Original Research Paper

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Removal of Ammonia Nitrogen in Wastewater by Indirect Mechanism Using Electrochemical Method with Platinum Electrode as Anode

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ABSTRACT

In this study, platinum (Pt) electrode was used as an anode to remove ammonia in the wastewater by electrolysis. The batch experiments were carried out with various ammonia concentrations in the synthetic wastewater at ambient temperature with and without chloride ions. The results indicated that the optimal condition was pH = 7.0 and current density of 10 mA/cm^2. The added NaCl concentration had a significant effect on both, the ammonia removal rate and energy consumption. The ammonia removal performance was 99% with initial ammonia concentration of 25 mg/L after 70 min electrolysis at i=20 mA/cm^2; pH 7.0 and 0.03% NaCl. The response functions described the correlation of four variables with NH_3 concentration, and NH_3 removal efficiency was determined by the response surface methodology (RMS), ANOVA tool with $R^2_{NH3-N} = 0.945$ and $R^2_{Eff} = 0.871$.

INTRODUCTION

Ammonia (NH_3) usually appears in the domestic, food industry and landfill leachate wastewater. Although ammonia is a nutrient for plants, it also causes the eutrophication in the surface waters. Moreover, ammonia is an inhibitor to the disinfection of drinking water as well as having an offensive smell and carcinogenic (Li & Liu 2009, Li et al. 2011, Malovanyy 2013). Therefore, nowadays, ammonia discharge standard has become increasingly stringent and ammonia treatment is increasingly concerned.

Ammonia can be removed by the anoxic process, air stripping, ion exchange and electrolysis. Among them, the electrolysis has some advantages as easy operation, good efficiency, friendly with environment, stable process and no secondary waste (Bunce & Bejan 2011, Candido et al. 2011, Kim et al. 2005, Kim et al. 2006, Li & Liu 2009, Li et al. 2009, Ma et al. 2012, Pérez et al. 2012). The ammonia treatment by this method depends on kind of electrode materials, operating conditions and especially, chlorine production (Bogatu et al. 2010, Bunce & Bejan 2011, Candido et al. 2011, De Vooys et al. 2001, Jeong et al. 2009, Kim et al. 2005, Kim et al. 2006, Li & Liu 2009, Li et al. 2009, Lin & Wu 1996, Ntengwel et al. 2010, Pérez et al. 2012).

The chlorine production decreased in the order of Ti/IrO$_2$ > Ti/RuO$_2$ > Ti/Pt–IrO$_2$ > BDD > Pt (Jeong et al. 2009).

In previous studies, the treatment performance and free chlorine production of the electrolytic process with Pt electrode is lower than other materials in both acid and alkali conditions (Kraft 2008, Kim et al. 2005, Candido et al. 2011). However, Pt electrode has the longest life. Lifetime of RuO$_2$, IrO$_2$ or IrO$_2$/RuO$_2$ is shorter than 1 year, meanwhile lifetime of Pt is longer than 7.5 years (Kraft 2008). So, if the performance of ammonia removal by Pt electrode can be improved, Pt will become a suitable material.

The oxidation mechanism of ammonia in the electrolysis with Pt anode is known by direct and indirect way (Bunce & Bejan 2011, De Vooys et al. 2001, Díaz et al. 2011, Kapalka et al. 2010, Kim et al. 2005, Kim et al. 2006, Li & Liu 2009, Marincic & Leitz 1978). In previous researches, they focused on the ammonia removal performance, free chlorine production and effect factors in the direct mechanism. The indirect mechanism is ambiguous.

RSM is well known in assessing the effect of parameters on treatment results (Spliid 2002). Namely, RSM has been successfully applied to different processes for optimization of the experimental design such as electro-oxidation with Ti/IrO$_2$ (Li et al. 2010), TiO$_2^-$ coated oxidation (Danion et al. 2004), electrolytic process using FePMo$_{12}$O$_{40}$ catalyst (Yue et al. 2014), electrochemical treatment of texttile dye wastewater using Fe electrode (Korbahti 2007) or electro-

oxidation of biologically treated landfill leachate (Zhang 2011). However, the application of RSM in electrochemical oxidation of ammonia with Pt electrode has not yet been reported.

Hence, in this study, Pt was used as anode to remove ammonia in a synthetic wastewater with and without chloride ions (indirect/direct mechanism) at acidic, neutral and alkaline conditions. Objectives of this study were: (1) to investigate ammonia removal efficiency by direct and indirect way; (2) to evaluate chlorine production; (3) to predict and explain clearly the indirect mechanism of ammonia removal process; (4) to predict ammonia removal efficiency, ammonia concentration in effluent and interaction effect of operating parameters on the electrolytic process by RSM and ANOVA tool.

MATERIALS AND METHODS

Synthetic Wastewater

The synthetic wastewater was prepared by dissolving $(NH_4)_2SO_4$ into tap water. Sodium chloride and sodium sulphate were added to create chloride ions and control TDS, respectively. Sodium hydroxide and sulphuric acid were used to adjust pH. The properties of synthetic wastewater are given in Table 1.

