



Bipolar membrane electrodialysis for generation of hydrochloric acid and ammonia from simulated ammonium chloride wastewater



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ARTICLE INFO

Article history:

Received 13 July 2015

Received in revised form

7 November 2015

Accepted 15 November 2015

Available online 22 November 2015

Keywords:

Ammonium chloride wastewater
Bipolar membrane electrodialysis
Influence factors

ABSTRACT

Simulated ammonium chloride wastewater was treated by a lab-scale bipolar membrane electrodialysis for the generation of HCl and $\text{NH}_3 \cdot \text{H}_2\text{O}$ and desalination. The influence of initial concentration of NH_4Cl , current density, salt solution volume, initial concentration of acid and base and membrane stack structure on the yields of HCl and $\text{NH}_3 \cdot \text{H}_2\text{O}$ was investigated. The current efficiency and energy consumption were also examined under different conditions. The results showed that, at the current density of 48 mA/cm^2 , the highest concentration of HCl and $\text{NH}_3 \cdot \text{H}_2\text{O}$ with initial concentration of 110 g/L NH_4Cl was 57.67 g/L and 45.85 g/L , respectively. Higher initial concentration of NH_4Cl was favor to reduce unit energy consumption and increase current efficiency of the BMED system. The membrane stack voltage of BMED increased quickly under constant current when the concentration of NH_4Cl contained in the solution of salt compartment was depleted below the “inflection point concentration” about 8000 mg/L . It means that the concentration of NH_4Cl below 8000 mg/L was no longer suitable for BMED because of higher energy consumption. The HCl and $\text{NH}_3 \cdot \text{H}_2\text{O}$ concentration increased more quickly following the increase of current density. When increasing the volume of NH_4Cl , the concentration of HCl and $\text{NH}_3 \cdot \text{H}_2\text{O}$ also increased. The high initial concentration of acid and base could improve the final concentration of them, while the growth rate was decreased. Compared with the BMED system with three compartments, the growth rate of HCl concentration with the two compartments was higher and its unit energy consumption was lower. It meant that the performance of the BMED system could be improved by optimizing operation conditions. The application feasibility of the generation of HCl and $\text{NH}_3 \cdot \text{H}_2\text{O}$ and desalination of ammonium chloride wastewater by BMED was proved.

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1. Introduction

Due to the fast development of economy and industrialization, industrial effluents from chemical fertilizer and rare earth

processes containing ammonium chloride have been continuously discharged into the environment and resulted in severe problems (Wang et al., 2014a; Huang et al., 2012). Ammonium chloride is one of the crucial sources of ammonia nitrogen and discharging of the ammonium chloride containing wastewater to the fresh water resources could lead to several environmental problems such as eutrophication, excessive propagation of algae and accumulation of chloride ion (Wang et al., 2014a; Li and Qiu, 2013). It was reported that more than 2.4 million m^3 of wastewaters containing ammonia chloride were discharged in 2013 in China. Therefore, ammonia nitrogen pollution has become a serious environmental concern in many newly industrialized countries and the development of effective technologies for removal of ammonia nitrogen is in high demand.

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The technologies of nano-filtration, reverse osmosis and biodegradation have been investigated and tried to apply in the treatment of ammonia chloride effluents (De Gisi et al., 2009; Pérez-González et al., 2012; Jin et al., 2013), but those methods still have some disadvantages. Huang et al., proposed an integrated method in which evaporation crystallization, ammonia stripping and break-point chlorination processes were involved to treat the ammonia chloride containing effluent discharged from rare-earth separation plants with various concentration (Huang et al., 2012). Nevertheless, suffering from high energy consumption, secondary pollution and high capital and process costs, this method was hindered for the wider applications. Some synergistic effects combining biodegradation and physical treatments were also applied to remove the ammonium nitrogen within the wastewater. The studies revealed that by incorporating anaerobic, anoxic and oxic processes with nano-filtration and reverse osmosis, 99.8% of ammonium nitrogen within the coking wastewater was removed (Jin et al., 2013). However, the concentrated wastewater after nano-filtration and reverse osmosis processes obtaining higher ammonium and chemical oxygen demand (COD) level requires further treatments. Ion exchange materials, such as weak and strong acid cation exchange resins, natural and synthetic zeolite in packed bed columns were used to concentrate the ammonium contained in municipal wastewater (Malovanyy et al., 2013). However, the concentrated liquid of ammonium also needs to be further treated before discharge. Although a lot of effort was paid to improve the treatment of industrial wastewater with ammonium chloride, the environmental pollution resulted from ammonium chloride effluent and wastewater resources has not been solved completely. Thus more efforts should have been devoted for the treatment of ammonia nitrogen wastewaters.

