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Biological nitrogen removal from landfill leachate using anaerobic–aerobic process: Denitritation via organics in raw leachate and intracellular storage polymers of microorganisms

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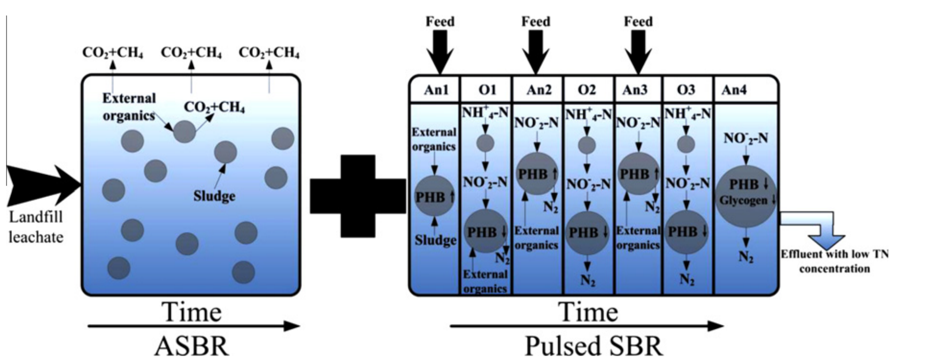
HIGHLIGHTS

- ▶ Advanced nitrogen removal of leachate was achieved without carbon source addition.
- ▶ Pulsed SBR took full advantage of organics in raw leachate for denitritation.
- ▶ PHB and glycogen were used as electron donor orderly for endogenous denitritation.
- ▶ A hypothesis showed that DNGAOs were responsible for endogenous denitritation.
- ▶ Endogenous DNR changed with type alteration of internal carbon source used.

GRAPHICAL ABSTRACT



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ABSTRACT

A system which combined ASBR with pulsed SBR (PSBR) was introduced to enhance COD and nitrogen removal from the real landfill leachate. ASBR was used to degrade the organics from raw leachate mainly. Three equal feeds mode was applied in PSBR operation. The results obtained from the joint operation period (157 days) show that the COD removal rate of ASBR was 83–88% under the specific loading rate of 0.43–0.62 gCOD $gVSS^{-1}$ day^{-1}. PSBR's operation can be divided into four phases according to the different influent NH_4^+-N which increased to 800–1000 mg L^{-1} finally, and total nitrogen (TN) removal rate of more than 90% with the effluent TN of less than 40 mg L^{-1} was obtained. PHB and glycogen can act as electron donor for endogenous denitritation orderly with the hypothetical function from DNGAOs. Consequently, the system achieved COD and TN removal rate of 89.61–96.73% and 97.03–98.87%, respectively, without any extra carbon source addition.

1. Introduction

Sanitary landfill is widely used in municipal solid waste (MSW) treatment throughout the world because of the convenience and low capital cost. As a result, the rainwater percolates through the waste material and the biodegradation of the MSW organic fraction generates a severely contaminated leachate, which is characterized by high concentration of organics, nitrogen, inorganic salts and heavy metals. If the landfill leachate has not been collected cautiously and discharge safely, it could be the potential pollution source that contaminates soil, surface water and groundwater (Nehrenheim et al., 2008).

At present, the chief methods to treat sanitary landfill leachate are physico-chemical and biological processes. Some researches (Uygur and Kargi, 2004; Park et al., 2001; Chiang et al., 2001) found

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that the high concentration of ammonium is the main reason for causing a low efficiency in biological treatment of landfill leachate and physico-chemical process is a necessary pretreatment. The most widely used physico-chemical processes are chemical precipitation (Ozturk et al., 2003), ammonium stripping (Castrillon et al., 2010) and reverse osmosis (Hasar et al., 2009). Although the physico-chemical processes have advantages of simplicity, efficient, etc., their benefits are counteracted by drawbacks like high operational costs and energy consumption. Consequently, the biological processes, such as ANAMMOX (Tao et al., 2012) and anaerobic–aerobic system (Peng et al., 2008), become the feasible technologies for removing organics and nitrogen from landfill leachate. Between them, anaerobic–aerobic system is more suitable for immature landfill leachate treatment. The results from previous studies (Peng et al., 2008; Huo et al., 2008; Hoilijoki et al., 2000) show that most COD and NH_4^+-N in the raw leachate can be removed via methanogenesis and nitrification, however, TN removal becomes an intractable problem because of the shortage of available carbon source.

In terms of immature leachate, the COD concentration could reach above 10,000 mg L^{-1}, and the COD/NH_4^+-N ratio could exceed 10:1. In the previous studies, anaerobic technique was used to convert the organics into CO_2 and CH_4 via methanogenesis, and aerobic/anoxic reactor was utilized to realize nitrification/denitrification. Consequently, the influent of aerobic/anoxic reactor is always with low COD/NH_4^+-N ratio because of the previous methanogenesis. As a result, organics in raw leachate were wasted and extra carbon source must be added in conventional anaerobic–aerobic system in order to obtain satisfying TN removal efficiency, otherwise only NH_4^+-N could be oxidized and NO_x^--N could not be denitrified thoroughly.

