH, applied current density, inter-electrode distance (IED), and reaction time, were investigated to achieve a higher removal of boron from water. The obtained results showed that pH was an important parameter affecting removal efficiency. Boron removal efficiency increased with increasing pH from 4 to 10 for process waters (PW) while maximum boron removal has been obtained at pH 7 for groundwater (GW). The highest boron removal efficiency (48%) was achieved at a pH value of 10 within 60 min of treatment time for PW. On the other hand, the maximum boron removal efficiency (30%) was obtained at pH 7 within 90 min of treatment time for GW. In order to evaluate the current density effects on the removal of boron, four different currents (1.5–3.0 A) were applied in experiments. These experiments were conducted at a constant voltage (10 V), a constant IED (1 cm) and an optimized pH value. The maximum boron removal efficiency was obtained at 2 A current density and 1 cm IED for both types of water. The IED was experimented in order to investigate the effect of this parameter on boron removal. The highest boron removal efficiency was obtained at 1 cm IED for both waters. Boron removal efficiency was higher at GW depending on the effluent boron concentration. Boron removal efficiency decreases with increasing boron concentration as, consistent with the literature. It can be concluded that the EC application as pre-treatment prior to RO is effective in reducing the boron concentration.

*Keywords:* Electro-coagulation (EC) process; Aluminum electrode; Boron removal; Operation parameters for EC

## 1. Introduction

Boron is widely scattered in nature found in the form of borates with a very low concentration. Boron is generated from natural sources or anthropogenic contamination to be widely distributed in the environment [1–3]. The amount of boron in surface and groundwater depends on the geochemical structure of the drainage area, and pollution potential of industrial and municipal effluents. Even though boron is an essential micro-nutrient for plants, it also has toxic effects on them when exceeded. Boron is used to increase or prevent the development of plants as the basic nutrient of many plants in variable sizes. Boron deficiency results are limited growth, yield loss and even death of the plant. In recent years, the concentration of boron in the drinking water sources has an increasing tendency due to wide spread applications in various industries, such as cosmetics, glass, ceramics, fertilizers and pharmaceuticals [4]. On the other hand, irrigation water that consists of high boron concentration causes certain environmental problems. Boron compounds passing to soil through irrigation water, surface waters or groundwater form many complexes with heavy metals (such as Pb, Cd, Cu, Ni, etc.), and these complexes are even more toxic than heavy metals forming them [5]. In the last few years, the significant increases in boron compounds concentrations' in surface waters have been observed. The World Health Organization (WHO) recommended limits of 0.3 mg/L and the European Union (UN) defined an upper limit of 1 mg/L boron for drinking water. However, at that time, the best available technologies that were used to treat boron in the water could not achieve a boron content of 0.3 mg/L, thus WHO raised the boron standard to 0.5 mg/L in 1998. Finally, the Drinking Water Quality Committee of WHO revised the boron guideline value in 2011 to 2.4 mg/L based on human health perspective [6,7].

Important boron reserves in the world are located in the United States and Turkey, while the remaining comes from Russia, China and South America. Turkey is known to have approximately 90 million tons of boron reserve that constitutes 70% of the total boron reserves of the world [8]. The boron in groundwater is naturally originated from the structure of the groundwater aquifer. The geochemical structure of the drainage area, and the impurities coming from the agricultural drainage areas, industrial and municipal effluents determine the concentration of boron in fresh waters [9]. Boron, which is in high concentrations in surface waters, originates from boron mines and boric acid production units that are involved in industrial activities. To eliminate the environmental effects of boron in the drinking and using waters, it is necessary to reduce it to the standard values.

The increasing of boron concentration in surface and ground water has triggered the development of effective technologies for boron removal researches. The most important and difficult issue is the selection of effective treatment method due to the amount of boron and chemical composition varying from place to place. The commonly used methods for water treatment such as coagulation, sedimentation and adsorption are not effective on the removal of high boron concentration, besides by biological or chemical treatment only small amounts of boron can be removed [10]. There is not a simple and economical treatment method for the removal of boron from the water. The boron removal from geothermal water by ion exchange resins was studied by Recepoglu and Beker [11], Badruk et al. [12,13], Kabay et al. [14,15], Ipek et al. [16,17] and Koseglu et al. [18]. In the recent years, membrane processes such as Donnan dialysis [19], electrodialysis [20] and reverse osmosis (RO) [21–23] with boron rejections of 91%–96% are used as well.

