Nitrogen source recovery from mature leachate via heat extraction technology: An engineering project application

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ABSTRACT

Large pool of ammonia in mature leachate is challenging to treat with a membrane bioreactor system to meet the discharge standard for pollution control of municipal solid waste landfills in China (GB 16889-2008) without external carbon source addition. In this study, an engineering leachate treatment project with a scale of 2,000 m³/d was operated to evaluate the ammonia heat extraction system (AHES), which contains preheat, decomposition, steam-stripping, ammonia recovery, and centrifuge dewatering. The operation results showed that NH₃-N concentrations of raw leachate and treated effluent from an ammonia heat extraction system (AHES) were 1,305–2,485 mg/L and 207–541 mg/L, respectively. The ratio of COD/NH₃-N increased from 1.40–1.84 to 7.69–28.00. Nitrogen was recovered in the form of NH₄HCO₃ by the ammonia recovery tower with the introduction of CO₂, wherein, the mature leachate can offer 37% CO₂ consumption. The unit consumptions of steam and power were 8.0% and 2.66 kWh/m³ respectively, and the total operation cost of AHES was 2.06 USD per cubic leachate. These results confirm that the heat extraction is an efficient and cost-effective technology for the recovery of nitrogen resource from mature leachate.

Key words: ammonia heat extraction system, comprehensive operation cost, mature landfill leachate, nitrogen source recovery

HIGHLIGHTS

- An leachate treatment engineering project with a scale of 2,000 m³/d was introduced.
- AHES was developed for nitrogen resource recovery from mature leachate.
- Nitrogen resource was recovered via the formation of NH₄HCO₃.
- The comprehensive operation cost of AHES was 13.11 RMB/m³ of leachate.

1. INTRODUCTION

Sanitary landfills are a widely used method for solid waste disposal (Renou et al. 2008; Rowe & Yu 2012; Xiong et al. 2018). Generally, most landfills have been on service for more than ten years in China (Hagman et al. 2008; Cortez et al. 2010). Thus, landfill management inevitably encounters the problem of mature leachate treatment, which is typically characterized by a low ratio of BOD₅/COD and high NH₃-N concentration (Cortez et al. 2011). The typical COD/TN ratio of mature leachate is around 1–3 (Pehkonen & Zhang 2002) to become the bottleneck of nitrogen removal by the nitrification-denitrification process.

Various chemical and physical processes have been developed to treat mature leachate, including air stripping, coagulation, flocculation, and advanced oxidation processes. However, the application of these technologies mainly requires significant capital investment in equipment and burdensome daily maintenance (Amor et al. 2015; de Almeida et al. 2019). Membrane bioreactor (MBR) systems employ a combination of an active sludge system and ultrafiltration unit, which has proven to be practical and useful for leachate treatment (Oller et al. 2011; Abuabdou et al. 2018). However, denitrification requires a large
amount of external carbon source, thereby limiting their application for mature leachate treatment (Bui Xuan et al. 2013; Amaral et al. 2017). Additionally, the high initial investment associated with conventional leachate treatment has led to a desire for novel technologies for NH$_3$-N removal or recovery, especially for mature leachate, as well as the wastewater (Le et al. 2021).

In the biological treatment process, an appropriate chemical oxygen demand/total nitrogen (COD/TN) ratio is preferred to enhance the denitrification of mature leachate. For example, reducing NH$_3$-N concentration of biological denitrification treatments influent could decrease the need for an external carbon source. Efforts to reduce operational cost and utilize nitrogen resources have increased the urgency to develop efficient NH$_3$-N recovery technologies for mature leachate (Zhang et al. 2013; Li et al. 2018). Presently, air-stripping and steam-stripping systems are the effective methods for the NH$_3$-H recovery from mature leachate. In the nitrogen air stripping process, a pH greater than 10 should be maintained in the stripping tower, and then adjusted downward to 7–8 for subsequent treatment, thus the cost is mainly the NaOH and H$_2$SO$_4$ reagents purchase. What’s worse, the addition of H$_2$SO$_4$ will react with Ca$^{2+}$, which generates CaSO$_4$ and causes membrane blockage phenomenon. And further, the yield of concentrate increases, which also decrease the effluent yield and increase the operation cost. Additionally, the removal efficiency of NH$_3$-N is greatly affected by temperature. Due to the problems of the complex control, unstable operation, difficult recovery of product and the secondary pollution, the air-stripping process for NH$_3$-N removal has only been employed in the Shenzhen Xiaping landfill leachate treatment plant at a scale of 1,500 m$^3$/d.

