AIR STRIPPING FOR AMMONIA REMOVAL FROM LANDFILL LEACHATE IN VIETNAM: EFFECT OF OPERATION PARAMETERS

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ABSTRACT

Leachate is the wastewater from landfill that contains various pollutants at high concentrations. The treatment of leachate requires a complicated wastewater treatment system including chemical, physico-chemical, biological, and advanced treatment processes. The high ammonia concentration of leachate usually causes inhibition for microorganism in biological treatment. Therefore, it is necessary to remove ammonia from the leachate to lower concentrations to make it suitable for further treatment processes. In this study, we designed an air stripper for removal of ammonia in both synthetic and leachate wastewater. The effects of pH, hydraulic loading rate (HLR), gas/liquid (G/L) ratio, and recirculation of liquid on the ammonia stripping efficiency were investigated. The results show that rising pH from 9 to 12 increased significantly ammonia removal efficiency, irrespective of the changes of G/L or HLR. At both HLR of 57.6 and 172.8 m^3/m^2.day, increase G/L ratio led to the enhancement of removal efficiency, getting the highest value of 56% at HLR of 172.8 m^3/m^2.day, pH 12, and G/L of 728. Furthermore, recirculating of leachate improved the stripping efficiency of ammonia up to 99.0% after three hours with the output concentration of 25.2 mg/L. The results from this study hence proved the effectiveness of air stripping as a pre-treatment process for ammonia removal from landfill leachate and suggested suitable operating conditions.

XỬ LÝ AMONI TRONG NƯỚC RỈ RÁC TỪ BÃI CHÔN LẤP TẠI VIỆT NAM BẰNG PHƯƠNG PHÁP TÁCH KHÍ: ẢNH HƯỞNG CỦA CÁC THÔNG SỐ VẬN HÀNH

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Nước rỉ rác
Bãi chôn lấp
Tách khí
Truyền khối
Amoni

TÓM TẮT

Nước rỉ rác là nước thải từ bãi chôn lấp chứa các thành phần ô nhiễm ở nồng độ cao. Do đó, việc xử lý nước rỉ rác cần một hệ thống phức tạp bao gồm các quá trình hóa học, hóa lý, sinh học và xử lý nâng cao. Nồng độ cao của amoniac trong nước rỉ rác ngăn cản sự phát triển của các vi sinh vật trong xử lý sinh học nên cần loại bỏ amoni xuống nồng độ thấp hơn, tạo điều kiện thuận lợi cho các quá trình xử lý tiếp theo. Trong nghiên cứu này, tháp tách khí để loại bỏ amoni từ nước thải tổng hợp và nước rỉ rác đã được thiết kế và thử nghiệm. Ảnh hưởng của pH, tải trọng thủy lực (HLR), tỷ lệ khí/lỏng (G/L), và thời gian tuần hoàn lên hiệu quả tách amoni đã được nghiên cứu. Kết quả cho thấy việc tăng pH từ 9 tới 12 đã tăng hiệu quả xử lý amoni dù hệ thống vận hành ở những tỷ lệ G/L hay HLR khác nhau. Tại HLR bằng 57.6 và 172.8 m^3/m^2.ngày, tăng tỷ lệ G/L nâng cao được hiệu quả xử lý, đạt 56% với HLR ở 172.8 m^3/m^2.ngày, pH 12, và G/L 728. Việc tuần hoàn nước rỉ rác đã cải thiện đáng kể hiệu quả tách amonia, lên tới 99.0% sau ba giờ, đạt nồng độ amoni đầu ra là 25.2 mg/L. Như vậy, kết quả từ nghiên cứu này đã chứng minh hiệu quả của phương pháp tách khí trong tiền xử lý amoni từ nước rỉ rác và đề xuất được các điều kiện vận hành phù hợp.

