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Advanced removal of organic and nitrogen from ammonium-rich landfill leachate using an anaerobic-aerobic system ☆

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A B S T R A C T

A novel system coupling an up-flow anaerobic sludge blanket (UASB) and sequencing batch reactor (SBR) was introduced to achieve advanced removal of organic and nitrogen from ammonium-rich landfill leachate. UASB could remove 88.1% of the influent COD at a volumetric loading rate of 6.8 kg COD$\cdot$m$^{-3}\cdot$d^{-1}. Nitritation–denitritation was responsible for removing 99.8% of NH_4^+-N and 25% of total nitrogen in the SBR under alternating aerobic/anoxic modes. Simultaneous denitritation and methanogenesis in the UASB enhanced COD and TN removal, and replenished alkalinity consumed in nitritation. For the activated sludge of SBR, ammonia oxidizing bacteria were preponderant in nitrifying population, indicated by fluorescence *in situ* hybridization (FISH) analysis. The Monod equation is appropriate to describe the kinetic behavior of heterotrophic denitrifying bacteria, with its kinetic parameters determined from batch experiments.

1. Introduction

Wastewater usually contains high concentrations of organics and ammonia, so nitrogen landfill leachate without treatment will pollute the environment seriously [1]. Cost-effective and highly efficient treatments for leachate are of great interest.

In terms of cost-effectiveness and reusability, biological methods predominate compared to other treatment methods, such as ammonium tripping, ozone oxidization and reverse osmosis [2–7]. In biological processes, organic and nitrogen in the leachate can be transformed into carbon dioxide and nitrogen gas, respectively, which means real removal of organic and nitrogen without secondary pollution. The biological process with an anaerobic–aerobic system is a feasible and sustainable technology for removing organic and nitrogen from landfill leachate [8–11]. In the majority of recently published papers, organic and ammonium removal higher than 90% could be achieved, but the total nitrogen (TN) removal efficiency is not high due to the shortage of carbon source available for denitrification [12,13].

In order to improve the nitrogen removal efficiency, nitritation–denitritation theory has been proposed in recent years, involving oxidation of ammonium to nitrite and then reduction to nitrogen gas. Compared with conventional biological processes, the nitritation–denitritation process can reduce the amount of aeration by 25% and carbon needed by 40% [14–16].

To enhance the denitritation efficiency, simultaneous denitritation and methanogenesis (SDM) was used in anaerobic reactor [17,18]. SDM has become an attractive technology for improving TN removal because it leads to better economic benefit.

In this study, a novel system coupling an up-flow anaerobic sludge bed (UASB) and sequencing batch reactor (SBR) is developed for organic and nitrogen removal from leachate. The main function of the UASB is to improve COD and TN removal through SDM. The SBR is operated under aerobic/anoxic mode to achieve ideal performance for nitrogen removal *via* nitritation–denitritation. Furthermore, batch experiments are conducted to determine the kinetic model for heterotrophic denitritation.

2. Materials and Methods

2.1. Reactor and operation

Fig. 1 shows the experimental system with an UASB and a SBR. The raw leachate sorted in the feed tank was used as the influent of UASB. An equalization tank was designed to meet the requirement of continuous effluent of UASB and intermittent influent of SBR, with leachate in the equalization tank utilized as the influent of SBR. The working volume of UASB and SBR was 3 L and 12 L, respectively, which is made of polymethyl methacrylate. For the SBR, dissolved oxygen (DO) and pH

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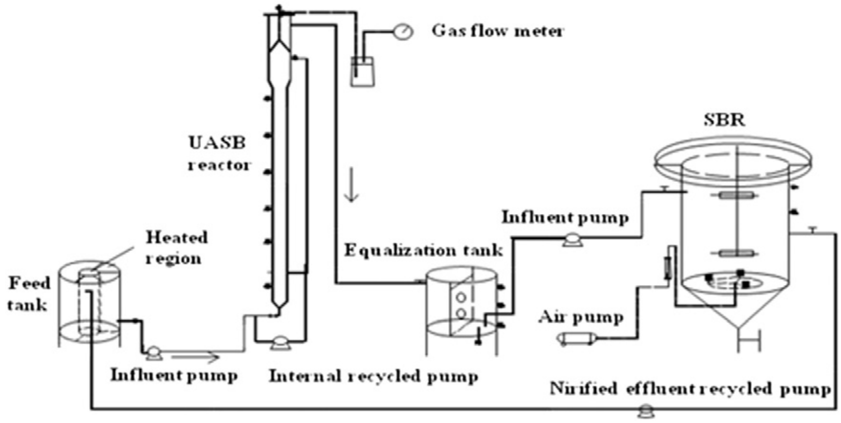

Fig. 1. Schematic diagram of the novel system coupling UASB and SBR.

meters, mechanical stirrer, and diffusers connected to an air compressor were set up. Through the temperature control apparatus, the operation temperature was controlled at (30 ± 2)°C for the UASB, while the SBR reactor was operated at ambient temperature of (20.5–31.4)°C. Nitrified supernatant in the SBR was returned to the UASB for denitrification with 300% of recirculation flow ratio.