As a consequence, it is necessary to develop a cost-effective technique for the removal of high concentration of NH$_3$-N in mature leachate. Due to the advantages of high effectiveness, simplicity and environmental sustainability, heat extraction technology has proven to be a valid alternative for ammonia nitrogen recovery from mature leachate (He et al. 2015; Yuan et al. 2016).

In an ammonia heat extraction system (AHES), steam is used as the carrier. The steam-stripping process has been determined to be more cost-effective than conventional air-stripping based on the energy cost of waste steam (Höhne & Niemeyer 2001; Xiong et al. 2018). Furthermore, the negative pressure steam-stripping process is a promising technology for recovering ammonia nitrogen in mature leachate because of the low-boiling point of water and the decomposition of HCO$_3^-$ under negative pressure (Teichgraber & Stein 1994). Our previous publication showed that an average NH$_3$-N recovery efficiency of 82.03% was obtained from mature leachate by using a pilot-scale (8–10 m$^3$/d) negative pressure steam-stripping technology (Xiong et al. 2018). However, for the verification of technical validation, this engineering AHES had a significant difference from the pilot-scale experiment because of the cost and facilities investment differential.

Aiming at nitrogen recovery and improving COD/TN of mature leachate, the AHES was established in this engineering project, based on the technical parameters of pilot-scale performance. The objectives of this study include (1) to evaluate the operational viability of heat extraction technology for nitrogen recovery from mature leachate in Shanghai Laogang Landfill, China; and (2) to assess the cost-effectiveness of the AHES in terms of material balance, energy consumption, and operational cost. The results of this study can also be applied to other nitrogen resource recovery engineering for the liquid digestate of food waste and sludge anaerobic digestion treatment.

2. MATERIALS AND METHODS

2.1. Characteristics of leachate

The mature leachate was collected from the Laogang municipal solid waste landfill in Shanghai, China (Figure S1). The physical and chemical properties of the leachate are listed in Table 1.

2.2. Technological process and operation

The workflow of AHES is presented in Figure 1. The raw leachate was pumped to a preheater and heated by the effluent of the steam-stripping tower, of which temperature is 80–90 °C. After preheating, the temperature of leachate was increased to 60–70 °C, and leachate entered the decomposition tower with the internal pressure being 6.96–9.57 psi. Saturated steam was simultaneously introduced to the decomposition tower for further heating the preheated leachate, dissociating HCO$_3^-$ and increasing pH. Furthermore, the effluent from the decomposition tower was pumped to the steam-stripping tower with an internal pressure of 8.12–10.73 psi, which are not an absolute or complete vacuum process. The ammonia steam was returned to the decomposition tower to maintain the temperature of 80–85 °C. The effluent ammonia steam from the decomposition tower was introduced to the condenser for gas-liquid separation. Ammonia steam was adsorbed by the
vacuum-ammonia absorption integrated device, therein, because of the system settings, there is no absolute vacuum. Finally, the ammonium bicarbonate was formed and subsequently precipitated from the supersaturated solution in the ammonia recovery tower with an operating temperature of 35.4–39.4 °C.

2.3. General layout and design parameters of AHES

The general layout is shown in Figure 2, and the actual engineering project and primary devices lists of AHES are shown in Figure S2 and Table S1, respectively. The engineering project was started in April, and the trial operation began in May 2018. Results from trial operation from May to August 2018 showed that the NH₃-N concentrations of the influent and effluent were 2,220–3,910 mg/L and 325–1,000 mg/L, respectively. The AHES was conducted for stable operation with a hydraulic loading of 2,000 m³/d for 30 days from April 19 to May 21, 2019. It should be noted that the steam-stripping tower was cleaned from May 1 to May 3, and the power was cut off on May 6. In this period, the treatment capacity was 1,000 m³/d. Therefore, the total treated leachate was 63,123 m³ during the performance test.

2.4. Actual NH₃-N concentration calculation

In this study, based on the mass balance and practical engineering project (2,000 t/d treatment scale) in Laogang MSW landfill, the actual NH₃-N concentration was calculated as the Equation (1). Wherein, the parameters based on the work from the

<table>
<thead>
<tr>
<th>Characteristics of the mature leachate used in this study</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Items</strong></td>
</tr>
<tr>
<td>COD</td>
</tr>
<tr>
<td>NH₃-N</td>
</tr>
<tr>
<td>Alkalinity</td>
</tr>
<tr>
<td>Hardness (Ca and Mg)</td>
</tr>
<tr>
<td>TSS</td>
</tr>
<tr>
<td>pH</td>
</tr>
</tbody>
</table>

Note: the sampling numbers of COD, NH₃-N, alkalinity, hardness, TSS, and pH analysis were 30, 30, 8, 4, 4, and 30, respectively.