Along with the widespread boron applications, boron waste increasing day by day pollutes the drinking water sources, and leads to a series of environment and health problems. Although there are different studies in the literature on the effects of the operating parameters, and treatment systems for boron removal from aqueous environments there have not been any recent current studies for removal of boron from waters with >2,000 mg/L boron concentration. It is suitable for the treatment of water to use hybrid systems, or to perform a pre-treatment process for waters containing high concentrations of boron. The aim of this study is to investigate the efficiency of the electro-coagulation (EC) as the pre-treatment process. In this work, the removal efficiency of boron from groundwater (GW) and process water (PW) has been achieved by EC using aluminum as anode and cathode. The effect of the operating parameters, including initial boron concentration, initial pH, current and treatment time on boron removal was investigated thoroughly.

## 2. Experimental method

### 2.1. Characteristics of waters

In this study, the pre-treatment of wastewater with high boron content through EC in order to increase the performance of RO membrane system has been investigated. The analyzed PW and GW were obtained from Kütahya, east side of Turkey. The characterizations of waters are given in Table 1.

### 2.2. EC setup

EC system consisted of an electro-chemical cell with 2,000 mL Plexiglas beaker made up of aluminum plate electrodes (12 cm × 7 cm × 0.3 cm) and DC power supply (GW INSTEK GPD 330S); (0-30 V, 0-3 A), Cole-Palmer, USA that has 0–3 A for current, and 0–30 V for voltage ranges for the run of experiments. A schematic diagram of the EC reactor is shown in Fig. 1. The magnetic stirrer was used at 300 rpm for the homogenization of water. pH and conductivity were monitored regularly by a pH meter (WTW pH 3110), and a conductivity meter (Hach, HQ40D Portable pH and Conductivity Meter, USA), respectively. The chemicals and reagents used during the experiments were of analytical grade.

All experiments were carried out at room temperature (25°C ± 1°C). Aluminum electrodes used as anode and cathode were placed vertically to be immersed in 1,800 mL of water. The effective surface area of each electrode was 140 cm². The EC cell was operated in mono-polar mode. In the optimization experiments of pH and current density, the distance between the anode and cathode (inter-electrode distance [IED]) was fixed at 1 cm. After the batch experiments were completed, the utilized aluminum electrodes were dipped in 6 N NaOH solution for 5 min, and further rinsed with deionized water to be dried for 15 min at 105°C

Table 1
Composition of waters

| Parameters | Process water | Natural water |
|---|---|---|
| Turbidity (NTU) | 2.37 | 9.50 |
| Conductivity (mS/cm) | 5.20 | 7.56 |
| pH | 7.10 | 3.45 |
| Elements (ppm) | | |
| B | 3,443 | 6,024 |
| $Na^+$ | 185 | 145 |
| $Mg^{++}$ | 450 | 750 |
| $NH_4^+$ | <0.125 | <0.125 |
| $SO_4^{--}$ | 2,260 | 4,500 |
| $K^+$ | 7.5 | 41 |
| $Ca^{++}$ | 360 | 550 |
| $Cl^-$ | 125 | 185 |
| $NO_3^-$ | <0.40 | <0.40 |
| $NO_2^-$ | <0.40 | <0.40 |
| $Br^-$ | <0.40 | <0.40 |
| $PO_4^{---}$ | <0.80 | <0.80 |

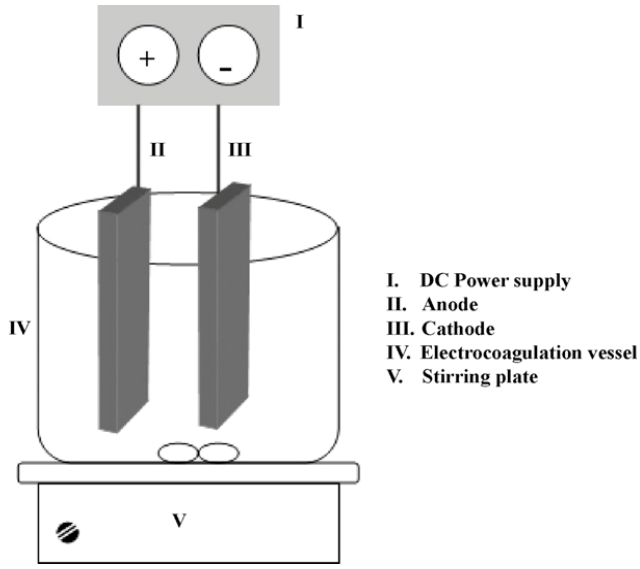


Fig. 1. Electro-coagulation set-up.

for removing all surface impurities and moisture before next usage.

