

STABILIZATION OF THE MUNICIPAL SOLID WASTE BY USING OF *EX SITU* SND AND *IN SITU* DENITRIFICATION BIOREACTOR LANDFILL IN A LONG-TERM OPERATION

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Abstract: The objective of this research was to investigate and compare stabilization of the municipal solid waste of *ex situ* simultaneous nitrification denitrification (SND)/*in situ* denitrification and *ex situ* nitrification/*in situ* denitrification bioreactor landfill in the long-term operations. Based on previous studies of 78 weeks, the two sets of laboratory-scale bioreactor landfill systems (an *ex situ* nitrification/*in situ* denitrification bioreactor and an *ex situ* SND/*in situ* denitrification bioreactor) continued to run up to weeks 105. The concentrations of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, $\text{NO}_2^-\text{-N}$, TN and COD in leachate were analyzed and monitored by standard methods. The results showed that *ex situ* SND/*in situ* denitrification was better than *ex situ* nitrification/*in situ* denitrification in the removal of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, TN and COD. The average effluent $\text{NH}_4^+\text{-N}$ concentration in F1 was 12.02 mg/L, and F2 was 8.20 mg/L. The average effluent $\text{NO}_3^-\text{-N}$ concentration in F1 was 542.80 mg/L, and F2 was 513.12mg/L. The average effluent TN concentrations in F1 was 555.27 mg/L, and F2 was 522.47 mg/L. The average effluent COD concentrations of F1 and F2 reached 153.10 mg/L and 116.99 mg/L. The concentrations of $\text{NH}_4^+\text{-N}$ and COD could meet the discharge standard established in the Standard for Pollution Control on the Landfill Site of Municipal Solid Waste (GB16889-2008), the COD concentration of F1 and F2 met the discharge standard (less than 100 mg/L) at week 104 and 100, respectively. But TN could not reach discharge standard because low $\text{NO}_3^-\text{-N}$ removal rate caused by the lack of carbon sources. Therefore, other effective measures or processes need to be adopted in order to remove $\text{NO}_3^-\text{-N}$ from landfill leachate.

INTRODUCTION

Landfill is one of the main treatment methods of domestic waste^[1]. However, the traditional landfill technology has some problems, such as long stabilization time, complex and difficult treatment of leachate components, low landfill gas production rate and difficult recycling^[2-4]. In view of the problems existing in traditional landfill, the bioreactor landfill technology with leachate recirculation as the main control measure came into being in the 1970s^[5]. Nitrogen removal bioreactor landfill technology that can

solve the problem of high ammonia nitrogen concentration of leachate from the source has attracted attention in the field of landfill and leachate treatment^[6-8].

One of the treatment scenarios for nitrogen removal was *ex situ* nitrification/*in situ* denitrification bioreactor landfill. The *ex situ* nitrification of leachate can be performed by various wastewater nitrification processes, such as SBR^[9-11], aerobic activated-sludge reactor^[12,13], and aged refuse bioreactor^[14,15]. The nitrified leachate is then recycled into the fresh-refuse bioreactor for

denitrification^[16]. Sun et al.^[17] combined the process of *ex situ* nitrification and *in situ* denitrification to remove nitrogen from municipal landfill leachate. The results showed that the average effluent ammonia concentration was 49 mg N L⁻¹, TN concentration was 93 ± 13 mg N L⁻¹ in landfill bioreactor. Wang et al.^[18] used the combined process of *ex situ* nitrification in an aged refuse bioreactor and *in situ* denitrification in a fresh refuse bioreactor, and found that after 146 days of operation, the effluent concentration of COD and NH₄⁺-N in fresh refuse bioreactor were 613 mg/L and 97.34 mg/L, respectively. The results of Zhong et al.^[19] showed that the COD gradually decreased to about 2000 mg/L, effluent ammonia of the denitrification column was higher than 300 mg N L⁻¹ about 250 days. The above researches indicated that *ex situ* nitrification/*in situ* denitrification bioreactor landfill has good nitrogen removal effect. But the COD, NH₄⁺-N or TN did not meet the discharge standards (less than 100, 25 or 40) at the end of the experiment. Therefore, *ex situ* nitrification/*in situ* denitrification should continue to be studied for a long time. Meanwhile, it has been reported that the high concentration nitrate produced in *ex situ* nitrification phase inhibit the methane production of refuse *in situ* denitrification bioreactor.