Figure 1 | Schematic diagram of the heat extraction technology for ammonia recovery. (note: P represents the pump).

Figure 2 | General layout of AHES.
authors’ previous research work (Xiong et al. 2018).

\[ QC_i = Q(1 + \alpha)C_e + 14/79 \cdot T \cdot (1 - \beta) + \gamma Q_{Cn} + Q_f C_f \quad (1) \]

Q: Water treatment capacity (m³/d);
Ci: NH₃-N concentrations of influent (mg/L);
α: Steam consumption, 8–10% of water treatment capacity;
Cₑ: NH₄-N concentrations of effluent (mg/L);
T: NH₄HCO₃ yield (kg/d);
β: NH₄HCO₃ moisture, 3–5%;
γ: Concentrated residue yield (m³/d), generally required 2–4% of water treatment capacity;
Cₙᵣ: NH₃-N concentrations in concentrated residue from decomposition tower (mg/L), generally 5,500–6,500 mg/L;
Qₙᵣ: Flow of non-condensable gas discharged from the system (m³/d);
Cᵣ: NH₃-N concentrations of non-condensable gas discharged from the system (mg/m³).

2.5. Analytical methods

Samples were collected at the influent and effluent of the decomposition tower and the pipe of the cryogenic cooler. pH was tested three times a day during the engineering operation using the glass electrode (PHBJ-260, INESA Scientific Instrument Co., Ltd., China). Leachate quality parameters (COD, NH₃-N, and alkalinity HCO₃⁻) were analyzed according to the guidelines of the Chinese Ministry of Environmental Protection. Specifically, COD tests were conducted using the closed reflux method. NH₃-N was measured using Nessler’s reagent photometry method, and NH₄HCO₃ was analyzed with the gravimetric method. During the engineering operation, CO₂ consumption was automatically recorded using a flowmeter (SM-R1-X, Shangdong Veici Gas Equipment Co., LTD., China). Pressures and temperatures were recorded every 6 hours at the top and button of towers.

3. RESULTS AND DISCUSSION

3.1. Ammonia recovery from leachate

During the stable operation, the hydraulic loading of AHES was 2,000 m³/d. Variations of the pH, NH₃-N concentration, and COD/NH₃-N ratio of influent and effluent are shown in Figure 3. The NH₃-N concentrations of influent were 2,215–2,720 mg/L, and values of COD/NH₃-N and the pH were 1.40–1.84 and 8.00–8.46, respectively. After heat extraction treatment, the NH₃-N concentrations of the effluent were dropped to 118–459 mg/L, and the pH (9.46–9.88) was higher than that of the influent.
In the AHES, NH$_3$-N recovery was realized via the following main steps (Xiong et al. 2018): (1) the preheated leachate entered the decomposition tower and achieved a high pH because of the bicarbonates and carbonates decomposition; (2) the effluent entered the steam-stripping tower for the desorption of the ammonia vapor; (3) ammonia vapor entered the condenser to desorb the gas from the liquid, and the condensate was recirculated. The ammonia was recovered in the ammonia recovery tower, and ammonium bicarbonate was obtained (Figure 4). Accompanied by NH$_3$-N removal and recovery, the ratio of COD/NH$_3$-N of the effluent increased to 7.68–28.00, with an average value of COD/NH$_3$-N of 13.73. Based on the practical engineering operation situation, the suitable value ranges of COD/NH$_3$-N for MBR is over 8.0, which is obtained from the engineering projects operated by Shanghai Municipal Engineering Design Institute (Group) Co., LTD.

3.2. Mechanisms involved in ammonia heat extraction system

The mechanisms involved could be separated into three processes.

Firstly, Due to the acid base behaviour of the HCO$_3^-$, which exists hydrolysis and ionization processes in the aqueous solution. The preheated raw leachate entered the decomposition tower from the top and underwent heat exchange with ammonia steam. Based on the phase equilibrium principle of CO$_3^{2-}$ (Xiong et al. 2018), the pH value of decomposition
tower effluent was 9.46–9.88 to motivate the decomposition of $\text{HCO}_3^-$ into $\text{CO}_3^{2-}$ and $\text{CO}_2$, increasing the concentration of free ammonia. The main chemical reactions are represented by Equations (2)–(4) in the decomposition tower.

