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Original research article

Boron removal from boric acid wastewater by electrocoagulation using aluminum as sacrificial anode

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ABSTRACT

Electrocoagulation (EC) using metallic aluminum as anode and cathode for boron removal from solution was studied. The electrolytic parameters included pH, current density, and initial boron concentration for optimizing the EC process. Experimental results showed that removal efficiency was increased by elevating pH from 4.0 to 8.0, and then decreased at higher pH. The electrolytic efficacy was increased with increasing current density from 1.25 to 5.0 mA cm^{-2}. With respect of energy consumption, 2.5 mA cm^{-2} of current density was acceptable for an effective EC of boron, while increasing boric acid from 10 to 100 ppm-B did not impair removal efficiency. NaCl as a supporting electrolyte promoted more anodic dissolution of aluminum from the electrode surface than that predicted by the Faraday's law. The optimal conditions under which 95% of boron was removed and less than 5 ppm-B remained in the electrolyte would be pH 8, four pairs of electrodes, and 2.5 mA cm^{-2} in 180 min as treating wastewaters containing 10–100 ppm-B. X-ray powder diffractometer and scanning electron microscope were used and results suggested that the irregular crystallites of hydroxide precipitates were composed of bayerite and boehmite phases simultaneously.

1. Introduction

Boron is available in the environment as calcium and sodium borates or as a result of anthropogenic pollution in the form of boric acid and borate salts [1]. Borate salts in solution exists as highly soluble poly-borate ions of high concentration, but it appears as a monomer with low concentration (< 25 ppm) [2]. Boron is widely used in many industries, such as the manufacture of borosilicate glass, detergents, semiconductor, cosmetics, flame retardants, fertilizers, and dyestuff production [3], and is unintentionally discharged into the environment during the manufacturing processes. The deficiency of boron in plants may result in reduced growth, loss of yield, and death [4]. However, raising levels of boron may be harmful to plants. Boron is toxic to human body with some symptoms, include nausea, vomiting, dermatitis, diarrhea, and lethargy [5]. Recently, World Health Organization has regulated an upper limit of 2.4 ppm-B for drinking water [6].

Several techniques have been developed for reducing boron in wastewaters, including ion exchange, adsorption, chemical precipitation, and reverse osmosis [7–11]. Electrocoagulation (EC) generates coagulants through creating metallic hydroxide from the electrochemically soluble anodes [12,13]. It is an efficient way for treating industrial wastewaters containing organic and inorganic pollutants, such as dye, organic matter, nitrate and arsenate [14–17]. EC is outstanding in some aspects as compared to conventional technologies, including high removal efficiency, compact treatment facility, and less sludge produced. This process involves three stages: (i) coagulants formation due to electrolytic oxidation of sacrificial electrodes, (ii) destabilization of the contaminants, and (iii) flocculation of insoluble particles. Meanwhile, hydrogen gas and oxygen are released from the cathode and the anode, respectively. The gas production is beneficial to rise the flocculated particles to the water surface [18,19].

At cathode side:

$$H_2O + e^- = 1/2H_2 + OH^-;\ E^0 = -0.828\ V\ (vs.\ SHE) \quad (1)$$

At the anode:

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$$Al^{3+} + 3e^- = Al; E^0 = -1.662\ V\ (vs.\ SHE) \quad (2)$$

$$Al(OH)_4^- + 3e^- = Al + 4OH^-; E^0 = -2.328\ V\ (vs.\ SHE) \quad (3)$$

$$O_2 + 4H^+ + 4e^- = 2H_2O; E^0 = 1.229\ V\ (vs.\ SHE) \quad (4)$$

Al(III) oxidized from anode would dissolve as several types of hydrolytic species depending upon pH [20]:

$$Al^{3+} + H_2O = AlOH^{2+} + H^+; logK_1 = -5.02 \quad (5)$$

$$2Al^{3+} + 2H_2O = Al_2(OH)_2^{4+} + 2H^+; logK_{22} = -6.3 \quad (6)$$

$$Al^{3+} + 2H_2O = Al(OH)_2^+ + 2H^+; logK_2 = -8.56 \quad (7)$$

$$Al^{3+} + 4H_2O = Al(OH)_4^- + 4H^+; logK_4 = -21.7 \quad (8)$$

$$7Al^{3+} + 17H_2O = Al_7(OH)_{17}^{4+} + 17H^+; logK_{17} = -48.8 \quad (9)$$

$$13Al^{3+} + 34H_2O = Al_{13}(OH)_{34}^{5+} + 34H^+; logK_{34} = -97.4 \quad (10)$$

During EC process, Al(III) speciation undergoes spontaneous formation of hydroxide precipitates which are supposed to effective adsorbents for boron components. The stability diagram of $Al(OH)_{3(s)}$ as a function of pH is as shown in Fig. 1.

$$Al(OH)_{3(s)} + 3H^+ = Al^{3+} + 3H_2O; logK_{sp} = 9 \quad (11)$$

To remove boron efficiently, conventional coagulation and precipitation processes use metal salts, and high quantity is applied at high pH values that normally exceed 12 and high temperature (> 60 °C) [8]. This work aimed to examine the feasibility of EC using aluminum as the sacrificial electrode for boron removal from the boric acid solution at controlled temperature (30 ± 0.3 °C). Parameters investigated included the pH, current density, and boron concentration for optimizing EC. Precipitates collected from the electrolytic cell after treating the boric acid were also characterized using X-ray powder diffractometer (XRD) and scanning electron microscope (SEM). Eventually, the efficacy of conventional chemical coagulation (CC) and EC using the same level of precipitants (i.e., Al hydroxide) was compared in parallel.