The mechanism of aluminum oxidation during EC process is shown below:

Anode:

$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^- \quad (1)$$

Cathode:

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)} \quad (2)$$

The forming polymeric species, such as $Al_6(OH)^{3+}_{15}$, $Al_{13}O_4(OH)^{7+}_{24}$, during oxidation of the sacrificial Al anode transform into $Al(OH)_3$ as given in equation below:

$$Al^{3+}_{(aq)} + 3H_2O_{(l)} \rightarrow Al(OH)_{3(s)} + 3H^+_{(aq)} \quad (3)$$

The form of $Al(OH)_3$ acts as a sweep floc and provides boron removal from wastewater through increasing its adsorption capacity. The possible mechanism of coagulation by $Al(OH)_3$ is given in Fig. 2.

### 2.3. Experimental procedure

Batch size experiments were operated for two different waters, that is, PW and GW. All the experiments started with feeding the water sample into the cell, then the initial pH was adjusted to the desired value (4, 6, 7, 8 and 10) using 1 N $H_2SO_4$ or NaOH. The supporting electrolyte was not used in the experiments due to the high conductivity of related studying waters. The electrical currency (1.5; 2.0; 2.5; 3.0 A) was applied to the cell where relative distance between the anode and the cathode (IED) (1.0; 1.5; 2.0 cm) was examined. 1,800 mL of samples were collected at 0; 15; 30; 45; 60; 90 and 120 min during the experiments, and pH was measured at the same time, respectively. The residual

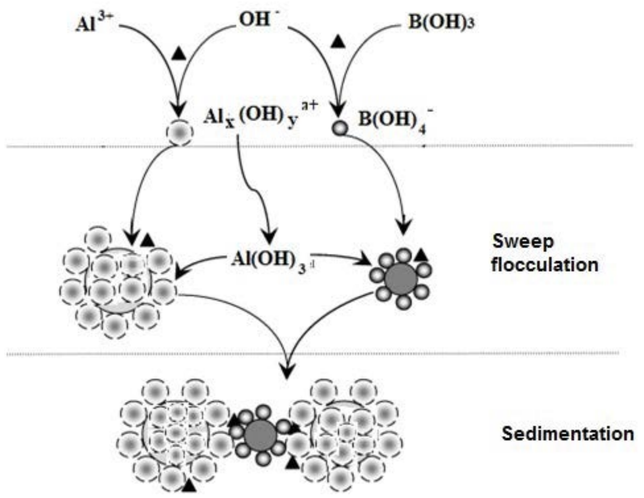


Fig. 2. Boron removal mechanism with $Al(OH)_3$ during electro-coagulation process.

boron concentrations in the samples were filtered in order to remove impurities through a filter paper (Millipore, Nylon Syringe Filter 0.45 µm, Membrane Solutions, USA) of 0.45 µm prior to ICP measurement. The boron measurement was done with Optima 7000 DV Inductively Coupled Plasma – Optical Emission Spectrophotometers (ICP-OES) device supplied by Perkin Elmer, USA. Each sample was analyzed three times, and standard deviations were recorded, respectively. Ion measurements were made by an ICS-3000 ion chromatograph.

## 3. Results and discussion

### 3.1. Effect of pH on boron removal efficiency

The removal efficiency of boron using aluminum electrodes depends on the pH value of the solution [24]. In order to determine the pH effect on boron removal efficiency, five different pH values were studied accordingly. The adjustment of pH was conducted through addition of 1 N NaOH, and 1 N $H_2SO_4$ solutions. The effect of pH on the removal of boron efficiency is shown in Fig. 3.