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1. Introduction

Leachate is a type of wastewater generated in landfills, formed by leakage of rainwater into landfills or due to the available moisture of waste accumulated in the bottom layer of landfill and seepage through the soil. In general, there are four main components in leachate, including (i) organic compounds such as dissolved organic substances, volatile fatty acids (acetic, propionic, butyric compounds), fulvic acid, humic acid, etc.; (ii) main inorganic ions: Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, NH_4^{+}, Fe^{2+}, Mn^{2+}, Cl^{-}, SO_4^{2-}, and HCO^{3-}; (iii) heavy metals: Cd^{2+}, Cr^{3+}, Cu^{2+}, Pb^{2+}, Ni^{2+}, and Zn^{2+}; (iv) xenobiotic organic compounds: aromatic compounds, phenols, pesticides, chlorinated aliphatics, plastics, etc. and oil-derived components of fuel: benzene, toluene, xylene, etc. Among these components, ammonia nitrogen is one of the pollutants of concern because its concentration is very high (800-5210 mg/L), even in leachate from old landfills [1]. The total ammonia nitrogen (TAN) up to 1000 mg/L can inhibit microbial activity, reducing the effectiveness of the biological based processes [2], [3]. Because it is generated from waste, leachate is very toxic and difficult to be handled, causing serious environmental pollution. It is known that with a certain amount of leachate absorbed into the soil, this wastewater can contaminate groundwater while if it follows into the canal, the water environment can be deteriorated. Therefore, leachate must be thoroughly treated before being discharged into the environment. To solve the problem of pollution from leachate, many technologies have been studied and applied, such as biological (anaerobic and aerobic), chemical-biological (Fenton-anaerobic-aerobic, and stripping-anaerobic-aerobic), physical, chemical oxidation, and membrane technologies.

Several studies have reported that air stripping is successful in removing ammonia from landfill leachate and many other wastewaters [4]-[9], such as those from the fertilizer industry [10], pig slurry [11], [12], anaerobic digestion effluent [13], [14] or source-segregated food waste [15]. The effectiveness of ammonia removal obtained in these studies was in the range of 90 to 99%. Ozturk et al. [6] used air-stripping to treat ammonia in leachate at the optimum pH of about 10, 11, and 12. The results showed that after 2 hours of aeration, the ammonia removal was 72% at pH 12 while it was nearly 20% at pH 10 and 11. Under continue aeration for the next 24 hours, the ammonia removal was at 45, 80, and 85% after 6, 12, and 17 hours, respectively [6]. In addition, Marttinen et al. [16] also used air-stripping tower with a 1.1-liter PVC column (6 cm in diameter and 40 cm high) filled by plastic materials, to remove ammonia from leachate. Experiments were performed at pH 11 at temperatures of 6, 10, and 20 °C and a flowrate of 2 or 10 L/h for 0, 6, and 24 hours. In the 24-hour test, the highest ammonia removal of 89% was achieved at pH 11, 20 °C and a gas flowrate of 10 L/h [5]. Furthermore, most studies on air stripping relied on small stripping units in which air was bubbled at flow rates of 1.2 to 300 L/h and only a small volume of leachate (0.8-4 L) was treated [5]-[7], [11], [17]-[19]. The ratio of G/L (m^3/m^3) varied for each reference which was 50-150 [9], 45-200 [20], 1250 to 2000 [21], 3480 [22], and 2000-6000 [23].

In Vietnam, different technologies for ammonia removal from landfill leachate have been investigated, including partial nitrification and denitrification in SBR [24], chemical precipitation [25] for Nam Son landfill leachate, combining the anoxic and attached growth processes at Phuoc Hiep landfill [26], completely autotrophic nitrogen-removal over nitrite - SBR process for Go Cat Landfill leachate [27], electrocoagulation and bio-filter for Nam Son Leachate [28], [29], and Fenton process followed by coagulation for Quang Hanh landfill [30]. Though the application of air stripping for ammonia removal in leachate has been reported from some research around the word, such research is rarely found in Vietnam, except for one report with limited information about the roles of respective treatment processes of pH adjustment with $CaCO_3$, air stripping, activated sludge, coagulation using $FeCl_3$, Fenton oxidation, sand filtration, and chlorine disinfection processes in Phuoc Hiep and Go Cat landfills [31]. Therefore, the objective of this study was to apply a pilot air stripping tower with Kaldnes packing material for removal of ammonia from synthetic leachate and the leachate collected from Go Cat Landfill.

2. Materials and methods

2.1. Synthesis of wastewater and leachate collection

Artificial wastewater was made from ammonium chloride (NH_4Cl) in tap water at different NH_4^+ concentrations. Real leachate was collected from Go Cat Landfill (Binh Hung Hoa Ward, Binh Tan District, Ho Chi Minh City). According to [27], this leachate is characterized as an old landfill leachate which was closed since 2007. The concentrated leachate was taken directly from the collection tank while the leachate diluted by rainwater was collected at storage pond.

