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A novel method for the removal of ammonia in hospital wastewater using flow electrolysis

Riyanto Riyanto[a,*], Muh. Supwatul Hakim[a], Indah Rohmah Wahyu Ningrum[a], Thorikul Huda[b]

[a]*Department of Chemistry, Faculty of Mathematics and Natural Science, Islamic University of Indonesia, Jalan Kaliurang KM 14, 5 Sleman, Yogyakarta, Indonesia 55584, Tel. +6287839295822; emails: riyanto@uii.ac.id (R. Riyanto), supwatul@gmail.com (M. Supwatul Hakim), 20923003@students.uii.ac.id (I.R. Wahyu Ningrum)*
[b]*Diploma Program in Chemical Analysis, Faculty of Mathematics and Natural Science, Islamic University of Indonesia, Jalan Kaliurang KM 14, 5 Sleman, Yogyakarta, Indonesia 55584, email: thorikul.huda@uii.ac.id*

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ABSTRACT

Ammonia in hospital wastewater is a very serious problem. Various methods have been used to treatment of ammonia in hospital wastewater. These various methods have many weaknesses such as high cost, uneconomical, complicated in processing and generating new waste. The flow electrolysis method is an efficient method, low cost, easy to process, does not produce new waste, and does not require advanced methods. Reducing NH_3 concentration in hospital wastewater by flow electrolysis method using carbon electrode has been done. This work was conducted using various electrode circuits (series and parallel), water flow circulation (3, 6, 9, and 12 times), and various voltages (10, 15, 20, and 25 V). Hospital wastewater sample before and after treatment was analyzed using a UV-Vis spectrophotometer at wavelength 640 nm. The results showed that the optimum condition used was a positive-negative parallel circuit at voltage 20 V and 12 times circulation with a degradation concentration of NH_3 (87.50%). The electrolysis flow method can be used to reduce NH_3 concentration in hospital liquid waste effectively.

Keywords: Ammonia; Wastewater; Hospital; Flow electrolysis

1. Introduction

Wastewater from the hospital contains high levels of organic and inorganic compounds, chemical compounds, pathogenic microorganisms that can cause diseases to public health [1]. The most important parameters in wastewater treatment are temperature, pH, total suspended solids, chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammonia (NH_3), and phosphate (PO_4^{3-}). One of the parameters can be used to determine the quality of hospital wastewater. Liquid waste containing NH_3 can affect human health because it has corrosive and irritating properties. Hospital wastewater contains ammonia concentration of up to 0.8 mg/L. Meanwhile, the parameter quality standard set by the government for water quality for ammonia is 0.1 mg/L.

Short-term exposure to NH_3 can cause nausea, vomiting, chest pain, headaches, lung damage, burns if inhaled if contact with the eyes will result in glaucoma and can even lead to blindness when in contact with the skin [2]. Ammonia (NH_3) in large quantities can be toxic and disrupt aesthetics because it is caused by an offensive odor and eutrophication in the surrounding area [3]. NH_3 can be formed through the breakdown of protein in the decomposition of waste so that in the waste storage an unpleasant odor comes from NH_3 [4].

* Corresponding author.

Ammonia is a chemical compound that is toxic to organisms in the water. In general, the ammonia content in wastewater has a high concentration. Most of the ammonia in wastewater is ammonium nitrogen (NH_4^+–N) which is converted into nitrate or nitrite nitrogen through the nitrification process [5]. Hospital wastewater contains a lot of organic compound contaminants, microorganisms, antibiotics, and viruses. Hospital wastewater has high values of BOD, COD, ammonia, and nitrogen [6,7].

Several researchers have carried out various methods of treating liquid waste containing ammonia, namely aeration to convert ammonia to nitrate [8,9]; ion exchange [10]; UV/chlorine process [11]; oxidation [12]; photobioreactor [13]; physicochemical treatment [14]; anaerobically digested [15]; coal-based activated carbons modified by oxidation [16]; co-adsorption onto natural zeolite [17] and electrochemical methods.

Hospital wastewater treatment can be carried out physically, chemically, or biologically. Biological processing can use microorganisms and higher plants. According to research by [18], hospital wastewater treatment using the phytoremediation method with aquatic plants was used to reduce various levels of toxic metals and organic substances. Physico-chemical method of hospital wastewater treatment such as filtering, coagulation, adsorption, sedimentation, neutralization, and electrolysis. More specifically, to reduce the concentration of ammonia in hospital wastewater management, several methods have been implemented, including bioremediation using nitrifying bacteria, zeolite absorption techniques, and the use of waterwheels to increase solvent oxygen in the water. However, this technique is not an effective and high-cost treatment [19].