In order to solve the above problems, *ex situ* simultaneous nitrification denitrification (SND)/*in situ* denitrification bioreactor landfill process has been conducted^[20,21]. The results showed that the *ex situ* SND/*in situ* denitrification process obtained a better leachate removal performance compared with the *ex situ* nitrification/*in situ* denitrification process, and the *ex situ* SND process can reduce the energy costs. Based on the experimental data from week 4 to week 30, it is predicted that NH₄⁺-N concentration would reach less than 25 mg/L after 63 weeks in *ex situ* nitrification/*in situ* denitrification (in F1) and 40 weeks in *ex situ* SND/*in situ* denitrification (in F2), the COD concentrations of leachate in F1 and F2 were expected to meet the discharge standard (less than 100 mg/L) after weeks 41 and 34, respectively^[20]. Effluent TN concentrations of leachate in F1 and F2 were expected to meet the

discharge standard (less than 40 mg/L) after weeks 54 and 35, respectively^[21]. However, the NH₄⁺-N concentration in F1 and F2 reached 52.71 mg/L and 54.74 mg/L at the predicted time. The COD concentration in FRBs failed to reach below 100 mg/L, F1 and F2 was about 696.96 mg/L and 396 mg/L at the predicted time, respectively. Although the TN concentration in FRBs reach below 40 mg/L, F1 and F2 was about 39.16 mg/L and 20.06 mg/L at the predicted time, respectively, TN concentration fluctuated in this stage and did not reach the standard discharge at week 78^[21]. Since the concentration of COD and NH₄⁺-N did not reach the standard in the previous studies, the experiment has been continued.

In this study, *ex situ* SND and *in situ* denitrification bioreactor landfill was investigated in a long-term operation lasted for 105 weeks based on previous study. The purpose is: (1) to investigate whether and when the effluent can meet the discharge standard established in the Standard for Pollution Control on the Landfill Site of Municipal Solid Waste (GB16889-2008). (2) Evaluate and compare the effects of *ex situ* nitrification/*in situ* denitrification and *ex situ* SND /*in situ* denitrification on landfill leachate.

MATERIALS AND METHODS

Fresh and aged refuse

Fresh refuse was obtained from a waste transfer station near Guilin University of Technology, Guilin, Guangxi, China. Sample compositions were adjusted based on the investigations of average composition of MSW made in Guilin City. These compositions comprised (by wet weight) organics (51.13%), paper (14.99%), plastic (4.45%), metal (0.14%), glass (2.84%), wood (2.30%), and others (24.16%). The large samples were cut into approximately 2-5 cm and were then mixed uniformly.

The 10-years old aged refuse utilized was obtained from the Chongkou Sanitary Landfill in Guilin, Southwest China. Non-biodegradable materials, such as stones, metals, glass, and plastic bags, were removed.

The fine fraction resembling soil with diameters of 4 -10 mm was selected because this refuse is an ideal bioreactor material. The properties of the fresh refuse and aged refuse were the same as those used in Sun et al. and Li et al.^[20,21].

Experimental setup

A schematic diagram of the experimental setup is presented in Fig. 1(a). Two fresh refuse bioreactors (FRBs), namely, F1 and F2, were made of a stainless steel cylinder with an inner diameter of 340 mm and height of 650 mm. Two aged refuse bioreactors (ARBs), namely, A1 and A2, were made of a polyvinyl chloride cylinder with an inner diameter of 390 mm and a height of 1000 mm. F1 and F2 were loaded from bottom to top with 50 mm gravel (<10 mm in diameter) followed by 400 mm fresh refuse and topped with 50 mm gravel (<10 mm in diameter). Meanwhile, A1 and A2 were loaded from bottom to top with 50 mm gravel (<10 mm in diameter) followed by 800 mm aged refuse and topped with 50 mm small gravel (<10 mm in diameter).

A combined process comprising *ex situ* nitrification (or SND) in an aged refuse bioreactor and *in situ* denitrification in a fresh refuse bioreactor was constructed. The leachate from F1 and F2 was introduced into A1 and A2, while the effluent from A1 and A2 was recirculated into F1 and F2 once per day, respectively. F1 and A1 constituted the *ex situ* nitrification and *in situ* denitrification process (Fig. 1(b)), while F2 and A2 constituted the *ex situ* SND and *in situ* denitrification process (Fig. 1(c)).

