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Advanced nitrogen removal from landfill leachate using real-time controlled three-stage sequence batch reactor (SBR) system

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HIGHLIGHTS

- A novel three-stage SBR process was firstly developed to treat landfill leachate.
- The real landfill leachate without diluted by tap-water was used in this study.
- The ratio of $NO_2^-:NH_4^+$ in the Anammox influent was not needed to be adjusted.
- The modified continuous filling mode minimized the nitrite inhibition effects.
- Quantitative PCR analysis of Anammox showed Anammox gene ratio was less than 5%.

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ABSTRACT

A three-stage sequencing batch reactor (SBR), comprising pretreating SBR (SBR_{pre}), nitritation SBR (SBR_{ni}), and anaerobic ammonium oxidation (Anammox) SBR (SBR_{ana}), was developed for the nitrogen removal from mature landfill leachate. The concentrations of ammonia and chemical oxygen demand (COD) in the leachate were 2000 ± 100 and 2200 ± 200 mg/L, respectively. About 100 mg/L of organic substance was removed from SBR_{pre} to reduce the negative effect on the Anammox process under real-time control. After acclimation for 40 days, the nitrite to nitrogen oxide ratio (NO_2^-/NO_x) in SBR_{ni} was above 0.95. The nitrogen removal efficiency reached 90% in SBR_{ana}, and nitrogen load rate and nitrogen removal rate were 0.81 and 0.76 kg N/(m^3 d), respectively. The continuous filling process was used to avoid the nitrite inhibition on the Anammox activity. The quantitative PCR analysis of Anammox indicated the average Anammox gene ratio increased from 0.23% to 4.77% after 220 days operation.

1. Introduction

Landfill leachate contains high concentrations of organic/inorganic contaminants and may cause severe environmental pollution if not treated properly. There are three classes of landfill leachate in terms of landfill age: the young, middle-age, and mature landfill leachate (Bernard et al., 1997). Mature landfill leachate contains a relatively high concentration of ammonia and low concentration of biodegradable organic substances. In general, the ammonia concentration is above 1000 mg/L, chemical oxygen demand (COD) is below 3000 mg/L, and a ratio of biological oxygen demand (BOD_5) to COD is below 0.1 (Chen, 1996; Kulikowska and Klimiuk, 2008). The low C:N ratio and low biodegradability of landfill leachate poses high challenges for treatment. The biological treatment (e.g. nitrification/denitrification) is used as the main treatment for landfill leachate (Renou et al., 2008). However, for the mature landfill leachate with low C:N ratio, the conventional nitrification/denitrification process needs an external carbon source, which makes the treatment systems complicated and expensive.

Anaerobic ammonium oxidation (Anammox) is a cost-effective process with a great potential system (Kalyuzhnyi et al., 2006; Mulder et al., 1995; Kartal et al., 2010). Compared to conventional nitrification/denitrification processes, Anammox does not need external carbon sources and the power consumption for nitritation-Anammox process can be reduced 50% substantially. Even though Anammox is suitable for the treatment of mature landfill leachate with a low carbon-to-nitrogen (C/N) ratio (Shen et al., 2012; Chamchoi et al., 2008; Ganigué et al., 2009), there are some obstacles for real-world application. First, biodegradable organic substances have an adverse effect on Anammox (Chamchoi et al., 2008; Dapena-Mora et al., 2007; Molinuevo et al., 2009), thus nitritation and COD removal should be achieved prior to Anammox. Second, the doubling time of Anammox bacteria is longer than 11 days (Schmidt et al., 2003), so that long sludge retention time

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(SRT) is needed to prevent the loss of Anammox sludge (Chamchoi and Nitisoravut, 2007). Third, a high concentration of nitrite inhibits Anammox (Strous et al., 1999; Lotti et al., 2012). The activity of Anammox decreases when the nitrite concentration in the landfill is higher than 100 mg/L (Strous et al., 1999), and operational modes should be modified to alleviate the adverse effects of nitrite.