\[
\begin{align*}
\text{HCO}_3^- + \text{H}_2\text{O} & \rightleftharpoons \text{H}_2\text{CO}_3 + \text{OH}^- \quad (2) \\
\text{H}_2\text{CO}_3 & \rightleftharpoons \text{CO}_2 + \text{H}_2\text{O} \quad (3) \\
\text{HCO}_3^- & \rightleftharpoons \text{H}^+ + \text{CO}_3^{2-} \quad (4)
\end{align*}
\]

In the engineering project, the internal pressure of the decomposition tower was 6.96–9.57 psi, and the corresponding boiling point was 80.6–87.6 °C. However, the actual detected temperatures at the top (import) and bottom (export) of the decomposition tower were 68.9–78.3 °C and 76.8–85.4 °C, respectively. It indicated the adequate decomposition reactions, and the preheated raw leachate was further heated.

Secondly, the saturated steam was then introduced under negative pressure, and the free ammonia was transferred into the vapor from leachate, and ammonia-containing steam was formed. These processes occurred in the steam-stripping tower. The internal pressure of the steam-stripping tower was 8.12–10.73 psi, and the corresponding boiling point was 83.7–90.6 °C. The actual detected temperatures at the top and bottom of the steam-stripping tower were 78.7–87.9 °C and 85.5–92.0 °C, respectively. The leachate was in a completely boiling state at the tower bottom. The external saturated steam further contributed to the desorption of free ammonia.

Finally, the ammonia-containing steam was returned to the decomposition tower for heat recovery and then was introduced into the condenser by the ammonia absorption device. Subsequently, the $\text{NH}_3\cdot\text{N}$ in steam was absorbed by water and interacted with $\text{CO}_2$ to form $\text{NH}_4\text{HCO}_3$ in the ammonia recovery tower. $\text{NH}_4\text{HCO}_3$ saturated solution that was discharged from the ammonia recovery tower was further cooled and centrifuged to obtain ammonium bicarbonate products, to realize nitrogen resource recovery. The involved reactions are shown in Equations (5) and (6).

\[
\begin{align*}
\text{NH}_3 + \text{H}_2\text{O} & = \text{NH}_3\cdot\text{H}_2\text{O} + \text{heat} \quad (5) \\
\text{NH}_3\cdot\text{H}_2\text{O} + \text{CO}_2 & = \text{NH}_4\text{HCO}_3 + \text{heat} \quad (6)
\end{align*}
\]

### 3.3. Factors affecting ammonia recovery

#### 3.3.1. Effect of temperature

The temperature was a significant factor for the equilibrium concentration of ammonia in the gas-liquid equilibrium. The solubility of $\text{NH}_3$ in wastewater decreased with increasing temperature under specific pressure, and the relationship between solubility and temperature is shown in Table 2. In AHES, the temperatures of preheated leachate and the effluent from the decomposition tower were 60–65 °C and 80–85 °C, respectively, leading to a high $\text{NH}_3\cdot\text{N}$ recovery efficiency. When dissolved in water, $\text{NH}_3\cdot\text{N}$ reacts to form $\text{NH}_3\cdot\text{H}_2\text{O}$, which is alkaline because of the ionization process represented by...
Equations (7) and (8)

\[
\text{NH}_3 + \text{H}_2\text{O} \rightleftharpoons \text{NH}_3\text{H}_2\text{O} \rightleftharpoons \text{NH}_4^+ + \text{OH}^-
\]  

(7)

\[
\text{NH}_3\text{H}_2\text{O} \rightleftharpoons \text{NH}_3 + \text{H}_2\text{O}
\]  

(8)

3.3.2. Effect of pH

The NH$_3$-N dissociation rate with respect to pH and temperature is presented in Table 3.

Most of the ammonia nitrogen in raw leachate was in the form of ammonium ion (NH$_4^+$) and free ammonia (NH$_3$), and the transformation process is represented by Equation (9) (Bonmati & Flotats 2003). At room temperature and pH 7.0, ammonia nitrogen exists in the form of NH$_4^+$. As pH increasing, NH$_4^+$ is dissociated to free ammonia. When the pH was 11.0, the proportion of free ammonia reached 98% and was not affected by temperature.

\[
\text{NH}_4^+ + \text{OH}^- \rightleftharpoons \text{NH}_3 + \text{H}_2\text{O}
\]  

(9)

3.3.3. Effect of pressure

In AHES, NH$_3$-N recovery is mainly based on the gas-liquid equilibrium principle (Adumitroaie et al. 2014). The gas-liquid equilibrium of the ammonia-water system under different pressures are represented in Table 4 (Tang et al. 2002). As seen from the equilibrium pressure table, the ammonia content in both the gas and liquid phases decreased with the decrease of pressure.