The electrochemical method is widely chosen as an alternative method in treating ammonia waste because it is more efficient. Several researchers have treated ammonia waste with electrochemistry, namely using Pt electrodes as anode [20]; Pt coated titanium [21]; pulse electrolysis [22]; zeolite-packed electrolysis reactor [23]; various cathodes and Ti/RuO_2-Pt anode [24]; copper [25]; vermiculite packed electrochemical reactor [26]. Several liquid waste processing techniques can minimize the negative impacts so that the waste becomes a more environmentally friendly waste. One of the methods is electrolysis and several studies have conducted research to reduce NH_3 concentration. According to the study of Liu et al. [27] the reduction of ammonia in sample solution using electrolysis method using Ti/RuO_2 and Ti/IrO_2 electrodes.

Based on these problems, it was necessary to have research to reduce the concentration of NH_3 in hospital wastewater. This work using the flow electrolysis method of an inert electrode, because NH_3 can be converted to N_2 and H_2. The advantages of this method were low-cost preparation and higher purity of hydrogen produced [28]. Besides, it does not require chemicals for hospital wastewater treatment and short-time analysis.

Another study according to research by [29] treatment of ammonia waste by electrolysis using titanium electrodes coated with ruthenium and iridium shows that ammonia removal in waste was caused by indirect oxidation of hypochlorite/chlorine compounds in electrolysis. The current efficiency in the study was less than 10% and the ammonia reduction reaction kinetics showed pseudo-first-order. The highest ammonia decomposition was obtained using a current density of 80 mA/cm^2, more than this value will decrease because the adsorption of ammonia on the surface of the electrode will be impeded by hydroxyl ions in solution [30].

In this paper, an innovation on ammonia waste treatment in hospital wastewater has been carried out. Ammonia was chosen as a parameter in this study because ammonia is a toxic compound and is difficult to process in hospital wastewater. The innovation of processing methods with flow electrolysis systems has the advantage that it is easier to process, does not require chemicals, and is efficient. In this study, carbon electrodes are used, because they are inert, stable, durable, and inexpensive.

2. Materials and methods

2.1. Materials

The experiment was carried out using hospital wastewater, carbon electrodes from the dry batteries, ammonium chloride (NH_4Cl) (Merck), phenol (Merck), ethanol 95% (pa) (Merck), sodium nitroprusside (Merck), trisodium citrate (Merck), sodium hydroxide (NaOH) (Merck) and deionized water.

2.2. Instruments

The equipment was used in this research included a series of electrolysis flow for hospital wastewater treatment, scanning electron microscopy-energy-dispersive X-ray (SEM-EDX) from Phenom Desktop ProXL, UV-Visible spectrophotometer (UH5300), a set of glassware.

2.3. Preparation and characterization of electrode

The carbon electrodes used in the study were obtained from waste dry batteries. Carbon rods was obtained from dry batteries waste by opening the zinc metal cover. Carbon rods that have been taken are cleaned of impurities, washed with distilled water, and dried in an oven at 105°C for 3 h. The carbon electrodes have been characterized by SEM-EDX.

2.4. Experimental procedures

2.4.1. Reactor flow electrolysis

Reactor flow electrolysis was made by cutting pipe to the size of 50 cm and then the pipe was perforated to 8 holes with a distance of 5 cm of each. The clean carbon from the dry batteries was used as the electrode. The pipe was connected to liquid hospital wastewater. The experimental setup for hospital waste treatment using flow electrolysis can be seen in Fig. 1.

2.4.2. Optimization of electrolysis variable

Hospital wastewater was electrolyzed in various voltage (10, 15, 20, and 25 V). The solution was taken each time based on circulation flow (3, 6, 9, and 12 times). The potential and circulation effects on ammonia degradation in hospital