Biomass can store excess organics as intracellular storage polymers under the dynamic conditions, in which the stored polymers like polyhydroxybutyrate (PHB) and glycogen can act as electron donor for denitrification (endogenous denitrification) when there is no available external carbon source presenting in wastewater (Beun et al., 2000; Majone et al., 1998; Vocks et al., 2005). For the purpose of realization of advanced nitrogen removal without extra carbon source addition, taking full advantage of organics in raw leachate and intracellular storage polymers of microorganism would be necessary. To authors' knowledge, advanced nitrogen removal form leachate through endogenous denitrification was never reported, as well as the exact types of internal carbon sources used in that.

This work presents a lab-scale anaerobic sequencing batch reactor (ASBR) combining pulsed sequencing batch reactor (PSBR) biological system adapting the removal of organics and nitrogen as a feasible process for the treatment of landfill leachate. The objective of this study is to investigate the possibility of achieving advanced nitrogen removal from landfill leachate without extra carbon source addition by utilizing the organics in raw leachate and intracellular storage polymers of microorganism. Organics and nitrogen removal performance of this biological system was monitored, and variation of nitrogen and intracellular storage polymers during the typical cycle was also investigated. Meanwhile a hypothesis was made to explain which group of microorganism is responsible for nitrogen removal using intracellular storage polymers.

2. Methods

2.1. Experimental lab-scale reactor

Fig. 1 shows the experimental system, which includes an ASBR and a pulsed SBR.

The raw leachate stored in the feed tank was used as the influent of ASBR. And leachate in equalization tank utilized as the influent of PSBR was a mix of effluent from ASBR, raw leachate and effluent from PSBR with a proper ratio in order to adjust the COD/NH_4^+-N ratio from 3 to 5, of these, the raw leachate and effluent of PSBR fed into the equalization tank through the bypass pipe and recirculation pipe, respectively. The working volume of feed tank and equalization tank was 9 L and 10 L for ASBR and PSBR which were made from polymethyl methacrylate. A pH meter and mechanical stirrer were installed inside the ASBR. For PSBR, a pH meter, DO meter, ORP meter, mechanical stirrer and three air diffusers were set up. Temperature adjusting was realized through temperature controller and heating band both for ASBR and PSBR.

2.2. Inoculums

The anaerobic activated sludge from Liulitun Landfill Leachate Treatment Plant (Beijing, China) was inoculated in the ASBR with initial MLSS and MLVSS of 19,780 mg L^{-1} and 8775 mg L^{-1}, respectively. On the other hand, inoculums for PSBR was aerobic activated sludge from a SBR mainly treating the mature leachate, whose initial MLSS, MLVSS and SV were 5365 mg L^{-1}, 4237 mg L^{-1} and 27%, respectively.

2.3. Raw leachate

Raw leachate for this experiment was supplied from the Liulitun MSW Sanitation Landfill Site (Beijing, China), and it was conserved at 4 °C to prevent the natural degradation of organics. The raw leachate, whose BOD_5/COD ratio and COD/NH_4^+-N ratio reached 0.5–0.7 and 7–9, respectively, with the comparably high concentration of COD, can be characterized as immature one. The average values of the principal chemical compounds concentration are summarized in Table 1.

2.4. Experimental procedure

The experiment was divided into start-up and joint operation period. In start-up period, ASBR and PSBR operated separately, then in the joint operation period, the system operated with ASBR linking to the PSBR. The operational modes of ASBR during these two periods were same. One ASBR standard cycle consisted of a rapid fill phase (10 min), an anaerobic phase (22.5 h), a settle phase (30 min), an effluent withdrawal phase (20 min), an idle phase (30 min), in which case 5 L of effluent was discharged, resulting in a hydraulic retention time (HRT) of 2 days. The MLVSS of ASBR was kept at 8000–9000 mg L^{-1} under the sludge retention time (SRT) of 27 days approximately. Temperature was maintained at 35 °C. During the joint operation period, the effluent of ASBR, raw leachate and effluent of PSBR went into the equalization tank under a proper volumetric ratio in order to obtain a demanded COD/NH_4^+-N (3–5), consequently, the effluent of ASBR would be exceeding, so it was used to do other researches in the lab.

In joint operation period, three equal feeds mode (i.e. three times of influent filling with the same volume) was applied for PSBR. Discharging of 3 L effluent every cycle led to 1 L influent filled for 3 times under three equal feeds mode. Fig. 2 shows the operational mode of PSBR. One standard cycle of PSBR could be concluded as follows: An1 → O1 → An2 → O2 → An3 → O3 → An4 → sedimentation → withdrawal as described. Every day one standard cycle was implemented. No extra carbon source was added at the end of O3, which led to implementation of denitritation in An4 with the internal carbon source (endogenous denitritation). All the three anoxic stages (except An4) lasted for 1 h, while the time of rapid fill phase, settle phase and effluent withdrawal phase was 2 min, 30 min and 10 min, respectively. However, the time of idle phase was not fixed. The MLSS and MLVSS of PSBR