3.3.4. Engineering advantages for ammonia recovery

In the 2,000 m$^3$/d ammonia recovery engineering project, the operation was automatic and continuity, which could ensure the ammonia recovery stability. However, in the pilot scale study with a scale of 8–10 m$^3$/d (Xiong et al. 2018), the operation was indirect and manual, which caused the low of operation efficiency. Meanwhile, the lack of preheater and steam return system led to the loss of ammonia. And the nonuniform of mass and heat transfer was the other drawback of the pilot scale.

3.4. Material consumption and balance analysis

3.4.1. Material balance analysis

During the stable operation of the AHES, the total amount of NH$_3$-N recovered and NH$_4$HCO$_3$ generated were 123.83 t and 495.92 t (moisture content of 97%), respectively. The Equation (1) could express material balance of NH$_3$-N. The observed removal efficiency of NH$_3$-N was calculated based on the daily treated water volume, NH$_3$-N concentrations of the influent and effluent, steam consumption, and the amount of NH$_3$-N removed by the concentrated residue. The actual removal efficiency was calculated based on the actual daily production of NH$_4$HCO$_3$. The difference between both calculations is shown in Figure 5.

Table 2 | The solubility of NH$_3$ in wastewater at different temperatures (wt%)

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>0</th>
<th>10</th>
<th>20</th>
<th>30</th>
<th>40</th>
<th>50</th>
<th>60</th>
<th>70</th>
<th>80</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solubility</td>
<td>47.3</td>
<td>40.6</td>
<td>34.6</td>
<td>29.1</td>
<td>24.0</td>
<td>19.0</td>
<td>14.4</td>
<td>10.0</td>
<td>6.1</td>
</tr>
</tbody>
</table>

Table 3 | The dissociation rates of NH$_3$-N for different pH and temperature values (wt%)

<table>
<thead>
<tr>
<th>pH</th>
<th>20 °C</th>
<th>30 °C</th>
<th>35 °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>9.0</td>
<td>25%</td>
<td>50%</td>
<td>58%</td>
</tr>
<tr>
<td>9.5</td>
<td>60%</td>
<td>80%</td>
<td>83%</td>
</tr>
<tr>
<td>10.0</td>
<td>80%</td>
<td>90%</td>
<td>93%</td>
</tr>
<tr>
<td>11.0</td>
<td>98%</td>
<td>98%</td>
<td>98%</td>
</tr>
</tbody>
</table>
In Figure 5, it is evident that the actual recovery efficiency of NH₃-N is lower than that of apparent value. The main reasons for this discrepancy are as follows: (1) the difference between the apparent and actual ammonia concentration in the concentrated residue from the decomposition tower. NH₃-N concentration was selected as 6,500 mg/L for the calculation of apparent ammonia recovery. However, the actual concentration of NH₃-N in the concentrated residue was closely related to the temperature of the bottom of the decomposition tower because of the different solubilities; (2) partial of ammonia loss was due to the non-condensable gas discharge. The non-condensable vapor generated from the system was purified by washing and pickling.

In the purification system, washing liquid was circulated, and the pickling liquid was discharged periodically, which caused a loss; (3) the other partial ammonia was discharged through the sewage system in the process of overhauling and cleaning.

### 3.4.2. Correlation analysis of ammonium bicarbonate yield and CO₂ consumption

According to chemical equilibrium Equations (10)–(12), 0.56 kg CO₂ is required for producing 1.0 kg NH₄HCO₃ theoretically.

\[
\text{NH}_3 + \text{H}_2\text{O} \rightleftharpoons \text{NH}_4^+ + \text{OH}^- \quad (10)
\]

\[
\text{CO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{HCO}_3^- + \text{H}^+ \quad (11)
\]

\[
\text{NH}_4^+ + \text{HCO}_3^- \rightleftharpoons \text{NH}_4\text{HCO}_3 \quad (12)
\]

In AHES, 495.9 t of NH₄HCO₃ was obtained during the 30-days performance tested, and the theoretical consumption of CO₂ was 276.2 t. In comparison, the actual CO₂ consumption was 168.2 t, corresponding to 63% of the theoretical value. It indicated that the alkalinity of raw leachate could contribute to the NH₄HCO₃ generation. In the operation process, CO₂ generated from raw leachate decomposition in the decomposition tower participated in the synthesis of NH₄HCO₃ in the ammonia recovery tower, and reduced CO₂ consumption. The alkalinitlies of influent and effluent were 12,540–14,324 mg CaCO₃/L and 3,600–5,443 mg CaCO₃/L, respectively, indicating that the raw leachate had a 37% CO₂ contribution for the NH₄HCO₃ production. Apart from water, power, steam, the purchased CO₂ consumption was also the main cost component. In the operation process, CO₂ generated from raw leachate decomposition in the decomposition tower participated in the synthesis of NH₄HCO₃ in the ammonia recovery tower, and reduced CO₂ consumption.