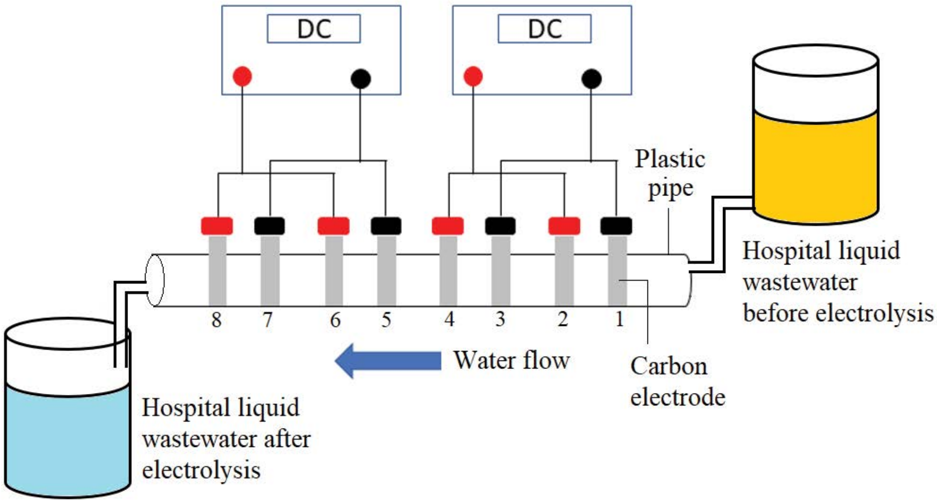

Fig. 1. Scheme of hospital wastewater treatment by flow electrolysis method, red wire (positive) and black wire (negative), Examples: positive-negative parallel (PNP) is the No. 1 electrode starts from black and is alternated with red. Based on the position of the electrodes and the relationship to the power supply, four variables were determined in this study (1) negative-positive parallel (NPP) is the No. 1 electrode starts from red and is alternated with black (2) positive-negative parallel (PNP) is the No. 1 electrode starts from black and is alternated with red (3) positive-negative series (PNS) is electrode No. 1–4 changed to red and the No. 5–8 black color (4) negative-positive series (NPS) is electrode No. 1–4 changed to black and electrode No. 5–8 red color.

wastewater were determined by analysis using a UV-Vis spectrophotometer.

2.4.3. Analysis of ammonia using UV-Visible spectrophotometer

Ammonia in hospital wastewater before and after treatment was determination using APHA method 4500-NH_3 [31]. Ammonia standard solution 10 mg N/L were pipetted 0, 5, 10, 20 and 40 mL and each put into a 100 mL volumetric flask, then distilled water is added to the limit mark so that the ammonia level is 0, 0.5, 1, 2 and 4 mg N/L. The UV-Vis spectrophotometer was optimized according to the instrument instructions for testing ammonia levels. 25 mL of ammonia solution was pipetted and inserted into the Erlenmeyer, then added 1 mL of phenol solution, 1 mL of sodium nitroprusside, and 2.5 mL of oxidizer solution and then homogenized. The Erlenmeyer was covered with plastic or paraffin film. The absorption was measured by UV-Vis spectrophotometer at wavelength 640 nm.

Sample (25 mL) was pipetted and put in 50 mL Erlenmeyer. Then added 1 mL of phenol solution, 1 mL of sodium nitroprusside, and 2.5 mL of oxidizer solution, then homogenized. The Erlenmeyer was covered with plastic or paraffin film. The absorption was measured at wavelength 640 nm by UV-Visible spectrophotometer.

3. Result and discussion

3.1. Characterization of electrode

The carbon contained in the dry batteries was used as an electrode in this work. Carbon electrodes were characterized using SEM-EDX where SEM was used to determine the surface morphology of the carbon electrode and EDX was used to determine the elements contained in the carbon electrode. The shape of the carbon electrode morphology can be seen in Fig. 2. The textural structure (Fig. 2) shows the morphology of the electrode before and after the electrochemical process to hospital wastewater degradation.

Fig. 2. shows the morphology of the carbon electrode with a magnification of 5.000 times (a) before and (b) after treatment. At a magnification of 5.000 times the morphology of the carbon before and after treatment showed that there were many pores, it was clear that the pores they had were very deep on the surface of the carbon. According to research by [14] that carbon has pores of uneven and unclean sizes. This is probably due to several factors, among others, the pores on the carbon were filled with other elements and there were impurities on the surface.

The carbon electrode was characterized using EDX to obtain information on the elemental composition contained in the carbon electrode used in this study. The results of EDX characterization can be seen in Fig. 3 and Table 1.

Fig. 3 shows the spectrum of the carbon electrode, it can be seen that each elemental composition contained in the carbon electrode can be seen in Table 1. Fig. 3 and Table 1 show the carbon electrodes (a) before and (b) after the treatment contain the highest element of carbon (C) about 94.18% and 86.31%, respectively. The element content of the electrode was not 100% pure of carbon but there were still other elements.

3.2. Effect of potential to the amount of circulation using the parallel position of the electrode

The results of treatment hospital wastewater samples with various voltage and circulation using the parallel position of the electrode to decrease the concentration of NH_3 can be seen in Fig. 4. Fig. 4a shows the results of hospital