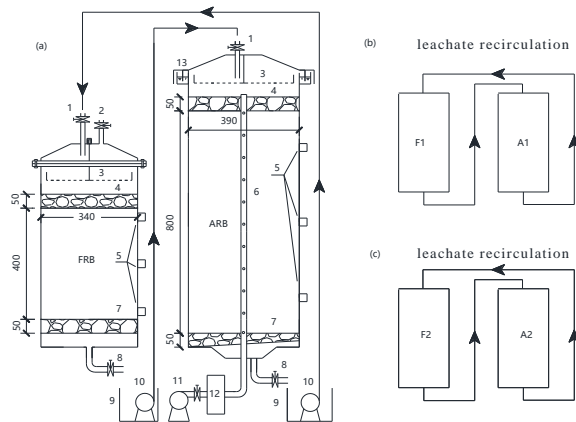


FIG. 1 The schematic of bioreactor landfill for nitrogen removal

1. leachate inlet 2. gas valve 3. liquid distributor 4. gravel layer 5. sample collection 6. perforated vent pipe 7. gravel layer 8. outlet valve 9. storage water tank 10. water pump 11. air compressor 12. gas flowmeter 13. sealing cover

Experimental operations

In this experiment, F1 and F2 were operated as *in situ* denitrification reactors. To secure the anaerobic conditions, the sealing cover was placed on top of the FBRs and the outlet valve at the bottom was closed. A1 was operated as an *ex situ* nitrification reactor. Based on the aeration rate of $2.01 \text{ m}^3/(\text{m}^3 \text{ refuse} \cdot \text{d})$ and aeration time of 8 h per day in the preliminary experiment^[22], the aeration rate of A1 in this study was set to $6.03 \text{ m}^3/(\text{m}^3 \text{ aged refuse} \cdot \text{d})$ for aeration time extended three times and was 24 h per day to guarantee the nitrification condition. The airflow rate of ARBs was measured by using the rotor flowmeter (LZB-3WB, KEDE Instrument Ltd., China) with a range of 40 ml/min to 400 ml/min, which was regularly calibrated with a soap bubble flowmeter (GILBRATOR-2, Sensidyne LP, American). A2 was operated as an *ex situ* SND reactor. Based on the effluent leachate quality and aeration rate in our previous experiment^[20,21], the aeration rate was set to $0.09 \text{ m}^3/(\text{m}^3 \text{ aged refuse} \cdot \text{d})$ and the aeration time was set to 3 h.

Experimental analysis

Leachate samples of approximately 50 mL were used for the analysis every week. The $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, $\text{NO}_2^-\text{-N}$, and COD of the leachate samples were analyzed according to the standard methods set by standard for pollution control on the landfill site of MSW (Ministry of Environmental Protection of the People's Republic of China, 2008). $\text{NH}_4^+\text{-N}$ was measured by using the Nessler's reagent spectrophotometric method, $\text{NO}_3^-\text{-N}$ was measured by using the ultraviolet spectrophotometric method, and $\text{NO}_2^-\text{-N}$ was measured by using the spectrophotometric method. COD was

measured by using the potassium dichromate volumetric method. The samples were stored in glass bottles at temperatures below 4 °C. The leachate samples were then filtered by using a qualitative filter paper, and the filtered samples were measured in triplicate.

RESULTS AND DISCUSSION

The data for weeks 1 to 30 of the experiment were obtained from the operation of bioreactors in the early study^[20]. The data for weeks 31 to 78 of the experiment were obtained from the operation of bioreactors in the mid-stage study^[21]. This experiment was conducted for weeks 85 to 105 as a late study.