The graphs present that the pH value had a significant effect on the removal efficiency of boron. The removal efficiency of process waters increased linearly with increasing pH from 4 to 10, and reached a maximum value at pH 10. On the contrary, the removal efficiency of boron in the groundwater increased up to pH 7, and reached a maximum value to be declined after a peak with further increase of the pH. It could be seen that the highest boron removal efficiency (48%) was achieved at pH 10 within 60 min of the treatment time for PW. On the other hand, for GW, the maximum boron removal efficiency (30%) was obtained at pH 7 within 90 min of the treatment time. Some studies reported the highest percentage of boron removal at pH 6–9 [1,25,26]. Literature data are compatible with GW in this study. Even though there is a certain amount of deviation for PW results, it is considered that water chemistry plays an important role. It could be concluded from experimental

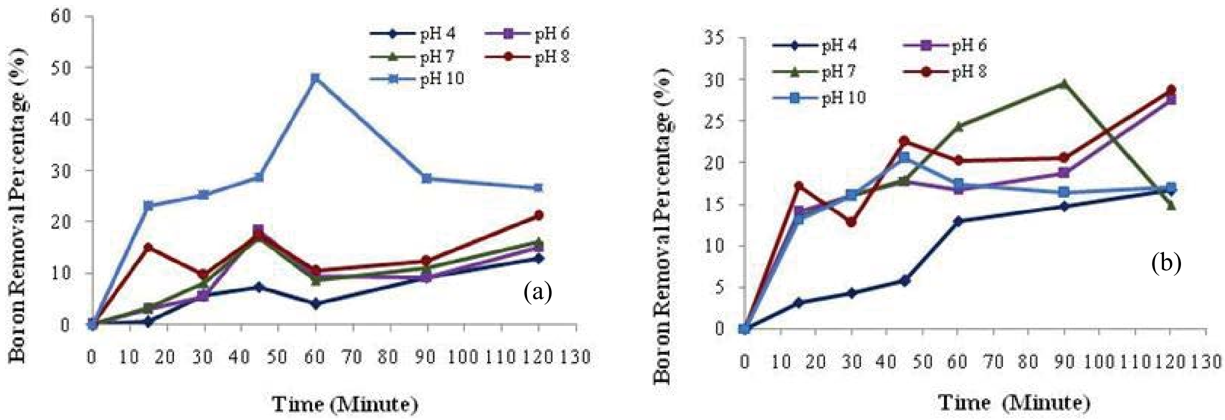


Fig. 3. Effect of pH on the removal of boron from (a) process water and (b) groundwater.

results that the quorum of all coagulant species are formed within the range of $Al(OH)_3$ is formed to be the main complex that is less soluble in water at pH 7–10, and aids in the removal of boron through coagulation.

The removal efficiency of boron from the solution depends on the pH of the solution. Different coagulants are formed in the solution, such as metal ionic species, monomeric hydroxide complexes and polymeric hydroxide complexes depending on the pH solution. The type and amount of these substances, and their distribution in solution are very important as each of them interact differently with contaminants and provide different clotting performances. In addition, the pH value significantly affects the physico-chemical properties of coagulants, such as: (i) solubility of metal hydroxides, (ii) electrical conductivity of metal hydroxides, and (iii) size of colloidal particles of coagulating complexes [27,28]. For this reason, neutral or alkaline solution is preferred for coagulation. The obtained experimental results in this study are in agreement with the literature [1,25,26].

The rate of dissolution of the anode is governed by Faraday's law of electrolysis. According to this law, the rate of anodic dissolution is directly proportional to the amount of current passed through the electrolyte. According to Faraday's law, theoretically 335.5 mg/L aluminum should be released at pH 10, 2 A current density and 60 min operation time. In this case for the determination of the aluminum amount in the water after operation time aluminum measured with ICP for each set of experiments. The ICP measurement results showed that the aluminum amount in treated water was not detectable. This situation probably can be explained by the fact that aluminum is consumed in the removal of other impurities in water.