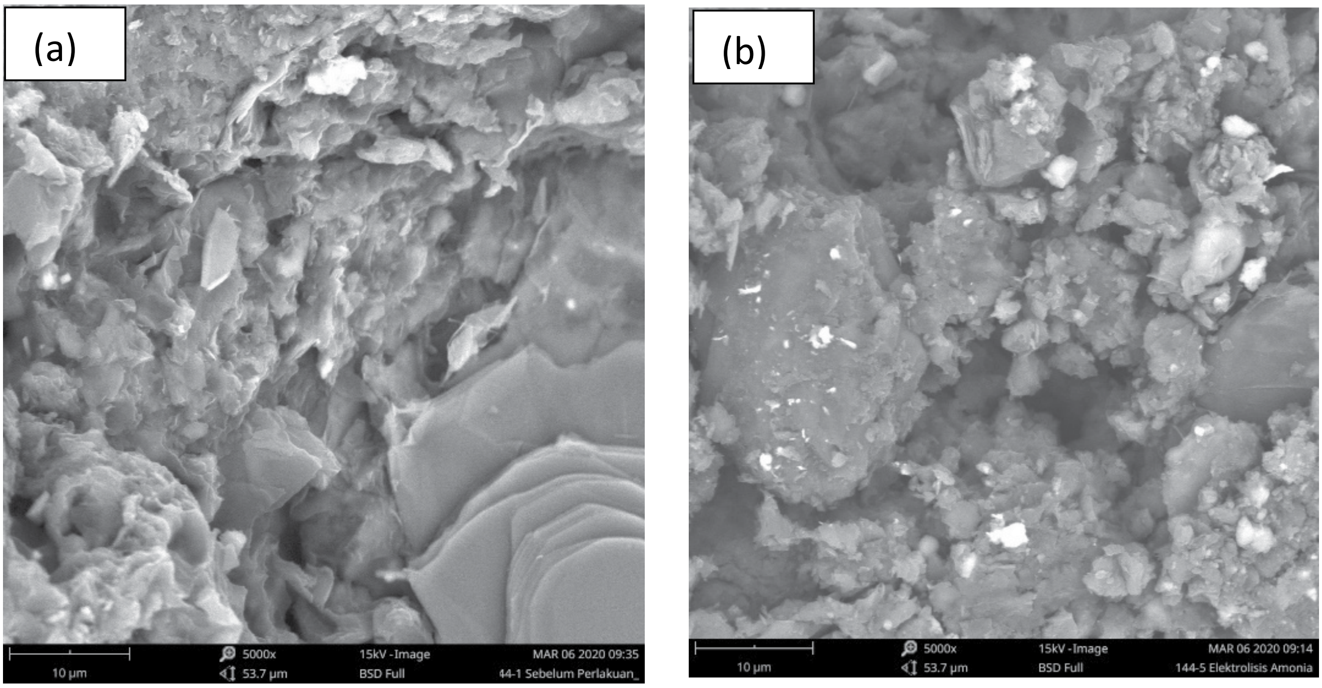

Fig. 2. SEM-EDX image of carbon electrode hospital wastewater electrolysis (a) before and (b) after with 5,000x magnifications.

Table 1
Elemental composition of carbon electrode before and after used for electrolysis of hospital wastewater

Element	Atom number	Weight (%)	
		Before (a)	After (b)
C	6	94.18	86.31
O	26	5.27	7.70
N	7	–	4.90
Si	13	0.27	0.28
S	16	–	0.26
Fe	26	–	0.18
Al	12	0.28	0.16
Ca	20	–	0.16
Mg	12	–	0.05

wastewater treatment using the flow electrolysis method with a negative-positive parallel (NPP) arrangement. The waste has hit the negatively charged electrode (cathode), then the waste has hit the positively charged electrode (anode). In the NPP system, there has been a water reduction reaction to form H_2 gas, then an ammonia oxidation reaction has occurred to form N_2 gas. The final result of the hospital wastewater treatment process with the NPP system is H_2 and N_2 gases. Hospital wastewater treatment with the NPP system resulted in ammonia degraded by an average of 45.88%. The variation in potential and the number of circulations in the NPP system did not show a significant difference in the % of integrated ammonia.

Fig. 4b shows the treatment of hospital wastewater using the positive-negative parallel (PNP) system. In the PNP system, hospital wastewater has hit a positively charged electrode (anode). At this stage, the waste containing ammonia will oxidize to form N_2 gas and in the next stage, the waste will hit the negatively charged electrode (cathode). Waste will be reduced at the cathode to form H_2 gas. The PNP system shows the best results for the degradation of ammonia in hospital waste. The results of electrolysis flow with a potential of 20 V and a total circulation of 12 cycles showed that the best degradation results reached 87.50%. Degradation of 87.50% was caused by oxidation of the waste followed by reduction. The PNP electrode arrangement with a potential of 20 V and a circulating amount of 12 is the best parameter in treating ammonia in hospital waste.