Anammox has been studied for nitrogen removal from mature landfill leachate (Liang and Liu, 2008), and was coupled with partial nitritation (PN) in an upflow anaerobic sludge blanket (UASB) (Liu et al., 2010; Sri Shalini and Joseph, 2012; Anfruns et al., 2013). Although the organic substances can be removed by PN, the ratio of nitrite to ammonia suitable for Anammox could not be maintained. Until now, the studies on the Anammox process for the treatment of mature landfill leachate either used diluted landfill leachate (Liu et al., 2010), or employed complicated operational modes to maintain the ratio of NO_2^-:NH_4^+ (Anfruns et al., 2013). Therefore, it is critical to investigate the advanced nitrogen removal process for the treatment of real mature landfill leachate without diluting or adding extra chemicals.

Sequencing batch reactor (SBR) has distinct advantages of space-saving, flexible operational mode, and auto-control capability (Chamchoi and Nitisoravut, 2007). It has been extensively used for biological nutrient removal (BNR) in municipal and industrial wastewaters (Wang et al., 2013). However, only a few researches applied SBR for Anammox process. In fact, SBR is suitable for Anammox enrichment due to its simplicity, efficient biomass retention, high stability over a long period of operation, and good BNR efficiency under substrate-shortage condition (Chamchoi and Nitisoravut, 2007; Strous et al., 1998). Therefore, this study aimed at developing a novel three-stage SBR process, comprising pretreatment SBR (SBR_{pre}), nitritation SBR (SBR_{ni}), and Anammox SBR (SBR_{ana}) for nitrogen removal from real mature landfill leachate. Specifically, the biodegradable organic substances were removed in the SBR_{pre} to reduce the negative effect on the Anammox process. Ammonia was oxidized to nitrite in the SBR_{ni}, and the effluents of the SBR_{pre} and SBR_{ni} were mixed together for the SBR_{ana}. In addition, the pH profiles and redox potential (ORP) profiles that could distinguish different operational stages (e.g. nitrification, dinitrification) in SBR cycles (Peng et al., 2008) were used for real-time control to achieve high Anammox efficiency and save operational energy. Finally, the Anammox microbial communities were determined using quantitative PCR and correlated with nitrogen removal efficiency and SBR operational conditions.

2. Methods

2.1. Experimental setup and operational procedure

The three-stage SBR system, comprising SBR_{pre}, SBR_{ni}, and SBR_{ana}, was made of polymethyl methacrylate with a total volume capacity of 39 L, distributed as 12, 12, and 15 L for the SBR_{pre}, SBR_{ni}, and SBR_{ana}, respectively (Fig. 1). The working volume capacities of the SBR_{pre}, SBR_{ni}, and SBR_{ana} were 10, 10, and 13 L, respectively (Fig. 1). The SBR_{pre} was equipped with pH meters, mechanical stirrers, and air diffusers. The SBR_{ni} was equipped with pH meters and air diffusers. The SBR_{ana} was equipped with a pH meter and a mechanical stirrer. An oxidation–reduction potential (ORP) meter was installed in the SBR_{pre} to monitor the aeration/anoxic status. During the aeration period, the aeration intensity was maintained at 100 L/h. The operational temperatures for SBR_{pre}, SBR_{ni} and SBR_{ana} were maintained at 25 °C, 25 °C and 35 °C using a temperature controller.

In the three-stage SBR system, the SBR_{pre} and SBR_{ni} were operated under the traditional mode: filling-aeration-settle-decant (5 h), whereas the SBR_{ana} was operated under the modified mode with continuous filling (5 h). The influent of the SBR_{ana} was the mixed effluents from the SBR_{pre} and SBR_{ni} based on a ratio of NO_2^-—N/NH_4^+—N of 1.3 (Fig. 1). The SBR_{ana} was operated under the modified mode: continuous filling (5 h) and stirring until the completion of the reaction. The terminal point of the Anammox was determined using the pH profile. The exchange volumetric rate of the SBR_{ana} was found to be 38%. The main operational conditions of the three SBRs in each period are shown in Table 1.