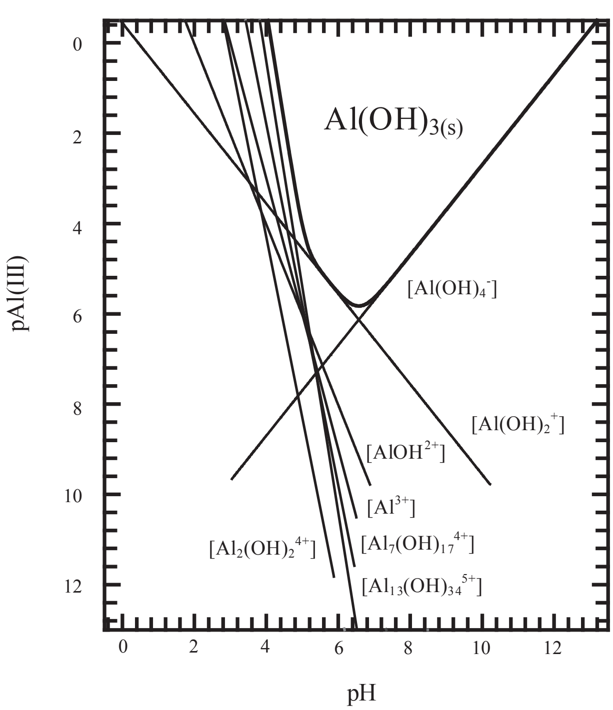

Fig. 1. Solubility diagram of aluminum hydroxide.

2. Materials and methods

2.1. Chemicals

All reagents were of analytical grade, and used without further purification. The synthetic wastewaters were prepared by dissolving the boric acid ($NaBO_3 \cdot 4H_2O$ powder, 97% purity, Panreac, Spain) into waters which were doubly deionized with a laboratory-grade RO-ultrapure water system (resistance > 18.3 MΩ cm). The pH was adjusted using sodium hydroxide (NaOH, Merck KGaA, Germany) and hydrochloric acid (HCl, SHOWA, Japan). The aluminum metal used as the anode and cathode was provided by Ming-Yuh Sci. Ins., Taiwan.

2.2. Experimental procedure

The batch experiment of EC was carried out in an electrolytic cell that was made of Pyrex glass with dimensions of 12 × 10 × 8 cm (WLH). Metallic aluminum as cathode and anode in pairs (10 × 10 cm) were placed at 1 cm interval and connected to a DC power supplier. Fig. 2 depicts the configuration of electrolytic cell. Given concentrations of boron and sodium chloride (NaCl, 99.5%, Showa) as background electrolyte were fed into reactor. All batch solutions were magnetically stirred at 300 rpm. The power supplier in a constant current mode was switched on to carry out experiments, during which the temperature were maintained at 30 ± 0.3 °C using a water bath. At specific time interval, samples withdrawn were filtered by 0.45 um filter, while precipitates were digested using 3 M HNO_3. The solution pH was measured and conditioned to desired value after every sampling. Boron and aluminum elements in filtrates and digests were analyzed by an inductively coupled-plasma emission spectrometer (ICP-OES, JY 2000-2, HORIDA). A SEM (JEOL JSM-6700F, Japan) and an XRD (Rigaku RX III, Japan) were used to determine the surface morphology and the structural phase of aluminum hydroxide, respectively.

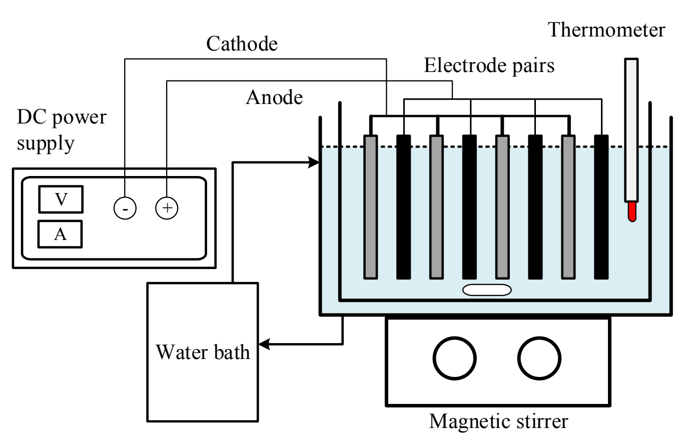

Fig. 2. Apparatus of electrolytic cell for electrocoagulation of boric acid using aluminum as electrodes.