3.3. Effect of potential to the amount of circulation using the series position of the electrode

The results of treatment hospital waste samples with various voltage and water circulation using the series position of the electrode to decrease the concentration of NH_3 can be seen in Fig. 5. Fig. 5a shows the electrode arrangement using the negative-positive series (NPS) system. A series system with an arrangement of four electrodes with a negative charge and the next 4 electrodes with a positive charge. In this system, hospital liquid waste containing ammonia will experience a reduction at 4 electrodes to form H_2 gas and oxidation at 4 electrodes to produce N_2 gas [30,35]. The NPS system produced a percentage of ammonia degradation of 24.56%. Fig. 5b shows the electrode arrangement using the positive-negative series (PNS) system. The PNS system uses an electrode arrangement of 4 positively charged electrodes (anode) and 4 negatively charged electrodes (cathode). The highest

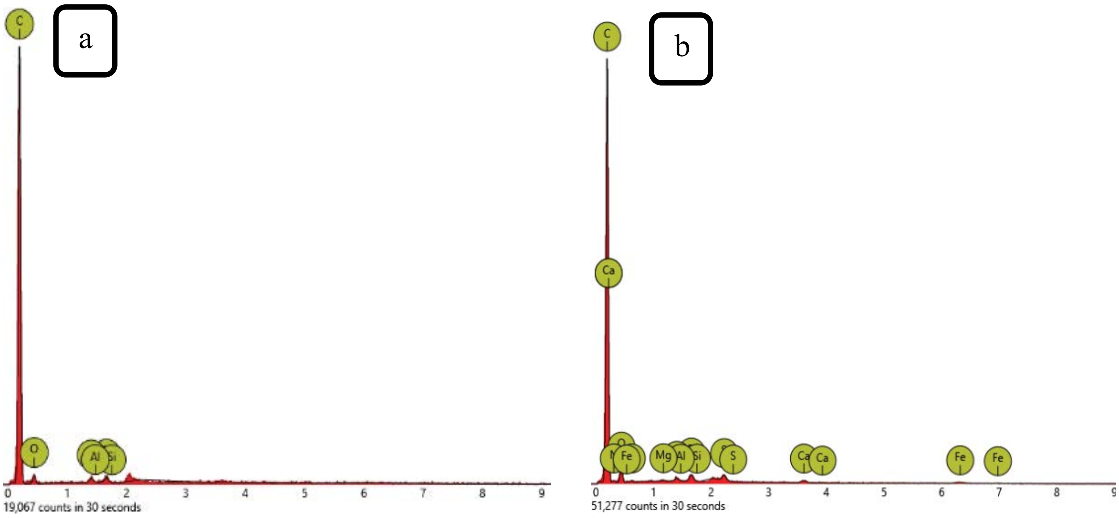

Fig. 3. EDX spectrum of carbon electrode (a) before and (b) after used for flow electrolysis.