$\text{NH}_4^+\text{-N}$

Fig. 2 shows the changes of leachate $\text{NH}_4^+\text{-N}$ concentration during the operation of the bioreactor landfill system. According to the leachate $\text{NH}_4^+\text{-N}$ concentration were fitted based on weeks 4 to 30, the $\text{NH}_4^+\text{-N}$ concentrations of leachate in F1 and F2 were expected to meet the discharge standard (less than 25 mg/L) after weeks 63 and 40, respectively^[20]. However, the $\text{NH}_4^+\text{-N}$ concentration in F1 and F2 exceeded emission standards and reached 52.71 mg/L and 54.74 mg/L at the predicted time, respectively^[21]. The $\text{NH}_4^+\text{-N}$ concentration only met the standard for pollution control on the landfill site of MSW (Ministry of Environmental Protection of the People's Republic of China, 2008) in F1 at weeks 49-52 and had an average effluent value of 19.29 mg/L, while the $\text{NH}_4^+\text{-N}$ concentration in F2 cannot reach below 25 mg/L and obtained a minimum value of 28.01 mg/L in week 49. According to Fig. 2(a), at the end of 78 week, the effluent concentration of $\text{NH}_4^+\text{-N}$ in FRBs did not meet the discharge standard, F1 was 48.03mg /L, F2 was 53.11mg /L. Fig. 2 (b) summarize the changes of $\text{NH}_4^+\text{-N}$ concentration reactor in this experiment. The concentrations of $\text{NH}_4^+\text{-N}$ in F1 and F2 during 85-105 weeks were both less than 25 mg/L, which met the landfill pollution control standards (Ministry of Environmental Protection of the People's Republic of China, 2008). The average effluent $\text{NH}_4^+\text{-N}$

concentration in F1 was 12.02 mg/L, while that of F2 was 8.20 mg/L. The $\text{NH}_4^+\text{-N}$ concentration in F2 was 5.57 mg/L, which was lower than that in F1 (3.85 mg/L) at the end of the experiment. The results suggested that *ex situ* SND/*in situ* denitrification was more effective than *ex situ* nitrification/*in situ* denitrification in eliminating ammonia.

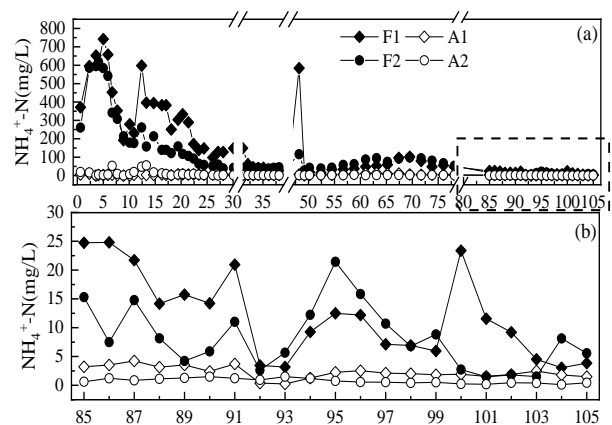


FIG. 2 Removal performance of $\text{NH}_4^+\text{-N}$ from leachates.

$\text{NO}_3^-\text{-N}$

The changes of $\text{NO}_3^-\text{-N}$ concentration during the operation is shown in Fig.3. $\text{NO}_3^-\text{-N}$ in the FRBs were relatively stable within 50-78 weeks, and the concentration was less about 30mg/L. The results showed that the FRBs had an excellent denitrification effect. Compared with week 50-78, the $\text{NO}_3^-\text{-N}$ concentration of the A reactor in 85-105 week increased significantly. This may be that the leachate contained high concentration of $\text{NH}_4^+\text{-N}$ in the early stage of the test, and the aged refuse adsorbed a large amount of $\text{NH}_4^+\text{-N}$ and converted into $\text{NO}_3^-\text{-N}$. With the progress of the test, the $\text{NH}_4^+\text{-N}$ concentration in the influent of the aged refuse reactor gradually decreased, and the saturated aged refuse desorbed, resulting in the increase of $\text{NO}_3^-\text{-N}$ concentration in the effluent of the aged refuse reactor. At the end of the experiment, the average effluent $\text{NO}_3^-\text{-N}$ concentration in reactor A2 was 533.41

mg/L, which was significantly lower than that of A1 (712.82 mg/L). Nitrogen removal by biological methods was divided into two stages, namely aerobic nitrification of ammonia to nitrate or nitrite and anaerobic/anoxic denitrification of nitrate to nitrogen^[10]. Due to the denitrification of A2 under low aeration conditions, NO_3^- -N produced by nitrification was reduced, so that the NO_3^- -N concentration of A2 was lower than A1. The average effluent NO_3^- -N concentration in reactor F2 was 513.12mg/L, which was lower than that of F1 (542.80 mg/L). This may be due to the lack of carbon sources leading to the difficulty of denitrification of NO_3^- -N from ARBs, resulting in the increase of NO_3^- -N concentration in FRBs^[23,24]. This was also the reason for the increase in TN concentration in Fig. 5.