2.2. Air stripping unit

An air stripping tower was designed with a 2 m high tube and a diameter of 0.09 m (Table 1). Kaldnes rings (25×25 mm) made of Polyvinyl Chloride with a specific surface area of 250 m^2/m^3 were used as the packing material in the tower. The height of packing material was 1.80 m. The tower was operated in batch mode at room temperature. As shown in Figure 1, the wastewater was conveyed by a dosing pump from the wastewater tank to the top of the tower. At this point the leachate was distributed evenly through the packing material and simultaneously contacted with air stream driven from the outside by an air blower. The treated wastewater was collected in a tank and its ammonia concentration was measured. In state#3 (Table 2), the treated wastewater was recirculated back to the inlet while samples were regularly taken for ammonia analysis.

Table 1. *Air stripping tower parameters*

Parameter	Unit	Value
Diameter	m	0.097
Height	m	2
Height of packing material	m	1.8
Air blower		
- Flow	m^3/h	3200
- Power	HP	2
- Pressure column	Pa	1000
Water pump		
- Pressure column	mH_2O	2.5
- Flow	m^3/h	40

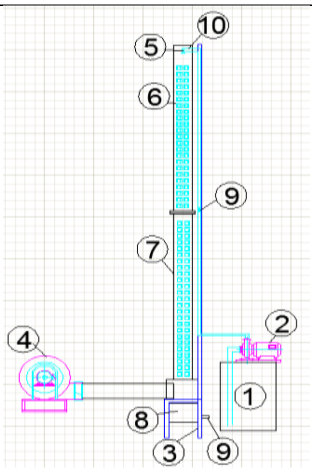

Figure 1. *Schematic diagram of the air stripping tower: (1) wastewater tank, (2) dosing pump, (3) frame, (4) air blower, (5) wastewater distribution system, (6) packing material, (7) column, (8) treated wastewater tank, (9) treated wastewater outlet, and (10) wastewater inlet*

2.3. Operating the tower

During the operation, it is necessary to control parameters pH and gas/liquid ratio (G/L) so that NH_4^+ in wastewater can be converted into NH_3 gas. pH of leachate was adjusted to 9, 10, 11, and 12 by slowly adding 30% NaOH solution. The pH raising process must take place slowly to prevent rising NH_3 too fast. G/L was controlled by air and water flowrates in which water flowrate was adjusted by throttle valve while air flowrate was monitored by an anemometer.

Table 2. *Operation modes of model*

Stage	Type of wastewater	Liquid flowrate (L/min)	G/L ratio (m^3/m^3)	Average $NH_4^+{}_{in}$ (mg/L)	pH
Stage #1	Synthetic wastewater	Q = 0.3 (L/min) (HLR = 57.6 (m^3/m^2.day))	G/L = 1802	3300, 3000, 1400, 500	pH: 9, 10, 11, 12.
	Leachate			4032, 3606, 3405	
Stage #2	Leachate	Q = 0.3 (L/min) (HLR = 57.6 (m^3/m^2.day))	G/L = 936 G/L = 1630 G/L = 2185	3780	pH: 9, 10, 11, 12.
	Leachate	Q = 0.9 (L/min) (HLR = 172.8 (m^3/m^2.day))	G/L = 312 G/L = 543 G/L = 728	3780	pH: 9, 10, 11, 12.
Stage #3	Synthetic wastewater	Recirculating 15, 30 min, 1, 2 and 3 h; (HLR = 172.8 (m^3/m^2.day))	G/L = 728	3080	pH: 12
	Leachate			2520	pH: 12
	Leachate			442	Initial pH (7.65)

**HLR: hydraulic loading rate*

Three stages of operation were designed as given in Table 2. The stage#1 was conducted in order to find the relation between input NH_4^+ concentrations (in both synthetic and real leachate), pH and removal efficiency. For the stage #2, effect of two hydraulic loading rates (57.6 and 172.8 (m^3/m^2.day)) and different G/L ratios on ammonia removal efficiency from real leachate was evaluated. In final stage (stage #3), the tower was operated at optimum values of HLR, G/L, pH found from previous stages and the liquid phase was recirculated at different periods of time (15 and 30 minutes; 1, 2 and 3 hours). Total liquid volume in this stage was 5 L for both synthetic wastewater and leachate.