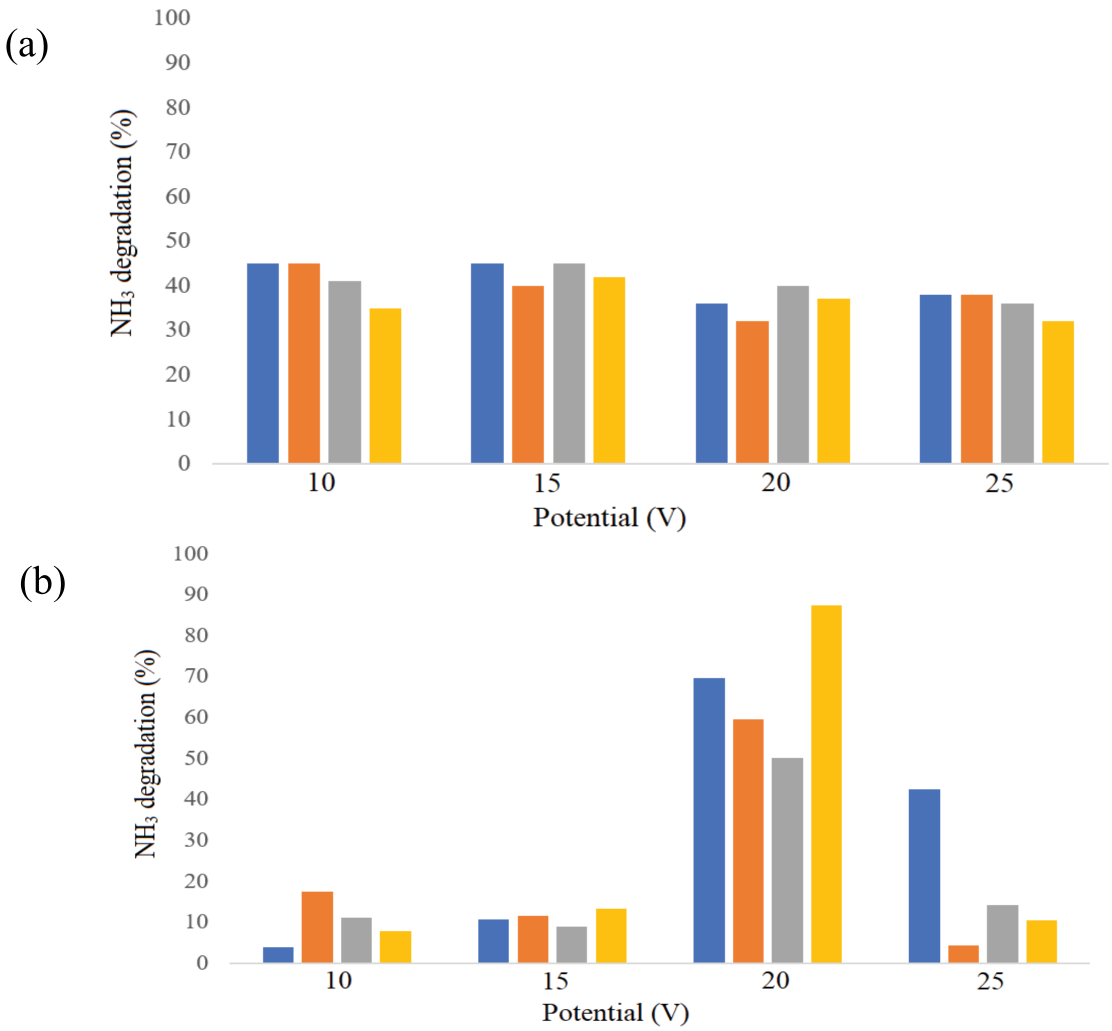

Fig. 4. Effect of potential (a) negative-positive parallel and (b) positive-negative parallel and the amount of circulation to percent degradation of NH_3 in hospital liquid wastewater, where ■ 3, ■ 6, ■ 9, and ■ 12 cycles.

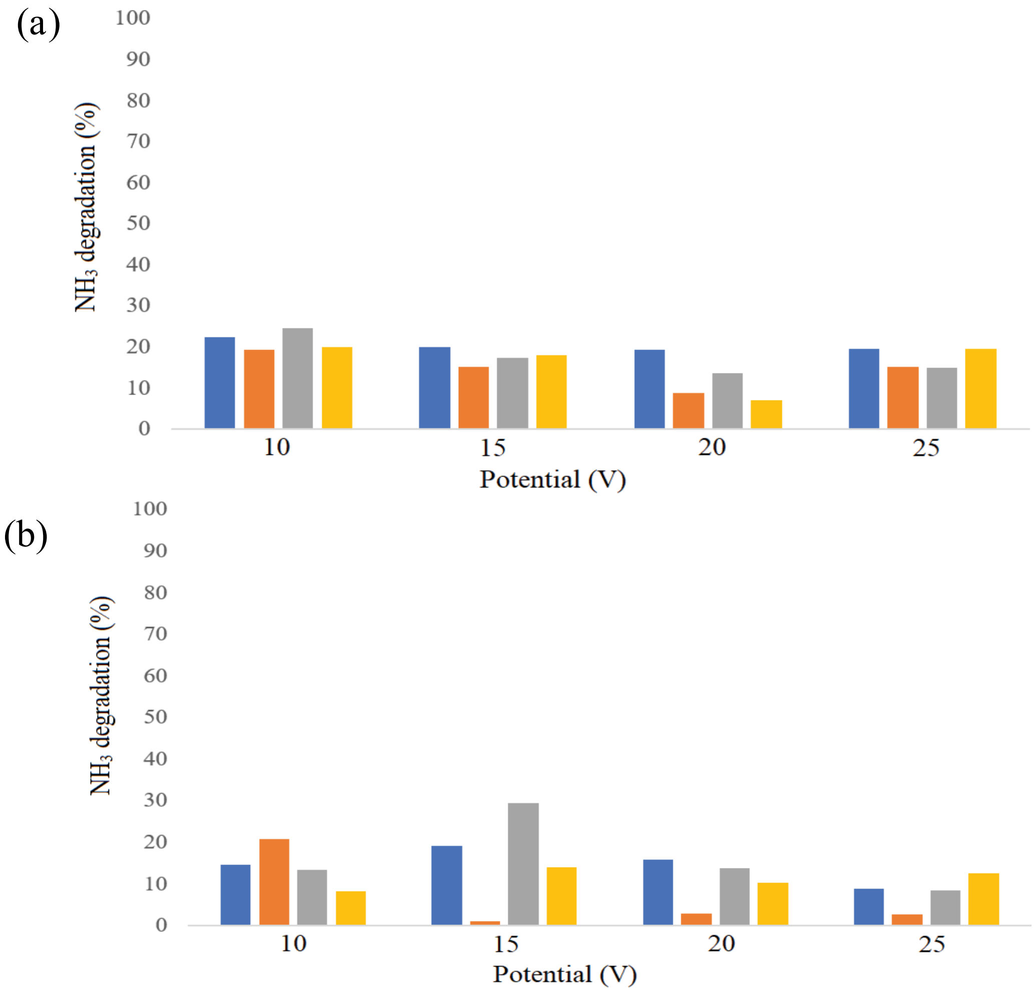

Fig. 5. Effect of potential (a) negative-positive series and (b) positive-negative series and the amount of circulation to percent degradation of NH_3 in hospital liquid wastewater, where ■ 3, ■ 6, ■ 9, and ■ 12 cycles.

ammonia degradation was obtained with the PNS system at a potential of 15 V and the number of circulations of 9 times.