What’s more, the dissociation of CO₃²⁻ and HCO₃⁻ dominated the total alkalinity of the raw leachate, which could be expressed by Equations (13)–(15). The hydrolysis rate of HCO₃⁻ increases with the increase of temperature and the decrease
with negative pressure, which confirms the CO₂ contribution of the raw leachate.

\[
\begin{align*}
\text{CO}_2^- + \text{H}_2\text{O} & \rightleftharpoons \text{HCO}_3^- + \text{OH}^- \\
\text{HCO}_3^- + \text{H}_2\text{O} & \rightleftharpoons \text{H}_2\text{CO}_3 + \text{OH}^- \\
\text{H}_2\text{CO}_3 & \rightleftharpoons \text{H}_2\text{O} + \text{CO}_2 \uparrow
\end{align*}
\]

(13)

(14)

(15)

3.4.3. Economic evaluation of negative pressure steam-stripping pretreatment

3.4.3.1. Water and power consumption. In AHES, water consumption occurs during ammonia adsorption, chemical washing, and solution preparation processes. The power consumption of the primary devices and unit power consumption of AHES are shown in Table 5 and Figure 6. Power consumption required to treat 1 m³ leachate range from 2.45 to 4.04 kW·h, with an average value of 2.66 kW·h. Because of the equipment maintenance in the 10–12th day, one of the AHES operation lines was taken off and the water treatment capacity drops sharply. However, the primary devices (such as cooling tower, inlet pump of steam-stripping tower, outlet pump, and so on), were shared in two production lines, which could not be taken off.

3.4.3.2. Steam and CO₂ consumption. The results for the steam consumption are shown in Figure 7, which indicated that the value of steam consumption/water treatment capacity was 8.0–9.0%, showing a negative correlation with the leachate treatment capacity. NH₄HCO₃ yield, theoretical and actual CO₂ consumption are shown in Figure S3.

3.4.3.3. Total operation cost. Apart from water, power, steam, and CO₂ consumption, a defoaming agent was used for the anti-clogging of the tower plate. In contrast, the sulfamic acid solution was used to clean the decomposition tower, ammonia recovery tower, preheater, and cryogenic cooler. Additionally, defoamer, sulfamic acid, and CO₂ were purchased. At the same time, an landfill gas power plant supplied steam. The total operating cost is shown in Table 6, and the unit operating cost was 2.06 USD/m³ of leachate, corresponding to 1.52 USD per 1 kg NH₃-N removed.

Comparatively, 1 kg NH₃-N removal in the MBR leachate treatment system requires at least 6 kg COD carbon source, of which the cost is about 0.63 USD. Thus, it can be estimated that AHES could save at least 2.25 USD/m³ of carbon source cost for leachate treatment, being a gospel for the subsequent MBR system. What’s more, it was calculated that the cost was
### Table 5 | Power consumption of primary devices in ammonia heat extraction system