2.4. Chemicals and parameters analysis

NH_4Cl, acid boric, acids, and bases used in this study were purchased at analytical grade. pH of wastewater was measured by Hanna Hi 8424 while the air flowrate was measured by an anemometer (Manometer Testo 435). Ammonia concentration in wastewater was analyzed according to Standard Methods 4500 NH3 B with duplicates for each analysis.

3. Results and discussion

3.1. Effect of pH and initial NH_4^+ concentration on NH_4^+ removal efficiency

Relationship between pH, initial ammonia concentration, and efficiency of ammonia removal in artificial wastewater and leachate is illustrated in Figure 2. As can be seen from Figure 2(a), NH_4^+ remove efficiency was increased obviously when pH increased and initial NH_4^+ concentration reduced. The highest efficiencies achieved at pH of 12 were 79, 70, 64, and 48%, corresponding to the initial concentrations of 500, 1400, 3000 and 3300 mg/L. According to reaction (1), this trend is reasonable because of rising pH led to the shift of the equilibrium of the reaction to produce more NH_3 into gas phase.

$$NH_4^+ \leftrightarrow NH_3 + H^+ \quad (1)$$

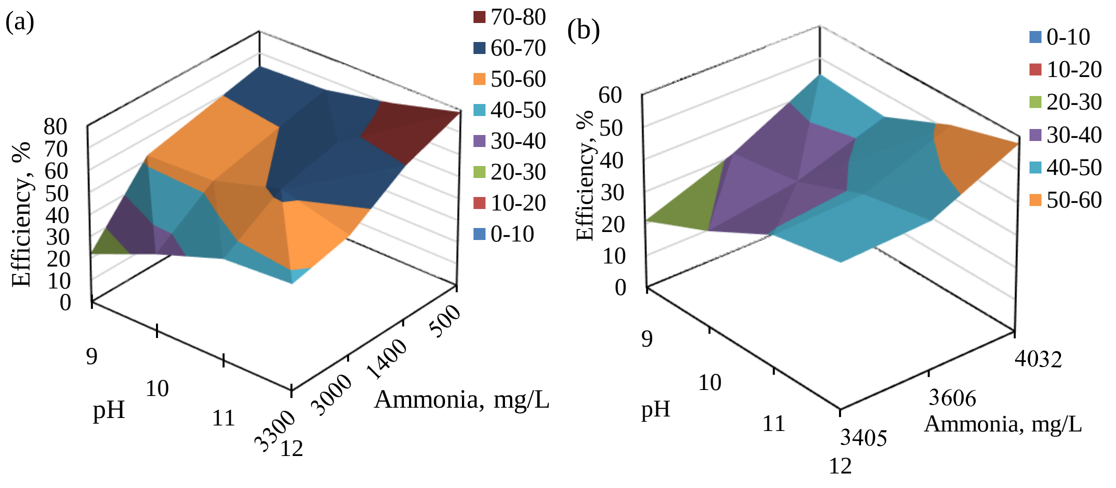

Figure 2. *Relationship between pH, initial ammonia concentration, and efficiency of ammonia removal in (a) artificial wastewater and (b) leachate*

The dependence of removal efficiency on pH of leachate was similar to that of synthetic wastewater, with the highest amount of NH_4^+ striped out at pH 12. This optimum pH was consistent with the results of pH value found by Ozturk, et al. [6] and Marttinen, et al. [16]. However, increasing initial NH_4^+ concentration of leachate from 3405, 3606 and 4032 mg/L resulted in the increasing of removal efficiency from 45, 46, and 58% at pH 12, respectively. This trend differed from that of synthetic wastewater which can be explained based on the free ammonia amount available in leachate but not in synthetic wastewater. As calculated via the equation (2) [32], leachate contained about 1-5% of free ammonia (FA) and the leachate with a higher ammonia concentration contains a higher FA content which was easily released at the pH of greater than 9. Therefore, the leachate with higher input concertation of NH_4^+ could achieve a higher removal efficiency.

$$FA = \frac{C_{NH_4^+} \times 10^{pH}}{\frac{k_b}{k_w} + 10^{pH}} \quad (2)$$

Where $C_{NH_4^+}$ is ammonia concentration in leachate and $\frac{k_b}{k_w} = \exp\left(\frac{6,344}{273+t}\right)$

3.2. Effect of hydraulic loading rate and gas/liquid ratio on NH_4^+ removal efficiency

The results achieved from the operation of stage #2 are illustrated on Figure 3. This experiment was conducted with two hydraulic loading rates (HLR) of 57.6 and 172.8 m^3/m^2.day, pH ranged from 9 to 12, at different G/L ratios. To increase G/L ratio, we used a fixed wastewater flowrate while increased air flowrate (Table 2).

