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Continuous-flow combined process of nitritation and ANAMMOX for treatment of landfill leachate

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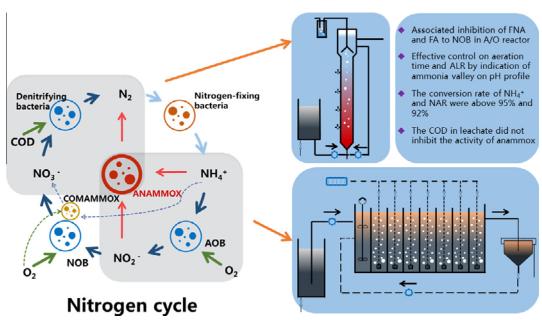
HIGHLIGHTS

- A continuous-flow process of nitritation and ANAMMOX was used to treat leachate.
- The mature landfill leachate without diluted by tap-water was used in this study.
- Associated inhibition of FA and FNA to NOB facilitate realization of nitritation.
- The conversion rate of NH_4^+-N and NAR in A/O were up to 95% and 92%, respectively.
- Quantitative PCR analyzed the proportions of AOB, NOB and anammox in A/O and UASB.

GRAPHICAL ABSTRACT



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ABSTRACT

Due to the difficulty in removing nitrogen from landfill leachate, a combined continuous-flow process of nitritation and anammox was applied to process mature leachate. The transformation rate of ammonia and nitrite accumulation ratio in A/O reactor were kept above 95% and 92% respectively through associated inhibition of free ammonia (FA) and free nitrous acid (FNA) to NOB. The total nitrogen volumetric load of anammox in an UASB reactor was brought up from 0.5 kg/(m^3·d) to 1.2 kg/(m^3·d) by gradually increasing influent substrate concentration and reducing hydraulic retention time (HRT). The results show that COD from mature leachate did not bring obvious inhibition effects to anammox. Under concentrations of influent ammonia and COD which were respectively 1330 mg/L and 2250 mg/L, the removal efficiencies of TN and COD reached 94% and 62% respectively. In the quantitative PCR reactions, the proportions occupied by AOB, NOB and anammox in A/O were 11.39%, 1.76% and 0.05% respectively; and proportions of those in UASB were 0.35%, 4.01% and 7.78% respectively.

1. Introduction

Disposal of solid waste in landfills is used most commonly in waste management, but generation of leachate cannot be avoided (Berge et al., 2007). The treatment of sanitary landfill leachate is one of the major obstacles for scientific community mainly due to the huge contents of ammonia and recalcitrant organic substances (Kulikowska and Klimiuk, 2008). Hence, it is necessary to develop an effective and cost-saving removal method so as to increase the approval degree of leachate treatment and realize its development.

At present, landfill leachate is processed mainly by physico-chemical and biological processes. Ammonium stripping (Zhang et al., 2012), reverse osmosis (Talalaj and Biedka, 2015) and chemical precipitation (Huang et al., 2016) are the commonest

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physicochemical processes for processing of leachate. However, the high cost caused by the huge chemical and energy consumption greatly hinders its practical applications. As a result, the biological processes such as the traditional nitrification–denitrification (Peng et al., 2008) and anammox (Miao et al., 2015) process become the feasible method because of high cost performance and high efficiency.

Characteristics of leachate vary greatly with the burial time. Leachate becomes "mature" after being buried for over 10 years (Singh and Tang, 2013), while the mature leachate contains a low concentration of biodegradable substances (BOD $\leqslant$ 500 mg/L), high amount of total nitrogen (above 1000 mg/L) and low ratio of BOD_5 to COD (below 0.1). However, the conventional nitrification–denitrification process usually shows low efficiency during processing of the mature leachate, wherein the organic carbon is insufficient for complete denitrification. Thus, the anammox progress is more suitable for the mature leachate treatment because of its chemoautotrophy and minimal surplus sludge production (Jetten et al., 2001). More than 100 wastewater treatment projects which take anammox as the biological nitrogen removal method have been practiced to treat N-rich wastewater such as leachate and digested water (Lackner et al., 2014). Nitritation and anammox combined process is a complete autotrophic nitrogen removal process which reduces energy consumption and requires no external carbon source in comparison with the conventional nitrification–denitrification process. (Ma et al., 2016)

However, realization of the combined nitritation and anammox process is mainly hindered by realization of nitritation. At present, some scholars have researched some operational conditions which may inhibit the growth of NOB more obviously in comparison with that of AOB, such as FA (free ammonia), FNA (free nitrous acid), low DO (dissolved oxygen) and high pH (Anthonisen et al., 1976; Rongsayamanont et al., 2014; Gu et al., 2012). Due to the high ammonia concentration, the nitritation realized in a continuous reactor has been discussed in the previous studies under inhibition of FA and FNA (Peng et al., 2008; Wu et al., 2015). In addition, in comparison with a one-stage process, a two-stage process is advantageous by the high nitrogen removal rate (NRR). High-rate performance with NRR of 76.7 kg N/(m^3·d) was reached in the lab-scale UASB reactors (Tang et al., 2011) in comparison with 0.06–1.80 kg N/(m^3·d) in a one-stage reactor (Van Hulle et al., 2010). The anammox reactor was separated from the nitritation reactor, so that the activity of anammox could be enhanced as no inhibition from dissolved oxygen existed in the nitrification reactor (Li et al., 2011).