2.2. Influent and seed sludge

During the acclimation period, the SBR system was fed with a synthetic wastewater consisting of KH_2PO_4 (10 mg/L), $CaCl_2{\cdot}2H_2O$ (5.6 mg/L), $MgSO_4{\cdot}7H_2O$ (300 mg/L), $KHCO_3$ (1250 mg/L), trace element solution I (EDTA 5000 mg/L and $FeSO_4$ 5000 mg/L), and trace element solution II (EDTA 1000 mg/L, H_3BO_4 14 mg/L, $MnCl_2{\cdot}4H_2O$ 990 mg/L, $CuSO_4{\cdot}5H_2O$ 250 mg/L, $ZnSO_4{\cdot}7H_2O$ 430 mg/L, $NiCl_2{\cdot}6H_2O$ 190 mg/L, $NaSeO_4{\cdot}10H_2O$ 210 mg/L, $NaMoO_4{\cdot}2H_2O$ 220 mg/L) (Sliekers et al., 2002). $NaNO_2$ and NH_4Cl solutions were added to supply nitrite and ammonium for Anammox activities. The pH of the influent was controlled at 7.5 ± 0.2.

After the acclimation period, the landfill leachate collected from the Liulitun Municipal Solid Waste (MSW) Sanitation Landfill Site (Beijing, China) was used as the feeding solution (Table 2). The raw landfill leachate was stored at 4 °C to preserve the characteristics of the landfill leachate.

The excess sludge taken from the existing lab-scale nitrification SBR and Anammox UASB systems treating domestic wastewater was used as the inocula for the aerobic SBR (SBR_{pre} and SBR_{ni}) and Anammox SBR (SBR_{ana}), respectively. The sludge from the lab-scale SBR was flocculent, while the sludge from the Anammox UASB system was the mixture of granular and flocculent. After being put in the three-stage SBR system, the mixed liquor suspended solid (MLSS) of the aerobic SBR and the Anammox sludge SBR were 3500 and 4200 mg/L, respectively.

2.3. Operational strategy of three-stage SBR process

The operational strategies for each unit in the three-stage SBR system were different. The SBR_{pre} had two periods: pre-A period (acclimation period) and pre-B period (combination with SBR_{ana}). In pre-A, the influent of the SBR_{pre} was a mixture of the raw mature landfill leachate and tap water. In pre-B, the influent of the SBR_{pre} was a mixture of the mature landfill leachate and effluent of the SBR_{ana}.

The SBR_{ni} had four periods: (i) ni-A period (acclimation period), (ii) ni-B period (stable operation period), (iii) ni-C period (increasing load period), and (iv) ni-D period (the combination with SBR_{ana}). In ni-A, ni-B, and ni-C periods, the influent of the SBR_{ni} was a mixture of the raw mature landfill leachate and tap water. In ni-D period, the influent of the SBR_{ni} was the effluent of the SBR_{pre}.

The SBR_{ana} had four periods: (i) ana-A period (acclimation for synthetic wastewater), (ii) ana-B period (increasing nitrogen load), (iii) ana-C period (adding the effluent of SBR_{ni}), and (iv) ana-D period (combination of the SBR_{pre} and SBR_{ni}). In ana-A and ana-B periods, the SBR_{ana} was fed with synthetic wastewater (Sliekers et al., 2002). In ana-C period, the nitrite in the influent of the SBR_{ana} was supplied by the effluent of the SBR_{ni}, and the ammonia was added in the form of NH_4Cl. In ana-D period, the influent of the SBR_{ana} was the mixed effluents from the SBR_{pre} and SBR_{ni}.

2.4. Analytical methods

The dissolved oxygen (DO), pH, ORP, and temperature were monitored using a pH/Oxi 340i analyzer (WTW Company,