The effect of circuit type, amount of circulation, and optimum potential to %degradation of NH_3 in hospital wastewater are shown in Table 2. The formula for determining degraded ammonia is shown in Eq. (1). Ammonia concentration was initially obtained from the results of ammonia analysis in hospital waste. After the hospital wastewater is treated by the flow electrolysis method, the final ammonia concentration (residue) is analysed using UV-Visible spectrophotometer.

$$\%NH_3\ \text{degradation} = \frac{[NH_3]\text{initial} - [NH_3]\text{residual}}{[NH_3]\text{initial}} \times 100\% \quad (1)$$

The performance of the flow electrolysis method for ammonia degradation in hospital wastewater will be more effective if the waste contains Cl^-. The result of ammonia decomposition will increase with increasing concentration of Cl^- ion [27,30]. The concentration of Cl^- ion has a linear relationship with the rate of degradation of ammonia in the electrolysis process, as well as current density [27]. Ion Cl^- added in the beginning will increase the degradation of ammonia. Ammonia electrolysis mechanism with the addition of Cl^-:

$$2Cl^- \rightarrow Cl_2 + 2e^-\ \text{(Anode)} \quad (2)$$

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^-\ \text{(Chemical solution)} \quad (3)$$

$$HOCl + 2/3NH_3 \rightarrow 1/3N_2 + H_2O + H + Cl^- \quad (4)$$

$$HOCl + 2/3NH_4^+ \rightarrow 1/3N_2 + H_2O + 5/3H + Cl^- \quad (5)$$

$$HOCl + 1/4NH_4^+ \rightarrow 1/4NO_3^- + 1/4H_2O + 3/2H^+ + Cl^- \quad (6)$$

Electrolysis mechanism without addition of Cl^- at the beginning of the electrolysis process:

$$2NH_3 + 6OH^- \rightarrow N_2 + 6H_2O + 6e^- \quad (7)$$

$$6H_2O + 6e^- \rightarrow 3H_2 + 6OH^- \quad (8)$$

$$2NH_3 \rightarrow N_2 + 3H_2 \quad (9)$$

3.4. Mechanism electro degradation of ammonia using flow electrolysis

According to the results of research by [32], the electrolysis of ammonia can produce hydrogen which can be used as fuel for fuel cells. Hydrogen produced from the electrolysis of a solution containing 1 M NH_3/5 KOH with a current density of 2.5 mA/cm^2, using an anode containing Rh 1 mg/cm^2 and Pt 10 mg/cm^2 at ambient temperature and pressure required 14.54 Watt h/g H_2 was produced. Electrolysis can be used to reduce the concentration of NH_3 water flow circulation method. This method can produce environmentally products, namely hydrogen (H_2) and nitrogen (N_2) from the NH_3 oxidation reaction. In this work, the materials used were easy to obtain, cheap, without adding chemicals, without filtering, and short-time analysis.

Electrolysis was a new way to effectively produce hydrogen from biomass, by converting acetate into hydrogen gas [33]. The process of electrolyzing ammonia to produce hydrogen and treating waste with ammonia content has been developed recently [34]. Mechanism electro degradation of ammonia using flow electrolysis is shown in Fig. 6.

$$2NH_{3(g)} + 6OH^-_{(aq)} \rightarrow N_{2(g)} + 6H_2O_{(l)} + 6e^- \quad E_0 = -0.77 \text{ V/SHE} \quad (10)$$

$$6H_2O_{(l)} + 6e^- \rightarrow 3H_{2(g)} + 6OH^-_{(aq)} \quad E_0 = -0.83 \text{ V/SHE} \quad (11)$$

The first reaction is an ammonia oxidation reaction that occurs at the anode, while the second reaction was a water reduction and occurs at the cathode. The overall reaction of the two reactions can be written by reaction *n* (12), while the magnitude of the standard cell potential of the two reactions was calculated from the standard potential of reduction minus the standard potential of oxidation.

$$2NH_{3(l)} \rightarrow N_{2(g)} + 3H_{2(g)} \quad E_0 = -0.06\text{V} \quad (12)$$