With above researches as the reference, this study adopted mature leachate which was obtained from a landfill in Beijing; an anoxic/aerobic (A/O) reactor and an up-flow anaerobic sludge blanket (UASB) were combined. The A/O treatment reduces COD and NH_4^+-N, while nitritation takes place in the A/O reactor. Further nitrogen removal is achieved in the subsequent UASB via anammox. By refluxing the effluent from the UASB-anammox to the A/O, the experiment investigated the removal of organic matter and nitrogen, partial nitrification, anammox and the key factor for realization of nitritation in the A/O reactor. By virtue of this technology, deep-level removal of organic matters, NH_4^+-N and total nitrogen (TN) can be accomplished simultaneously without addition of any carbon source.

2. Materials and methods

2.1. Experimental apparatuses

Fig. 1 shows an experimental system, which comprises an A/O and an UASB. The A/O and UASB reactors were made of Plexiglas, wherein their effective capacities were 10.5 L and 5.5 L respectively. The A/O reactor was divided equally into 8 cells, wherein the first cell was used for anoxic treatment and the rest ones were used for aerobic treatment.

2.2. Influent and inoculums

The mature landfill leachate collected from the Liulitun Municipal Solid Waste (MSW) Sanitation Landfill Site (Beijing, China) was used as a feeding solution and it was preserved at 4 °C to prevent the natural degradation of organics. The main characteristics of the influent were shown in Table 1.

In the increase of nitrogen loading of UASB-anammox, synthetic wastewater was used as the influent of the UASB reactor. The composition of the synthetic wastewater contained NH_4^+-N ($(NH_4)_2SO_4$), NO_2^--N ($NaNO_2$) and a medium solution. The ratio of influent NH_4^+-N to NO_2^--N was kept at 1:1.32. The medium solution contained the following nutrients (/L): 0.03 g of KH_2PO_4, 0.14 g of $CaCl_22H_2O$, 0.5 g of $KHCO_3$, 0.14 g of $MgSO_47H_2O$, and 1.0×10^3 L of trace element solutions A and B according to van de Graaf et al. (1996). The trace element solution A contained (/L): 5 g of EDTA and 5 g of $FeSO_4$; and the trace element solution B contained (/L): 15 g of EDTA, 0.43 g of $ZnSO_47H_2O$, 0.24 g of $CoCl_26H_2O$, 0.99 g of $MnC1_24H_2O$, 0.25 g of $CuSO_45H_2O$, 0.22 g of $NaMoO_42H_2O$, 0.19 g of $NiC1_26H_2O$, 0.21 g of $NaSeO_410H_2O$, and 0.014 g of H_3BO_4.

The sludge obtained through the lab-scale nitrification SBR and used for the treatment of mature landfill leachate was taken as the inoculum for the A/O reactor. Concentration of the mixed liquor suspended solids (MLSS) was 3500S) w mg/L.

2.3. Analysis methods

2.3.1. Conventional analysis items

Values of pH, DO and temperature were monitored using a pH/Oxi 340i analyzer (WTW Company, Germany). Values of MLSS, MLVSS, NH_4^+-N, NO_3^--N, NO_2^--N and COD were measured according to the standard methods (APHA, 1995). Total nitrogen was analyzed by a TN/TOC analyzer (MultiN/C3000, Analtikjena A G, Germany).

2.3.2. DNA isolation and polymerase chain reaction (PCR)

2.3.3.1. Quantitative PCR. Abundances of all the bacterial and anammox DNAs were determined by real-time PCR using an MX3000P Real-Time PCR system (Stratagene, La Jolla, CA) and a fluorescent dye SYBR-Green approach. The primers for all the bacteria and anammox in the real-time PCR were 341f-534r and Amx368f-Amx820r. The primers for Nitrospira, Nitrobacter and AOB in the real-time PCR were 338f-685r, FGPS872f-FGPS1269r and amoa1f-amoa2r, respectively. The amplification was performed in the 20 μL reaction mixtures which consist of SYBR Green exTaq (10 lL, Takara, Dalian, China), ROX Reference Dye 50 (0.3 μL), each primer (0.3 μL, 10 mmol/L), and DNA template (2 μL, 1–10 ng). The program is completed by the following steps: first step for 3 min at 95 °C, second step with 40 cycles for 30 s at 95 °C, third step for 30 s at 56 °C, and fourth step for 30 s at 72 °C.

2.3. Calculation

The efficiency of NH_4^+-N, NO_2^--N, NO_3^--N, TN and COD removal was calculated according to Eq. (1):

$$\text{Efficiency } (\%) = \frac{C_{inf}\ (mg/L) - C_{eff}\ (mg/L)}{C_{inf}(mg/L)} \times 100 \quad (1)$$

where: C_{inf} refers to the concentration of NH_4^+-N (or NO_2^--N and NO_3^--N, etc.) in the influent and C_{eff} refers to the one in the effluent.