Electrodes

Platinum (Pt) and stainless steel (SS316) were used as the anode and cathode, respectively. The working geometric surface area of each electrode was $100\, cm^2$ (L × W = 10 cm × 10 cm).

Bench Scale

A polyacrylic rectangular tank (L × W × H = 15 cm × 12 cm × 20 cm) contained 2.5 L of the synthetic wastewater. Pair of Pt-SS316 electrodes was immersed in the electrolytic solution with 1 cm distance between them without separated membrane. A laboratory DC power supply (Sunchang Electronics Co., LTD., South Korea) having voltage in the range of 0-30 V and current in range of 0-20 A, was connected to the electrodes to supply the direct current.

The wastewater was circulated by a 5 litre/min pump and mixed by a 150 rpm magnetic stirrer to increase the uniformity of sample. pH and temperature were continuously observed by pH meter and thermometer, respectively.

Experimental Procedures

Schematic diagram of the set up of experimental process is shown in Fig. 1.

Direct electro-oxidation: The synthetic wastewater samples containing TDS in the range of 300-500 mg/L, 25 mg/L of NH_3 initial concentration, without NaCl were electro-oxidized at different pH values (4.0, 7.0 and 10.0) and 20 mA/cm^2 of current density.

Indirect electro-oxidation: The experiments were carried out with the same conditions as in the direct electro-oxidation but 0.03% NaCl was added into the synthetic wastewater.

During these experiments, the ammonia, nitrate, nitrite concentrations, pH and temperature were measured versus time.

Chlorine production: The chlorine production and pH were determined during experiments without ammonia in the synthetic wastewater sample at different conditions: pH values from 4.0 to 10.0, current density from 5 to 20 mA/cm^2 and NaCl concentration from 0.03% to 1%.

Analytical Methods

The pH, temperature and TDS were measured by pH meter (Denver/UB-10, Germany), OKAYA Handy Thermo (T200, Japan) and conductivity meter (ORION Model 130, Germany), respectively. Total chlorine, free chlorine, ammonia, nitrate and nitrite concentration were analysed by the spectrophotometer (DR 2800, USA). Received data were analysed by using ANOVA tool of Microsoft Excel and software Mode 5.

RESULTS AND DISCUSSION

Direct electro-oxidation: After 80 minutes of electrolysis,

Table 1: Characteristics of synthetic wastewater used in the experiments.

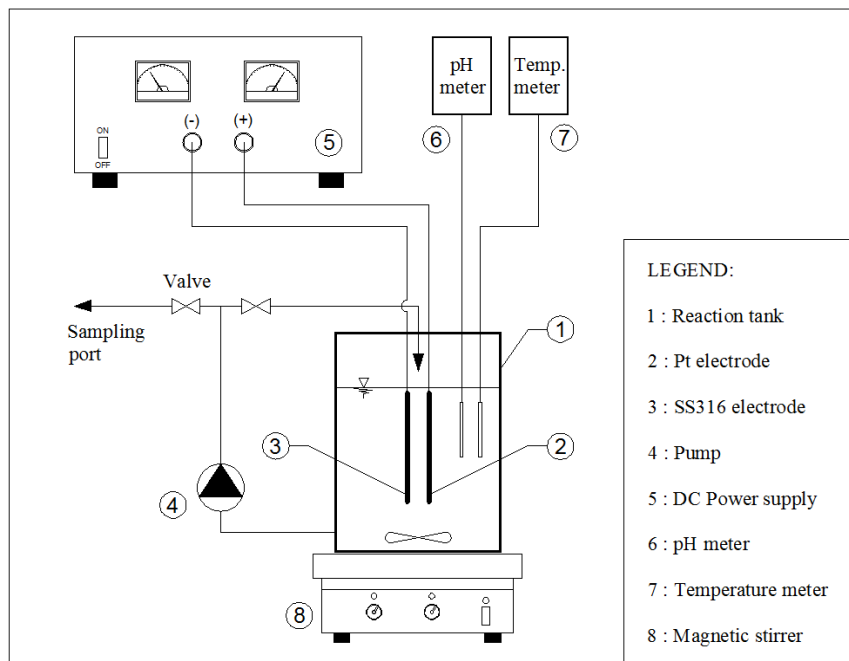| Parameters	Units	Synthetic wastewater
pH		4.0-10.0
Ammonia (NH_3)	mg/L	4-25
Nitrite (NO_2^-)	mg/L	0
Nitrate (NO_3^-)	mg/L	0-1
TDS	mg/L	400-1500
Chloride ion (% added NaCl)	mg/L	0-600 (0-0.1%)
Temperature	°C	20-25

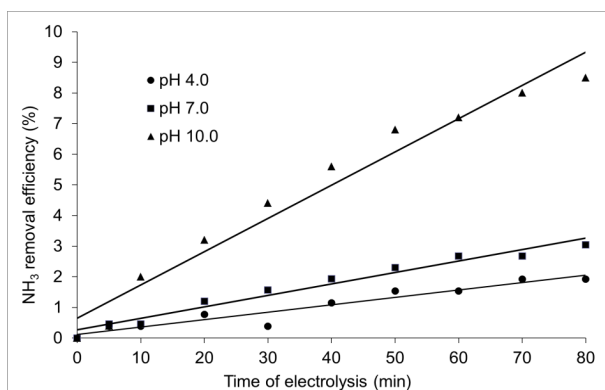Fig. 1: Schematic diagram of the electro-oxidation system for ammonia removal.