<table>
<thead>
<tr>
<th>No.</th>
<th>Items</th>
<th>Rated capacity (kW)</th>
<th>Amount</th>
<th>Run time (h)</th>
<th>Energy consumption (kWh)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Centrifuge</td>
<td>30</td>
<td>1</td>
<td>8</td>
<td>240</td>
</tr>
<tr>
<td>2</td>
<td>Packer</td>
<td>25</td>
<td>1</td>
<td>8</td>
<td>200</td>
</tr>
<tr>
<td>3</td>
<td>Cooling tower</td>
<td>15</td>
<td>1</td>
<td>24</td>
<td>360</td>
</tr>
<tr>
<td>4</td>
<td>Inlet pump of decomposition tower</td>
<td>11</td>
<td>2</td>
<td>24</td>
<td>528</td>
</tr>
<tr>
<td>5</td>
<td>Sewage pump</td>
<td>7.5</td>
<td>2</td>
<td>2</td>
<td>30</td>
</tr>
<tr>
<td>6</td>
<td>Inlet pump of steam-stripping tower</td>
<td>11</td>
<td>2</td>
<td>24</td>
<td>528</td>
</tr>
<tr>
<td>7</td>
<td>Outlet pump</td>
<td>15</td>
<td>2</td>
<td>24</td>
<td>720</td>
</tr>
<tr>
<td>8</td>
<td>Reflux pump</td>
<td>1.5</td>
<td>2</td>
<td>24</td>
<td>72</td>
</tr>
<tr>
<td>9</td>
<td>Absorption circulating pump</td>
<td>18.5</td>
<td>5</td>
<td>24</td>
<td>2,220</td>
</tr>
<tr>
<td>10</td>
<td>Discharge pump</td>
<td>4</td>
<td>1</td>
<td>8</td>
<td>32</td>
</tr>
<tr>
<td>11</td>
<td>Mother liquid pump</td>
<td>4</td>
<td>1</td>
<td>8</td>
<td>32</td>
</tr>
<tr>
<td>12</td>
<td>Circulating pump for ammonia recovery</td>
<td>4</td>
<td>1</td>
<td>24</td>
<td>96</td>
</tr>
<tr>
<td>13</td>
<td>Pickling circulating pump</td>
<td>11</td>
<td>1</td>
<td>1</td>
<td>11</td>
</tr>
<tr>
<td>14</td>
<td>Agitator of crystal slurry tank</td>
<td>7.5</td>
<td>2</td>
<td>24</td>
<td>360</td>
</tr>
<tr>
<td>15</td>
<td>Agitator of mother liquid</td>
<td>7.5</td>
<td>1</td>
<td>24</td>
<td>180</td>
</tr>
<tr>
<td>16</td>
<td>Induced draft fan</td>
<td>15</td>
<td>1</td>
<td>24</td>
<td>360</td>
</tr>
<tr>
<td>17</td>
<td>Circulating pump for purification tower</td>
<td>4</td>
<td>1</td>
<td>24</td>
<td>96</td>
</tr>
<tr>
<td>18</td>
<td>Circulating pump for cooling water</td>
<td>45</td>
<td>1</td>
<td>24</td>
<td>1,080</td>
</tr>
<tr>
<td>19</td>
<td>Draught fan for cooling water</td>
<td>15</td>
<td>1</td>
<td>24</td>
<td>360</td>
</tr>
<tr>
<td>20</td>
<td>Air compressor</td>
<td>4.8</td>
<td>1</td>
<td>24</td>
<td>115.2</td>
</tr>
<tr>
<td>21</td>
<td>Vacuum absorption pump</td>
<td>37</td>
<td>1</td>
<td>24</td>
<td>888</td>
</tr>
</tbody>
</table>

**Total installed power** 8,268.2

### Figure 6 | Unit power consumption of the ammonia heat extraction system.
Compared with conventional MBR system, the comprehensive investment was higher of AHES, while the N resource revovery was an obvious advantages.

3.5. Utilization of recovered ammonia

Ammonia recovered from leachate could be utilized as follows: (1) nitrogen resource recovered in the form of ammonium bicarbonate from centrifuge could be marketed as commercial-grade ammonium bicarbonate; (2) ammonia obtained from the condenser of the selective non-catalytic reduction (SNCR) process of municipal solid waste incineration plant, because of the less negative impact thereof than urea on boiler efficiency (Gholami et al. 2020). Commercial ammonia used in the SNCR process usually needs to be diluted to 5–15 wt% to ensure that the ammonia spray gun has enough jet power and atomization effect. In this engineering project, the concentration of ammonia water obtained from condenser was 10 wt%, which was suitable for direct application in SNCR.

3.6. Pollution control for AHES

Exhaust gas mainly contained non-condensable gas and ammonia escape. These non-condensable components were treated via the process of ‘acid absorption + pickling purification + activated sludge purification’, and the purified gas was induced to

262.21 USD/t for NH₄HCO₃ recovery. Compared with conventional MBR system, the comprehensive investment was higher of AHES, while the N resource revovery was an obvious advantages.