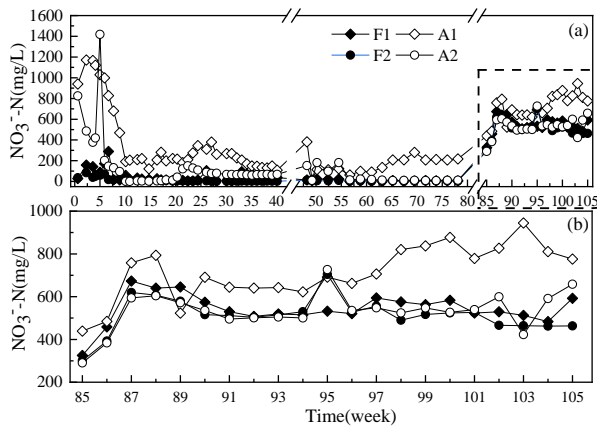


FIG. 3 Variation of NO_3^- -N concentration in leachate

NO_2^- -N

The changes of NO_2^- -N concentration during the operation of the bioreactor is shown in Fig.4. The effluent NO_2^- -N concentration of A1 and A2 reactors remained at a low level during 50-78 weeks. The concentration of NO_2^- -N in this experiment was also relatively low compared with that at 50-78 weeks. At the end of the experiment, the concentration of NO_2^- -N in A2 was slightly higher than that in A1, the average concentration of NO_2^- -N in A1 was 0.69 mg/L, and that in A2 was 0.85mg/L. These findings can be attributed to

the short-cut denitrification of A2 under low aeration conditions^[25]. The average concentration of NO_2^- -N in F1 was 0.45 mg/L, and that in F2 was 1.14mg/L. Nag et al.^[26]believed that the formation of nitrite was confirmed in the process of denitrification, which could stimulate the conversion of NO_3^- -N to N_2 gas under low aeration conditions.

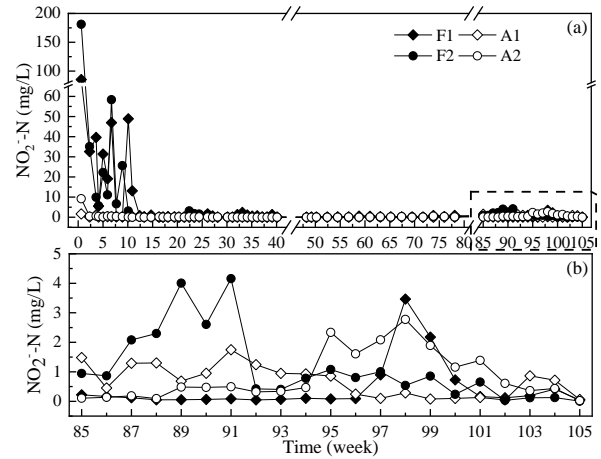


FIG. 4 Variation of NO_2^- -N concentration in leachate

TN

The variation of TN concentration in leachate during the entire operation of the bioreactor is shown in Fig. 5. TN could meet the 25 mg/L standard of the Chinese Emission Standards for pollution control on MSW landfill sites (GB16889-2008) at the predicted time. Specifically, F1 and F2 were 39.16 and 20.06 mg/L after 54 and 35 weeks, respectively. However, TN concentration fluctuated and did not reach the emission standards for a long time. The effluent TN concentration of leachate in ARBs was higher than that of influent in 85-105 week. These findings may be due to organic matter reaching adsorption saturation at this stage, and some pollutants were washed away^[27]. At the end of the test, the average effluent TN concentrations of F1 and F2 reached 555.27 mg/L and 522.47 mg/L, respectively. Sufficient carbon source was the key for denitrifying

bacteria to remove nitrogen efficiently^[28-30]. The poor removal effect of TN may be because the ammonia nitrogen in the influent generates nitrite and nitrate in ARBs, but only a small part of nitrogen was metabolized by microorganisms, and most of it was still retained in the leachate. Nitrate cannot be denitrified due to the lack of carbon source after recirculation to the FRBs, resulting in nitrate accumulation. This can be verified from Fig.3.