Fig. 2: Ammonia removal efficiency by direct way at pH 4.0, 7.0 and 10.0.
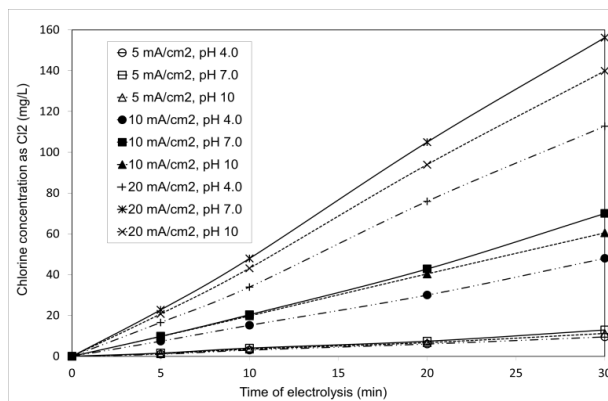

Fig. 3: Variation of chlorine production versus electrolysis time, pH and current density with NaCl concentration of 0.03%.

ammonia removal efficiencies were 8%, 3% and 2% corresponding to the alkaline, neutral and acidic mediums (Fig. 2). These results agreed with previous studies (Kim et al. 2005, Kim et al. 2006). In acidic and neutral medium, NH_4^+ was dominant and not oxidized by direct electro-oxidation. In alkaline medium, NH_3 was dominant, that was removed by direct-oxidation to nitrogen and the air tripping process with many fine bubbles produced in the electrolysis.

Chlorine production: The chlorine production was the main contributor in the ammonia decomposition by indirect oxidation. Result of experiments showed that the chlorine pro-

duction capacity of Pt anode-electrode depends on pH, current density, electrolysis time and chloride concentration. Mass of chlorine production was proportional to these parameters.

Fig. 3 and Fig. 4 illustrate that the chlorine production was highest in the neutral medium. It proportionally increased to current density from 5 to 20 mA/cm² and NaCl concentration from 0.03-0.1%. Especially, the energy consumption reduced when NaCl concentration increased. Additionally, the longer the electrolysis time was, the higher chlorine production was.
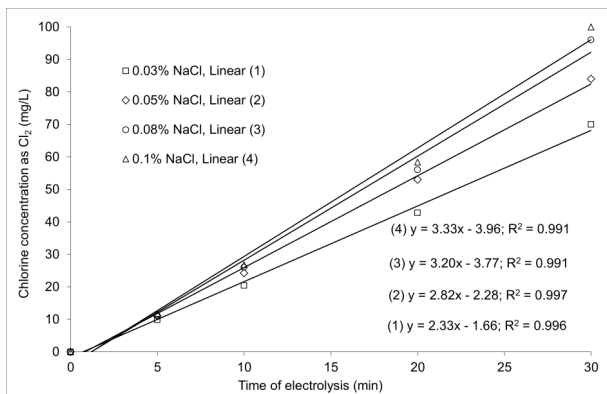
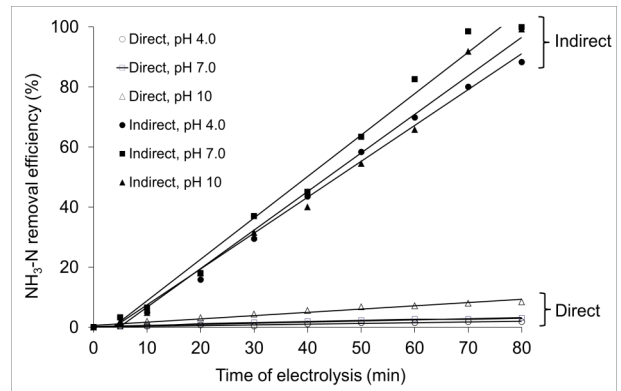Fig. 4: Variation of chlorine production versus NaCl concentration and electrolysis time at pH 7.0 and 10 mA/cm^2.