### 3.2. Effect of current on removal efficiency

Current is a very significant operating parameter for EC that is controlled directly by current density. Mass transferring from electrodes to form coagulant dosage and the formation of bubble for flotation in EC process is determined by current density [29,30]. In order to evaluate the current density effects on the removal of boron, four different currents (1.5; 2.0; 2.5; 3.0 A) was investigated during the experiments. These experiments were conducted at a constant voltage (10 V), a constant inter-electrode distance IED (1 cm), and at an optimized pH value for each types of waters (pH 10 for PW and pH 7 for GW). Fig. 4 illustrates the boron removal efficiency as a function of different current densities within 120 min of the operation time. An increase in boron removal efficiency was observed for all current densities during the experiment period. It can be seen that the maximum boron removal in PW was determined to be 20% in the case of 2 A, while it remained at the level of 10% in other currents. Similar results were observed for GW max. boron removal efficiency was found as 30% for 2 A and 20% for other currents. These experimental results illustrated that 2 A current density is an optimal value on the removal of boron with EC from water with high boron content. The obtained experimental results in this study are coinciding with the literature data [31]. Al-Raad et al. [31] reported that current density values higher than the optimum current density value cause the formation of hydroxyl radicals and these radicals increase the pH value. It is well known that the coagulant breaks down in high pH solutions, which then leads to a reduction in the performance of the EC process. The applied current density could be limited in order to avoid excessive oxygen evolution, as well as to eliminate other adverse effects such as heat generation [31].

### 3.3. Effect of IED

Inter electrode spacing is an important parameter in EC. The distance between the electrodes is another important operational parameter for the required effective surface area in the EC systems. In order to investigate the influence of the IED between electrodes on boron removal, three different distances (1, 1.5 and 2.0 cm) between electrodes were applied. Depending on the inter-electrode spacing, boron removal efficiency is given in Fig. 5.

As seen in Fig. 5, as the distance between the electrodes decreases, the boron removal efficiency has increased. Boron removal efficiency decreased from 16% to 11% and 27% to 10% after 120 min for PW and GW, respectively, when the distance between the electrodes increase from 1 to 2 cm. Experimental data have shown that reducing the distance between the electrodes improves the boron removal efficiency. Because of decreasing the IED between electrodes, result is increasing the rate of aluminum dissolution in solution depending on low resistance of electrodes and enhancing the boron removal efficiency. Besides,

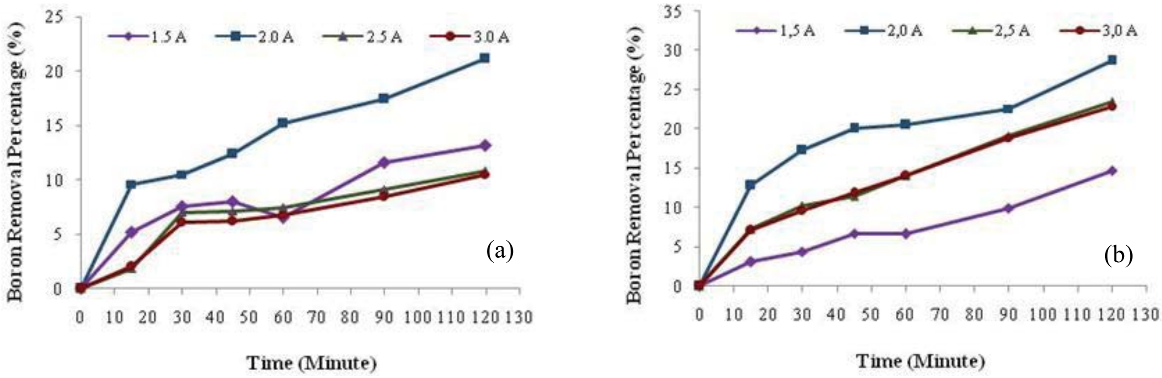


Fig. 4. Effect of current density on the removal of boron from (a) process water and (b) groundwater.