In consistent with the results from Section 3.1, the increase of pH from 9 to 12 significantly increased the removal efficiency of NH_4^+, irrespective of the changes of G/L or HLR, getting the highest values at pH 12. Under pH 12 and the HLR of 57.6 m^3/m^2.day, increase G/L ratio from 936 to 1630 led to the increase of removal efficiency, i.e. from 40 to 54%. This is explained based on the Equation (3) [23]. Accordingly, when G/L increases, concentration of ammonia in the output (C_e) will reduce or removal efficiency will increase. At this point, the working line shifts to the equilibrium line. However, the removal efficiency was unchanged when we further increased G/L ratio from 1630 to 2815 (i.e. 54 and 54%, respectively).

$$(G/L) = (P_T/H) \times (C_0 - C_e)/C_0 \quad (3)$$

Where H is Henry's constant for ammonia, P_T is total pressure, C_0 and Ce is the input and output concentrations of ammonia, G/L is the minimum ratio of gas and liquid.

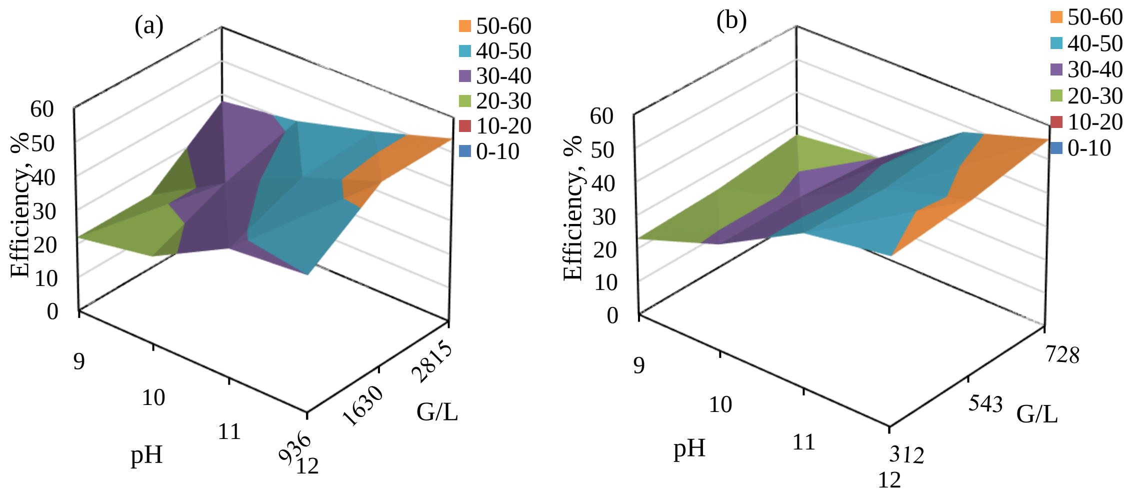

Figure 3. *Relationship between pH, G/L, and efficiency at (a) Q = 0.3 L/min, HLR = 57.6 m^3/m^2.day and (b) Q = 0.9 L/min, HLR = 172.8 m^3/m^2.day*

For the case of HLR of 172.8 m^3/m^2.day and G/L ratio of 312, 543 and 728, the removal efficiency was slightly changed from 50, 52, to 56%, respectively, which is in consistent with the results of [13, 21]. We hence selected HLR of 172.8 m^3/m^2.day, G/L ratio of 728 at pH 12 due to the induced highest removal efficiency and wastewater treatment capacity. The G/L ratio of 728 in this study was higher than those from [9] (i.e. 50-150), [20] (i.e. 45-200) but smaller than the values applied in [21] (i.e. 1250-2000), [22] (i.e. 3480), and recommended in [23] (i.e. 2000-6000).