Fig. 5: Comparison of NH$_3$ removal efficiencies versus electrolysis time between direct and indirect electro-oxidation, at various pH, 25 mg/L of initial NH$_3$ concentration, 20 mA/cm^2 of current density and 400 mg/L of TDS.
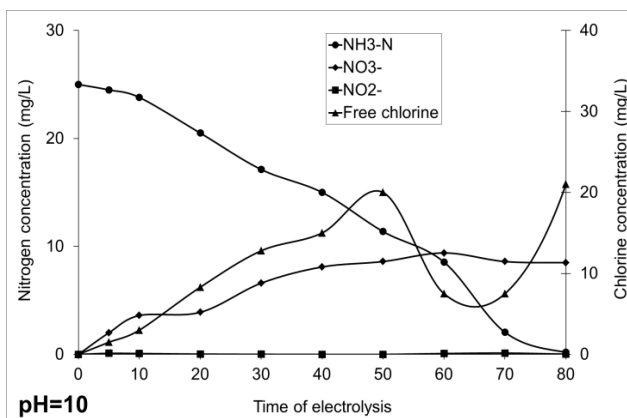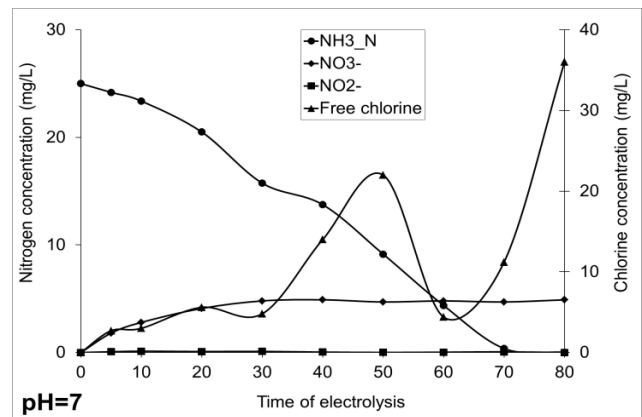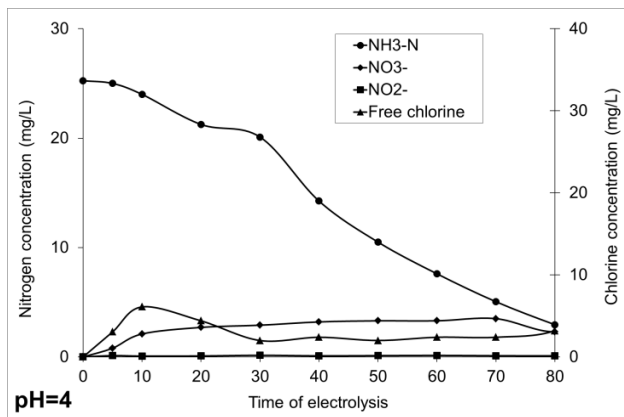

Fig. 6: Comparison of NH$_3$ removal efficiencies between pH=4.0, pH=7.0 and pH=10.
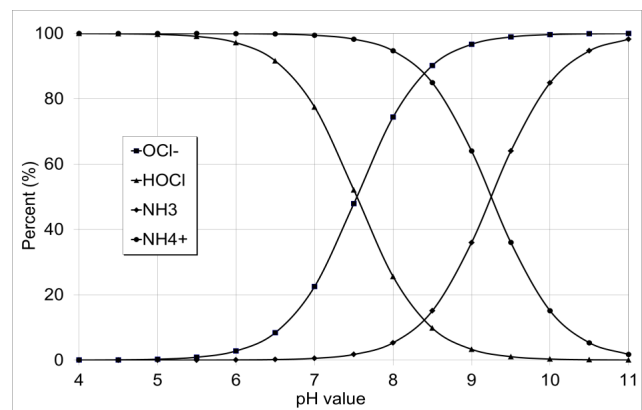
Fig. 7: Distribution of HOCl/OCl$^-$ and NH$_3$/NH$_4^+$ at 25°C as function of pH.

Indirect electro-oxidation: In Fig. 5, the obtained results presented that NH$_3$ removal efficiency by indirect mechanism was significantly higher than the other. The removal performances were 99.9%, 99.5% and 88.3% with respect to