Based on Table 2 shows that the initial concentration of NH_3 to these four parameters was different. It was unstable nature of NH_3 and waste before treatment. The optimum condition for flow electrolysis of hospital wastewater was a positive-negative parallel circuit with the highest %degradation 87.50% at 20 V and 12 times water circulation. It is caused by electrolyzed of NH_3 firstly. Anode can oxidize NH_3 and cathode can reduce water. Besides, voltage and water circulation were influenced to reduce the concentration of NH_3. Higher voltage and water flow circulation increase redox reaction to the solution. Based on the research of [35] which the greater the stress applied to the tool, the faster the NH_3 oxidation reaction. According to [33] the reactions that occur in the electrolysis of NH_3 are:

Anode:

$$2NH_{3(g)} + 6OH^-_{(aq)} \rightarrow N_{2(g)} + 6H_2O_{(l)} + 6e^- \quad (13)$$

Cathode:

$$6H_2O_{(l)} + 6e^- \rightarrow 3H_{2(g)} + 6OH^-_{(aq)} \quad (14)$$

Total reaction:

$$2NH_{3(l)} \rightarrow N_{2(g)} + 3H_{2(g)} \quad (15)$$

Therefore, this work was conducted using the flow electrolysis method using carbon electrodes. The parameters applied to the flow electrolysis device were using a positive-negative parallel circuit parameter with a voltage of 20 V and 12 times water flow circulation because these parameters were the optimum conditions to reduce the concentration of NH_3.

The flow electrolysis method in this work can be used to reduce the concentration of NH_3 in the hospital wastewater. Based on this work, many factors can be affected to the results such as various voltage, water flow circulation, flow rate, number of carbon electrodes, the surface area of electrodes, and lack of interaction time between the hospital liquid waste samples and carbon electrodes.

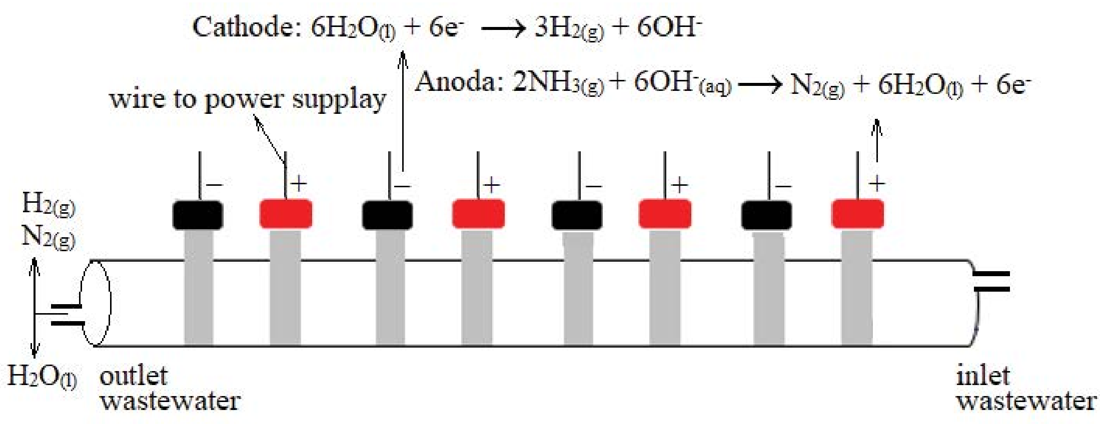

Fig. 6. Mechanism electro-degradation of ammonia using flow electrolysis.

Table 2
Effect of circuit type, amount of circulation, and optimum potential to %degradation of NH_3 in hospital wastewater

Circuit	Optimum potential (V)	Optimum cycles	Initial concentration NH_3 (mg/L)	Residual concentration NH_3 (mg/L)	%Degradation
NPP	15	9x	6.76	3.70	45.88
NPS	10	9x	6.76	5.10	24.56
PNP	20	12x	6.76	0.84	87.57
PNS	15	9x	6.76	4.77	29.44

4. Conclusion

In this work, a novel new method of flow electrolysis was used to reduce the concentration of ammonia in hospital wastewater. The characterization of carbon electrodes before and after treatment using SEM-EDX with a magnification of 5,000 times shows that the morphology of the electrodes appears too deep pores and the EDX results of the electrodes contain the highest element was carbon about 94.18% and 86.31%, respectively. The optimum conditions to reduce the concentration of NH_3 were positive-negative parallel circuit with voltage 20 V and 12 times water flow circulation was 87.50%. Flow electrolysis method using carbon electrodes was alternatively to reduce NH_3 concentration in hospital wastewater. Waste treatment techniques with flow electrolysis are strongly influenced by the waste flow rate and the number of electrodes used. The research proposal to improve this method is to study the effect of the number of electrodes and flow rate.

Acknowledgments

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