Being a new type of electrodialysis technology, bipolar membrane electrodialysis (BMED) has received intensive research attention due to its high efficiency to produce acid and base from the neutral salt under the direct electric field, in which the bipolar membrane with the interface layer is able to split water into H^+ and OH^- ions under the direct electric field (Mafé et al., 1998). Fruitful researches have been conducted using this BMED technology for the production of organic acids (Bailly, 2002; Quoc et al., 2011; Lameloise and Lewandowski, 2012). It was reported that succinic acid could be prepared using BMED with high current efficiency over 90% and low unit energy consumption below 2.3 kWh/kg (Fu et al., 2014). Moreover, the production of tartaric acid from BMED method has overcome several disadvantages of the traditional methods such as high labor intensity, low recovery and notorious solid pollution ($CaSO_4$) (Xu, 2002; Zhang et al., 2009). In the recent years, much attention has been paid to the application of BMED technology in the treatment of industrial effluents (Wei et al., 2011; Wang et al., 2014b; Graillon et al., 1996; Rehouma et al., 2013). It was reported that the metal washing step water could be treated by BMED for the regeneration of inorganic acid and base, and the concentration of acid and base was up to 1.76 N and 2.41 N, respectively (Tran et al., 2015). The industrial NaCl stream was treated by BMED technology with the desalination of 99%, and both of HCl and NaOH produced from the industrial saline water could attain the concentration between 1.5 and 2.0 M simultaneously (Ghyselbrecht et al., 2014, 2013). These studies presented that BMED technology had a good application prospect in organic acid

preparation and industrial saline wastewater treatment. However, reports about the treatment of ammonium chloride effluents by BMED technology have not been found up to now.

In this study, simulated ammonium chloride wastewater was treated by a lab-scale BMED for the generations of HCl and $NH_3 \cdot H_2O$ and desalination at the same time. The influence of initial concentration of NH_4Cl , current density, saline wastewater volume, initial concentration of acid and base and membrane stack structure on acid and base generation by BMED was investigated. The current efficiency, energy consumption, acid and base concentrations under different conditions were examined to test the application feasibility of BMED technology in the treatment of simulated ammonium chloride wastewater and the optimized conditions of the system were obtained.

2. Materials and methods

2.1. Reagents and ion exchange membranes

The following reagents were used in the experiments: ammonium chloride (solid, $\geq 99.5\%$), sodium sulfate (0.05 mol/L), hydrochloric acid (0.05 mol/L) and ammonia solution (0.05 mol/L) were used to prepare the initial solutions used in salt, acid and base compartments, respectively. Sodium hydroxide (0.5 mol/L), silver nitrate (0.1 mol/L), methyl orange (IND), phenolphthalein (IND) and potassium chromate (10 g/L) were used in the chemical analyses. All the reagents were produced by Xilong chemical Co. Ltd.

Three ion exchange membranes (anion exchange membranes JAM-10, bipolar membranes BP-001 and cation exchange membrane PE-001) were used in the experiments, in which JAM-10 and BP-001 are provided by Guangya Chemical Co. Ltd, China. PE-001 is provided by Shanghai Xiangfeng Water Treatment Equipment Co. Ltd, China. The main characteristics of ion exchange membranes provided by the manufacturers were presented in Tables 1 and 2.

BMED apparatus used in the experiments was provided by Sanyuanbada Technology Development Co. Ltd, China. The style of bipolar membrane configuration in this study was BM-A-C-BM namely three-compartment system including salt, acid and base compartments shown in Fig. 1(a). It could be seen clearly from Fig. 1(a) that the simulated ammonium chloride wastewater, HCl solution, ammonia solution and electrode rinsing solution were circulated in the three-compartment system driven by four peristaltic pumps, respectively. Each ion exchange membrane had an effective surface area of 50 cm^2 and the membrane triples were 10 in a stack. Electrodes included a titanium electrode coated with ruthenium oxide as the anode and a stainless steel electrode as the cathode. Anions (Cl^-) and cations (NH_4^+) migrated through anion exchange membranes into acid compartments and cation exchange membranes into base compartments under a direct electric field, respectively. Then the Cl^- and NH_4^+ combined with H^+ and OH^- generated from water splitting at the interface of bipolar membrane (BM) to produce HCl and $NH_3 \cdot H_2O$.

The ammonia, as a weak electrolyte generated in base compartment, can lead to the high solution resistance of base compartment to increase energy consumption of the BMED system. Therefore, the BMED system with two-compartment including salt and acid compartments was also employed and shown in Fig. 1(b). The style of bipolar membrane configuration was BM-A-BM, in

Table 1
The main characteristic of mono-polar ion exchange membranes used in the experiments.

Membrane	Thickness (mm)	Water content (%)	Ion exchange capacity (meq/g)	Area resistance (Ωcm^2)	Transport number (%)
Cation exchange membrane (PE-001)	0.3	35	2 ± 0.02	≤ 4.5	> 98
Anion exchange membrane (JAM-10)	0.24–0.26	20–24	1.6–1.7	7–10	≥ 96

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