the neutral, alkaline and acidic conditions. These results agreed with Kim et al. (2005, 2006). According to Kim et al. 2006, the conversion yield of ammonia ions to nitrogen increased in the indirect way due to the hypochlorous acid
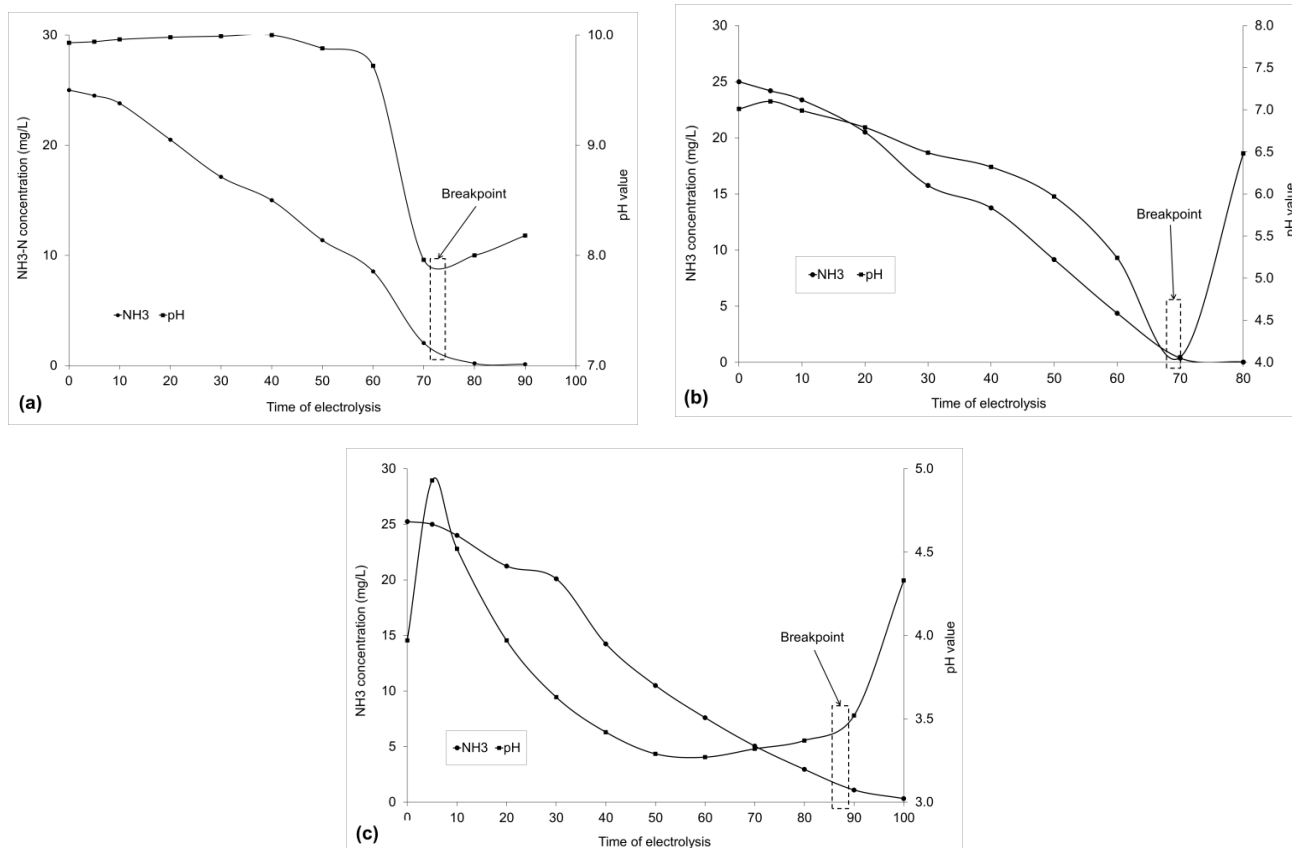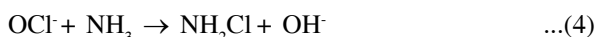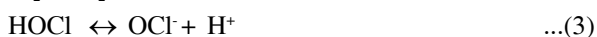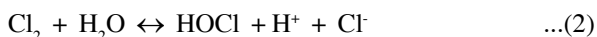
Fig. 8: Variation of pH versus the electrolysis time with 25 mg/L of initial NH_3 concentration, 0.03% NaCl, and 20 mA/cm^2 of current density.

or hypochlorite ion produced by the chlorination of Cl$^-$ at the anode. In the neutral medium, NH_3 removal efficiency was higher than others because the chlorine production was highest (Fig. 6).

The mechanism of ammonia removal by indirect electro-oxidation is different in the neutral, alkaline and acidic mediums. In the alkaline medium, the mechanism of ammonia removal includes adsorption/oxidation (direct way), oxidation by chlorine and air stripping. The predominant forms of ammonia nitrogen and chlorine in the bulk solution at pH 10 are NH_3 and OCl$^-$, respectively (Fig. 7). These equations were restated as:

$$2Cl^- + 2H^+ \rightarrow Cl_2 + H_2 \qquad \qquad ...(1)$$

$$Cl_2 + H_2O \leftrightarrow HOCl + H^+ + Cl^- \qquad ...(2)$$

$$HOCl \leftrightarrow OCl^- + H^+ \qquad \qquad ...(3)$$

$$OCl^- + NH_3 \rightarrow NH_2Cl + OH^- \qquad ...(4)$$

According to Eq. (4), the reaction of chloramine production only released OH$^-$, resulting in increased pH (Karukstis & Van Hecke 2003). Karukstis and Van Hecke indicated
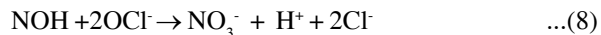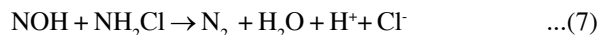
that the reaction between NH_2Cl and OCl$^-$ also occurred to create dichloramline :

$$OCl^- + NH_2Cl \rightarrow NHCl_2 + OH^- \qquad ...(5)$$

With catalyst of OH$^-$ in the alkaline medium, the dichloramine decomposition occurred via the way proposed by Morris & Wei (1969):