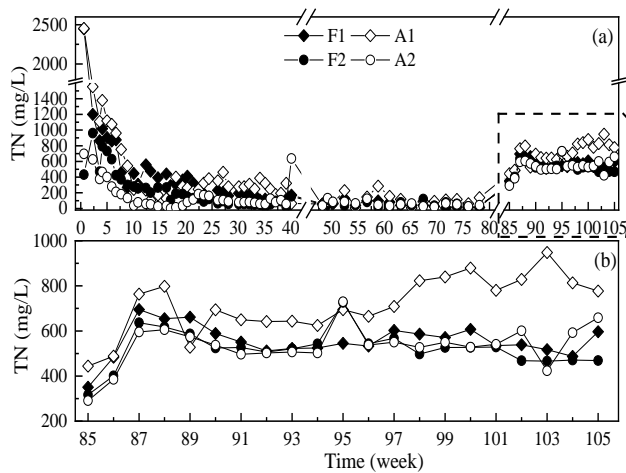


FIG. 5 Removal performance of TN concentration in leachate

COD

The change of leachate COD concentration during the whole operation of the bioreactor is shown in Fig. 6. The COD concentration of *ex situ* SND/*in situ* denitrification was significantly lower than that of *ex situ* nitrification/*in situ* denitrification. From week 1 to 30, The COD concentration of each bioreactor landfill showed a trend of fluctuation and decline. At 31-105 weeks, COD concentration is basically around 100-300 mg/L in FRBs (Fig. 5). Although the COD concentration in F1 and F2 failed to reach the discharge standard at the expected time after weeks 41 and 34^[21], the COD concentration of F1 and F2 can meet the discharge standard (less than 100 mg/L) in the Standard for Pollution Control on the Landfill Site of Municipal Solid

Waste (GB16889-2008) at week 104 and 100, respectively. The average effluent COD concentrations of F1 and F2 reached 153.10 mg/L and 116.99 mg/L, respectively. At week 105, the COD concentration of F1 and F2 reached 92.11 mg/L and 64.32 mg/L, respectively. Results of the experiment indicate that the COD concentrations of leachate from F2 decreased more rapidly than that those from F1. Therefore, *ex situ* SND/*in situ* denitrification can accelerate the degradation of the refuse.

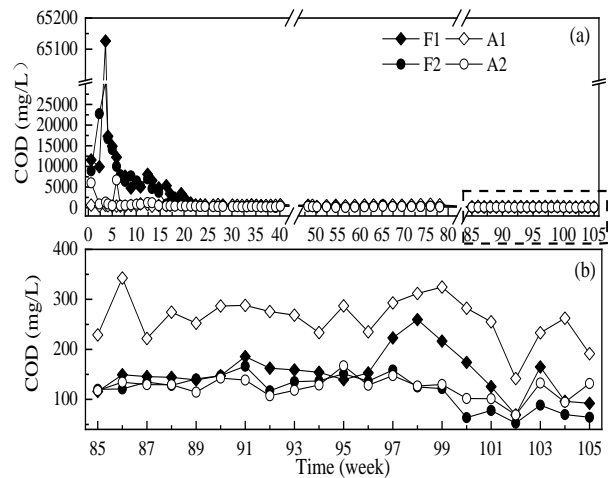


FIG. 6 Removal performance of COD in leachate

CONCLUSION

Compared with the *ex situ* nitrification/*in situ* denitrification process, the *ex situ* SND/*in situ* denitrification process obtained lower concentrations of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, TN and COD in the experiment. The $\text{NH}_4^+\text{-N}$ concentration in F2 was 5.57 mg/L, which was lower than that in F1 (3.85 mg/L) at the end of the experiment. The COD concentration in F2 was 64.32 mg/L, which also was lower than that in F1 (92.11 mg/L). *Ex situ* SND/*in situ* denitrification process could effectively remove the $\text{NH}_4^+\text{-N}$ and COD concentration in leachates. The $\text{NH}_4^+\text{-N}$ and COD concentrations could meet the discharge standard established in the Standard for Pollution Control on the Landfill Site of Municipal Solid Waste (GB16889-2008). *Ex situ* nitrification/*in*

situ denitrification and *ex situ* SND/*in situ* denitrification could accelerate the stabilization of refuse, and *ex situ* SND/*in situ* denitrification was better. However, the treatment performance of TN this process was unsatisfactory. Further research is required to develop effective measures or processes that can effectively treat leachates with low high concentrations of NO_3^- -N.

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