3.3. Effect of recirculation on NH_4^+removal efficiency

The results from the previous sections showed that the NH_4^+ removal efficiency from the air stripping tower ranged from 48-79% for synthetic wastewater and 45-58% for leachate. To enhance the stripped amount of ammonia, recirculation of wastewater (5 L) was applied. As can be seen from Figure 4, operation with the artificial wastewater at initial NH_4^+ concentration of 3080 mg/L could yield the efficiency from 90% at 15th minute to 99% at the 120th minute. At the same time, the NH_3 concentration calculated in the gas phase decreased considerably from 1297 to 179 mg/m^3. A similar trend of change in removal efficiency was found for the leachate containing 2520 mg/L of ammonia, from 81% at 15th minute to 99% at 120th minute. The NH_4^+ output concentration was 25.2 mg/L which is approximately equal to the allowable value in column B (i.e. 25 mg/L) from national technical regulation on wastewater of the solid waste landfill sites (QCVN 25:2009/BTNMT). This efficiency is higher compared to those obtained from previous studies, e.g. 98% with the operation time of 4 to 9 days [20], 95.5% for 3 hours [22]. During this period, the NH_3 concentration dropped from 956 to 97 mg/m^3, but was still higher than the value recommended in air quality – maximum allowable concentration of hazardous substances in ambient air (i.e. 0.2 mg/m^3, TCVN 5938:2005) or the allowable value given in national technical regulation on industrial emission of inorganic substances and dusts (i.e. 50 mg/m^3, column B, QCVN 19: 2009/BTNMT).

Though pH adjustment improved significantly the NH_4^+ removal efficiency but this step consumes chemicals and requires the neutralization of the wastewater after the treatment to facilitate the next treatment steps. For the rainwater diluted leachate with a low concentration of NH_4^+ (442 mg/L), we further tested the air stripping without pH adjustment and found the removal efficiency was 8, 18, 40, 74, and 91% at 15th, 30th minute, 1st, 2nd, and 3rd hour, respectively. Hence, recirculating the leachate could be considered as an effective pretreatment step improved the removal efficiency.

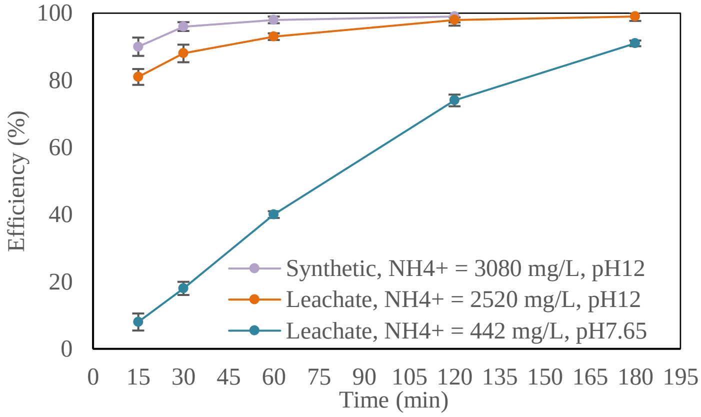

Figure 4. *Effect of recirculation time on NH_4^+ removal efficiency from artificial wastewater and leachate (error bars present standard deviations, n = 3)*

4. Conclusions

In this study, air stripping for ammonia removal in synthetic wastewater and leachate was investigated under various operating conditions of pH, initial ammonia concentration, hydraulic loading rate, gas to liquid ratio, and recirculating time. As a result, the increase of pH from 9 to 12 led to the significant increase of ammonia removal efficiency, irrespective of the changes of G/L or HLR, with the highest ammonia stripping achieved at pH12. For both hydraulic loading rates of 57.6 and 172.8 m^3/m^2.day, rising G/L ratio resulted in the improvement of removal efficiency, up to 56%. Under the HLR of 172.8 m^3/m^2.day, pH 12, G/L of 728 with liquid recirculation, the leachate containing ammonia at 2520 mg/L was stripped out 99% of ammonia for three hours. The final concentration of ammonia was 25.2 mg/L which is about equal to the allowable value from the discharging standard of leachate. The results from this study hence proved the effectiveness of air stripping in ammonia removal from leachate and the optimum operating conditions were suggested. Further investigation is needed for recovery the amount of ammonia stripped and released into gas phase so that this gas stream can meet the requirement to discharge into the air.

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