$$NHCl_2 + H_2O \rightarrow NOH + 2H^+ + 2Cl^- \qquad ...(6)$$

Followed by the reactions to produce the end products:

$$NOH + NH_2Cl \rightarrow N_2 + H_2O + H^+ + Cl^- \qquad ...(7)$$

$$NOH + 2OCl^- \rightarrow NO_3^- + H^+ + 2Cl^- \qquad ...(8)$$

The mechanism of this process was obviously confirmed through the change of pH versus time (Fig. 8a). In the first 40 min, pH slightly increased due to the release of OH$^-$ from the chloramine production reactions (4), (5) more than the release of H$^+$ from ammonia removal reactions (6), (7), (8). When the reactions of chloramine production were entire, OH$^-$ was no longer created, whereas H$^+$ was continuously created from the ammonia removal reactions. So the pH value quickly reduced during this period. Finally, after the proc-
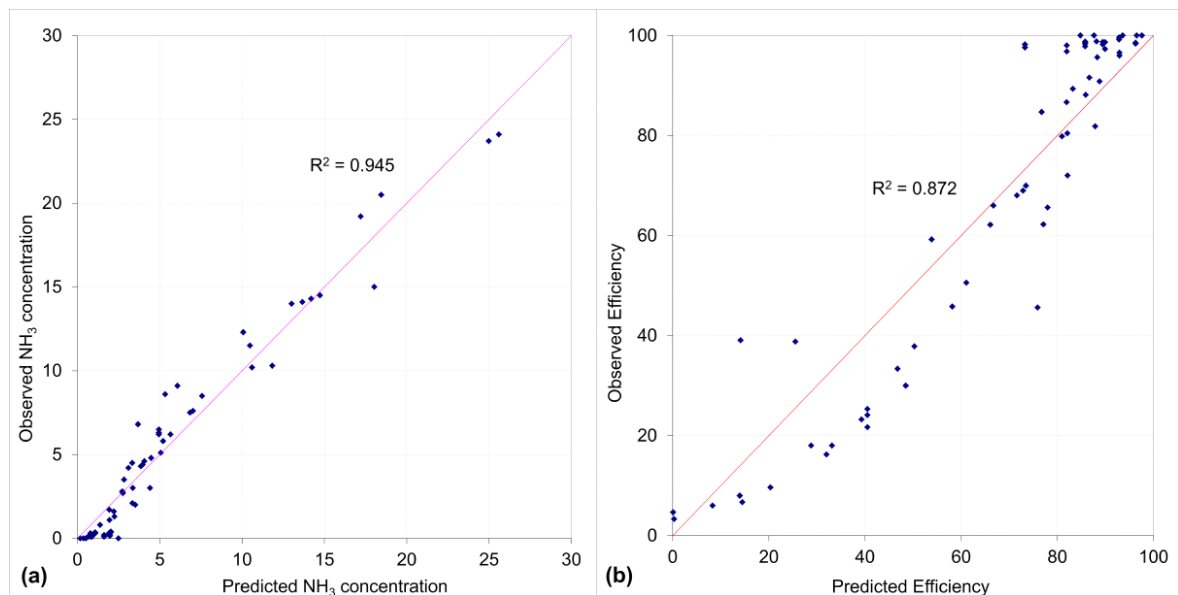
Fig. 9: Predicted versus observed values plot for (a) NH_3 concentration and (b) NH_3 removal efficiency.

ess of ammonia removal or breakpoint was at the end, pH value increased again because there was only the chlorine production process. As a result, the lowest peak of pH would be the breakpoint of electro-oxidation at which ammonia nitrogen was completely removed.

In the neutral medium, oxidation by chlorine was the main mechanism. Air stripping does not occur in this case. NH_4^+ ions were partly decomposed to nitrogen by the OH radicals generated at the electrode surface under the oxygen evolution (Kim et al. 2005). At pH 7.0, NH_4^+ were the predominant species and ratio $HOCl:OCl^-$ was balanced with 1:1. Monochloramine (NH_2Cl) was formed from reactions of HOCl, OCl^- and NH_4^+ as follows (Kim et al. 2005):
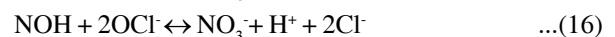
$$OCl^- + NH_4^+ \leftrightarrow NH_2Cl + H_2O \qquad ...(9)$$

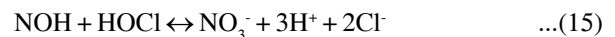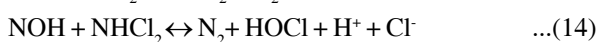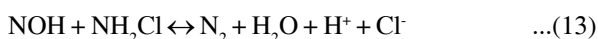$$HOCl + NH_4^+ \leftrightarrow NH_2Cl + H^+ + H_2O \qquad ...(10)$$
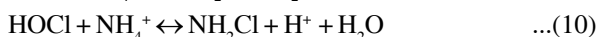
When $Cl_2:NH_3$ weight ratio increased to 3:1, the reaction of NH_2Cl with HOCl was faster than OCl^- due to the oxidation strength of HOCl higher than OCl^-.