### Table 6 | The total operation cost of AHES during the stable operation

<table>
<thead>
<tr>
<th>Name</th>
<th>Total consumption</th>
<th>Unit consumption</th>
<th>Unit price</th>
<th>Unit cost (USD /m³ leachate)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Defoamer consumption</td>
<td>2.85 t</td>
<td>0.045 kg/m³</td>
<td>2.36 USD/kg</td>
<td>0.11</td>
</tr>
<tr>
<td>2. Sulfamic acid consumption</td>
<td>8.20 t</td>
<td>0.130 kg/m³</td>
<td>1.02 USD/kg</td>
<td>0.13</td>
</tr>
<tr>
<td>3. CO₂ consumption</td>
<td>168.20 t</td>
<td>2.66 kg/m³</td>
<td>0.14 USD/kg</td>
<td>0.38</td>
</tr>
<tr>
<td>4. Steam consumption</td>
<td>5,040.00 t</td>
<td>80.00 kg/m³</td>
<td>0.013 USD/kg</td>
<td>1.00</td>
</tr>
<tr>
<td>5. Water consumption</td>
<td>2,520.00 t</td>
<td>40.00 kg/m³</td>
<td>0.55 USD/kg</td>
<td>0.02</td>
</tr>
<tr>
<td>6. Power consumption</td>
<td>167,639.00 kWh</td>
<td>2.66 kWh/m³</td>
<td>0.16 USD/kWh</td>
<td>0.42</td>
</tr>
</tbody>
</table>

Comprehensive operation cost
2.06

Note: the unit consumption was based on the total treatment capacity of 63,123 m³ leachate.
2–3 m below the aerobic section of the MBR system in the leachate treatment plant. Ammonia pollution seriously threatens human health and adversely impacts environmental security (Roman et al. 2013). Therefore, effective measures should be undertaken to control the escape of NH₃ during normal operations, maintenance, and accident conditions. Additionally, ammonia would escape from the processes of storage, dehydration, transportation, and packing of ammonium bicarbonate saturated solution, as well as the affiliated mother liquor pool in AHES. Correspondingly, the main control measures included absorption by water, sealing the devices, and wastewater collection.

During the operation of the decomposition tower, CO₃²⁻ was generated because of HCO₃⁻ dissociation. Subsequently, CO₃²⁻ interacted with Ca²⁺ and Mg²⁺ and formed precipitates (CaCO₃ and MgCO₃), which should be removed periodically. Besides, due to the high NH₃-N concentration (5,500–6,500 mg/L) of concentrated residue, appropriate measures should be implemented to inhibit odor emission. In this engineering project, a sedimentation tank and buffer tank were utilized. The concentrated residue was initially discharged to the sedimentation tank via pumping. The supernatant then flowed to the buffer tank through gravity and was then pumped to the collecting tank. The sludge at the bottom of the sedimentation tank was transported to the landfill for disposal.

Besides, drainage ditches equipped with drainage pipes were built around the AHES, which serviced the wastewater discharged from the maintenance process of devices. After that, the wastewater was disposed with leachate.

### 3.7. Implication and outlook

The imbalance of COD/TN is the critical limiting factor for mature leachate. The carbon source addition is necessary to realize the TN discharge standard by nitrification-denitrification processes, which is not only the high cost but also a lack of storage pool capacity. The innovation AHES developed in this study can solve the problem via the transformation of ammonia nitrogen into ammonium bicarbonate so that significantly reduce the nitrogen load. Moreover, the recovered ammonium bicarbonate product obtained was pure white crystal, which has the moisture and purity of 5% and 99.99%, respectively. Therefore, ammonium bicarbonate product has good quality and can be used as organic fertilizer. The results of the engineering project show that the AHES has demonstration significance and innovation for high ammonia wastewater treatment. While, the influence of raw leachate quality and secondary pollution on AHES operation stability should be emphasized.

### 4. CONCLUSIONS

High NH₃-N in mature landfill leachate and a low ratio of COD/NH₃-N result in operational difficulties and high treatment costs. This engineering project successfully recovers nitrogen resources from mature leachate by using AHES. (i) The results of the 30-days operation showed that NH₃-N concentrations of raw leachate were reduced from 1,305–2,485 mg/L to 207–541 mg/L, the removal rate was 62–90%. Thereby, the COD/NH₃-N ratio increased from 1.40–1.84 to 7.69–28.00 (with an average of 13.73), being a suitable range for biological nitrogen removal by the MBR system. (ii) Nitrogen resource was recovered via the formation of NH₄HCO₃ in the ammonia recovery tower with the introduction of CO₂. Based on the material balance of NH₄HCO₃ production, the mature leachate can offer 37% CO₂ consumption. (iii) The total operation cost of AHES was 2.06 USD per cubic leachate, equal to 1.52 USD/kg of NH₃-N removal. AHES could save at least 2.24 USD/m³ of carbon source cost in the subsequent MBR system. The innovation AHES developed in this study, free ammonia removed from leachate by forming NH₄HCO₃, which has various applications in industry and agriculture. In the engineering operation process for mature landfill leachate treatment, a relatively low unit operating cost of the MBR system with AHES pretreatment could reduce energy consumption prominently.

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### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

### REFERENCES


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