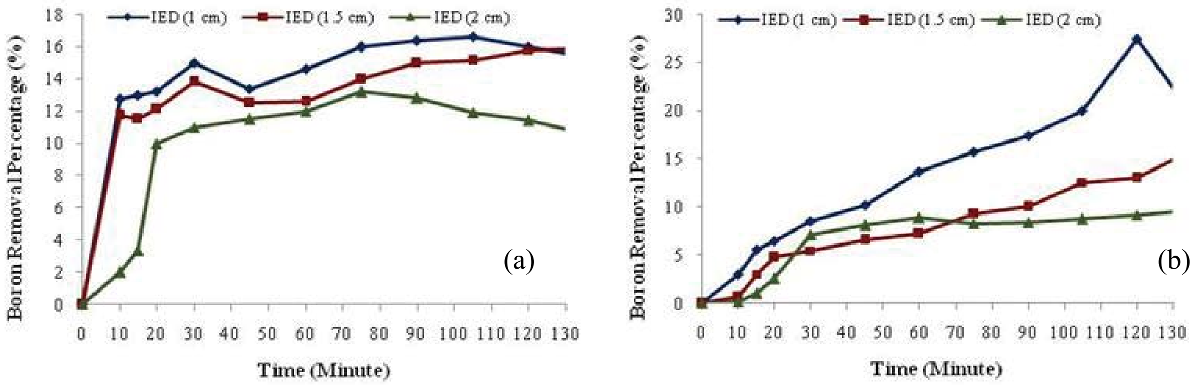


Fig. 5. Effect of IED on the removal of boron from (a) process water and (b) groundwater.

decreasing the IED could enhance the flotation process due to limiting the generated bubbles in a narrow space and results in higher removal efficiencies. An over-abundant distance between anode and cathode decreases notably the formation of flocs. Vasudevan et al. [32] reported that short inter electrode distance decreases electrical energy consumption, decreases resistance of motion depending on the shorter travel pathway and enhance process efficiency. The obtained result in this study is in agreement with data obtained in other works by Zeboudji et al. [33]. The results of this study are summarized in Table 2.

As can be seen from Table 2, the EC process is also capable of preventing clogging in the membranes through removing the silicon. Therefore, the EC as a pre-treatment process on membrane systems can be highly recommended for both increased boron performance and inhibition of clogging in

Table 2
Optimization results of operating conditions

| PW | Boron | | | | Silicon | | | |
|---|---|---|---|---|---|---|---|---|
| | pH 7 | | pH10 | | pH7 | | pH10 | |
| | Initial conc. (mg/L) | Removal (%) | Initial conc. (mg/L) | Removal (%) | Initial conc. (mg/L) | Removal (%) | Initial conc. (mg/L) | Removal (%) |
| | 6,024 | – | 6,024 | – | 113.4 | – | 113.4 | – |
| 30 min | 5,548 | 8.0 | 4,510 | 25.1 | 52.68 | 53.55 | 48.3 | 57.4 |
| 60 min | 4,846 | 19.6 | 4,190 | 30.0 | 48.24 | 57.5 | 28.7 | 74.5 |
| 120 min | 4,759 | 21.0 | 3,132 | 48.0 | 44.47 | 60.78 | 1.51 | 98.66 |
| Groundwater | | | | | | | | |
| | 3,443 | – | 3,443 | – | 24.87 | – | 24.87 | – |
| 30 min | 3,354 | 2.6 | 3,354 | 2.58 | 17.76 | 28.6 | 3.13 | 87.4 |
| 60 min | 2,930 | 14.9 | 2,930 | 14.89 | 12.66 | 49.0 | 2.01 | 91.9 |
| 120 min | 2,427 | 29.5 | 2,497 | 27.50 | 5.41 | 78.0 | 1.26 | 94.7 |

membrane systems. Silicon can interact with aluminum hydroxide to form highly dispersed colloids that are not removed by filtration systems due to preferential adsorption.

## 4. Conclusion

EC experiments were carried out with two different waters (PW and GW) having significantly high boron concentrations. The operating parameters were optimized in order to achieve the best boron removal efficiency. The removal rate was 48% for PW at pH of 10, the treatment time of 60 min, current density of 2 A, and IED of 1 cm. The boron removal rate was found to be approximately 30% for GW at pH of 7. Experimental results have shown that the boron content in waters decreases over time.

The high performance of the EC revealed that it could easily be used as a pre-treatment process for reduction of boron concentration to lower levels before membrane systems are operated. The cartridge filters used prior to membrane systems and/or membrane filters used prior to RO systems (micro-filtration or ultra-filtration) will be sufficient to protect the RO system following EC. The results of this study have clearly showed that the EC process is an operable technique for boron removal even at high concentrations of boron in water.

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