$$HOCl + NH_2Cl \leftrightarrow NHCl_2 + H_2O \qquad ...(11)$$
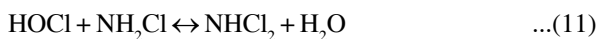
According to Saunier & Selleck (1979), in the neutral medium, the ammonia removal by breakpoint chlorination only occurred following the reactions given below. In this progress, it does not create NCl_3.
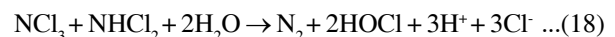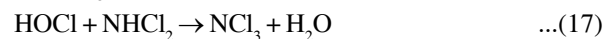
$$NHCl_2 + H_2O \leftrightarrow NOH + 2H^+ + 2Cl^- \qquad ...(12)$$
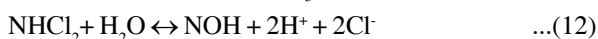
Dichloramine decomposition was followed to the reactions as below:

$$NOH + NH_2Cl \leftrightarrow N_2 + H_2O + H^+ + Cl^- \qquad ...(13)$$

$$NOH + NHCl_2 \leftrightarrow N_2 + HOCl + H^+ + Cl^- \qquad ...(14)$$

$$NOH + HOCl \leftrightarrow NO_3^- + 3H^+ + 2Cl^- \qquad ...(15)$$

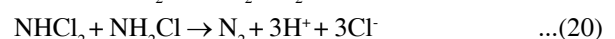$$NOH + 2OCl^- \leftrightarrow NO_3^- + H^+ + 2Cl^- \qquad ...(16)$$

The above mechanisms could be confirmed through the curve of pH change (Fig. 8b). During the ammonia removal reaction, the pH values would reduce because a mole of removed NH_4^+ would release 4 moles of H^+, whereas the electrolysis consumed only 3 moles of H^+. After that, pH values increased and it could be explained with the same way.

In the acidic medium, most ammonium ions were decomposed by free chlorine (indirect way). In this condition, the predominant forms of ammonia and chlorine were NH_4^+ and HOCl, respectively. The reactions follow the equations (1), (2), (10) and (11). Trichloramine was created from reaction between HOCl and $NHCl_2$ (eq. (17)). However, in the absence of free chlorine, NCl_3 was decomposed to HOCl (eq. (18)) or was otherwise lost through oxidation/reduction of NCl_3 (Black & Veatch Corp. 2010):

$$HOCl + NHCl_2 \rightarrow NCl_3 + H_2O \qquad ...(17)$$

$$NCl_3 + NHCl_2 + 2H_2O \rightarrow N_2 + 2HOCl + 3H^+ + 3Cl^- \quad ...(18)$$

Followed by reactions to produce the end products:

$$HOCl + 2NH_2Cl \rightarrow N_2 + H_2O + 3H^+ + 3Cl^- \qquad ...(19)$$

$$NHCl_2 + NH_2Cl \rightarrow N_2 + 3H^+ + 3Cl^- \qquad ...(20)$$

$$NHCl_2 + 2HOCl + H_2O \rightarrow NO_3^- + 5H^+ + 4Cl^- \qquad ...(21)$$

The mechanism of this process could also be proved by the change of pH value and free chlorine over time of electrolysis (Fig. 8c).

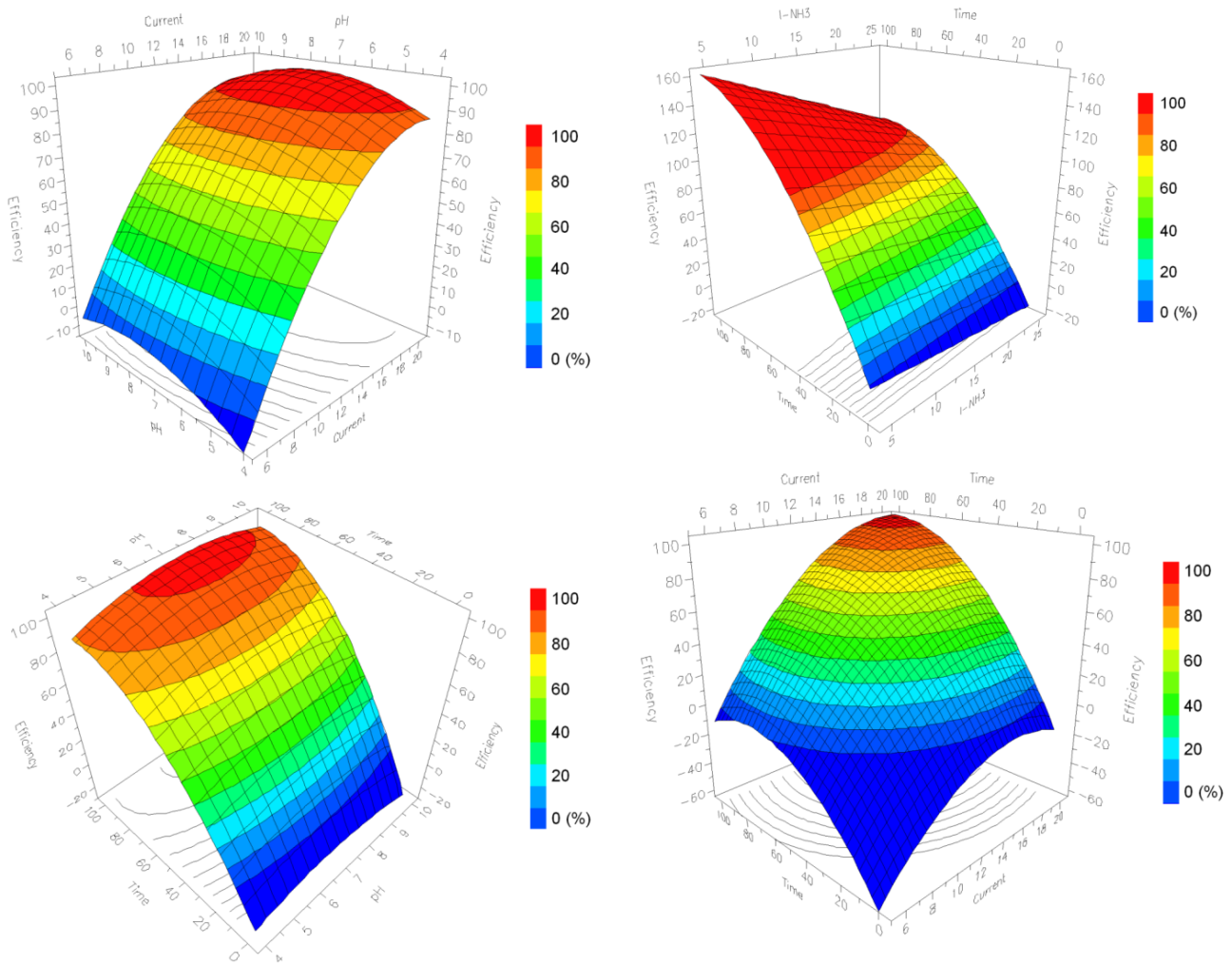Based on these results, we recognized that the pH value

Fig. 10: The response surface plots developed based on the RSM equations.

is an important parameter to monitor the time of breakpoint in the electro-oxidation process of ammonia.

RMS design to predict the ammonia concentration in effluent and removal efficiency: The response surface methodology (RMS) based on the Box-Behnken design was applied to investigate the effect of four independent variables on the response functions as well as predicting results of treatment process from initial parameters. The independent variables were current density (X_1), pH value (X_2), initial NH_3 concentration (X_3) and electrolysis time (X_4). The response functions were NH_3 concentration of effluent (Y_{NH3}) and NH_3 removal efficiency (Y_{Eff}). The coefficients of the response functions for different dependent variables were determined correlating the experimental results with the response functions by using ANOVA tool. The response functions of NH_3 concentration and NH_3 removal efficiency were presented by following equations:

$$Y_{NH3} = 15.446 - 0.959X_1 - 2.203X_2 + 1.060X_3 - 0.125X_4 - 0.003X_1X_2 - 0.05X_1X_3 - 0.006X_1X_4 - 0.009\,X_2X_3 - 0.001\,X_2X_4 - 0.005\,X_3X_4 + 0.052X_1^2 + 0.163X_2^2 + 0.020\,X_3^2 + 0.001\,X_4^2\ (R^2 = 0.945)$$

...(22)

$$Y_{Eff} = -136.882 + 14.368X_1 + 15.913\,X_2 - 2.151X_3 + 2.001X_4 - 0.057X_1X_2 - 0.05X_1X_3 - 0.061X_1X_4 + 0.043\,X_2X_3 + 0.027\,X_2X_4 + 0.021\,X_3X_4 + 0.052X_1^2 + 0.163X_2^2 + 0.020\,X_3^2 + 0.001\,X_4^2\ (R^2 = 0.872)$$

...(23)

In addition, the coefficients of these response functions and the correlation of the experimental results with them were also determined by using Mode 5 regression program (Version 5, 1999) and received similar result with ANOVA tool. These R^2 values illustrate good agreement between the calculated and observed results (Fig. 9). The response

surface plots, developed based on the RSM equations (Eq. 22, 23), are shown in Fig. 10.

CONCLUSIONS

In this study, the ammonia removal efficiency was so high by indirect way with 99% ammonia removed. The mechanism of indirect ammonia electro-oxidation was obviously defined and demonstrated as a process of breakpoint chlorination. The pH, current density and NaCl concentration significantly affected the ammonia removal efficiency. RMS design was proven to be suitable for predicting ammonia removal geometry and evaluating effect of operating variables on the treatment performance.

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