In addition, hydraulic retention time of the UASB was 24 h. The SBR had a cycle time of 12 h, with 8 h aerobic, 0.5 h settling, 0.5 h SNS recycling, 2 h anoxic, 0.5 h settling, and 0.5 h decanting periods. The exchange volumetric rate was 50%.

2.2. Landfill leachate

Raw leachate from the Liulitun Municipal Solid Waste Sanitation Landfill Site (Beijing, China) was used as the wastewater in this experiment. The main characteristics of the leachate are shown in Table 1.

2.3. Inoculums

Granulated anaerobic sludge from a methanogenic reactor of a beer factor (Beijing, China) was inoculated in the UASB. Inoculums for SBR were aerobic activated sludge from a lab-scale oxidation ditch treating municipal wastewater, which performs nitrogen removal *via* nitrification–denitrification well. During this experiment, the mixed liquor suspended solid (MLSS) concentrations of UASB and SBR were approximately 55000 and 3500 $mg\cdot L^{-1}$, respectively.

2.4. Analytical methods

Chemical oxygen demand (COD), ammonium (NH_4^+-N), nitrate (NO_3^--N), nitrite (NO_2^--N), MLSS and volatile MLSS (MLVSS) were measured according to the standard methods [19]. TN was determined with a TN/TOC analyzer (Multi N/C 3000, AnanltikjenaAG, Germany). Temperature, DO and pH were monitored using pH/Oxi 340i analyzer (WTW Company, Germany).

Fluorescence *in situ* hybridization (FISH) was performed as specified in Amann [20]. Oligonucleotide probes used in this study were EUBmix for the detection of all bacteria, Nso1225 for ammonia-oxidizing β-Proteo bacteria, Ntspa 662 for *Nitrospira*, and Nit3 for *Nitrobacter*. The images of FISH samples were captured using an OLYMPUS-BX52 fluorescence microscope (Japan). The quantitative analysis of FISH images was performed using Leica QWIN software, where the relative abundance of each group was determined in triplicate as mean percentage of all bacteria.

2.5. Batch tests

Batch experiments were carried out to determine the kinetics of heterotrophic denitritation. In each test, 500 ml of nitritation sludge taken from the parent SBR was transferred to batch reactor. Initial nitrite concentrations were adjusted to desired values of 5, 10, 20, 40, 60, 80 and 100 $mg\cdot L^{-1}$ by adding 10 $mg\cdot L^{-1}$ $NaNO_2$ solution. Ethanol was added to the sludge, resulting in an initial C/N ratio in the reactor higher than 4.0. Higher C/N ratio was used to ensure that denitritation was not limited by carbon source. The pH value was kept approximately constant to 7.0 ± 0.05 through manually adding 0.5 $mol\cdot L^{-1}$ HCl solution. Temperature was controlled at (27 ± 0.4)°C using a water jacket. The MLVSS concentration was controlled at (1250 ± 110) $mg\cdot L^{-1}$. The rate of nitrite reduction was determined from the measured nitrite profile using linear regression.

3. Results and Discussion

3.1. Performance of the UASB-SBR system on landfill leachate treatment

3.1.1. Organic removal

As shown in Fig. 2, during the whole operation period lasting for 113 days, the COD in the raw leachate was (6830 ± 541) $mg\cdot L^{-1}$, corresponding to an average organic loading rate of (6.8 ± 2.3) kg $COD\cdot m^{-3}\cdot d^{-1}$. A significant decrease in UASB influent was caused by the dilution of returned nitrified supernatant. The effluent COD of UASB decreased to (802 ± 124) $mg\cdot L^{-1}$ and most of the organic matters was removed by denitrification and methanogensis. The low influent biodegradable COD in the SBR made nitrification rapid and complete. The final effluent COD of the system was below (319 ± 82) $mg\cdot L^{-1}$ and the residual COD mainly involved refractory organic matters. The COD removal efficiency of the system was (95.2 ± 1.2) %. The contribution of the UASB and SBR to total COD removal efficiency

Table 1
Characteristics of the leachate used in this study

Items	pH	$COD/mg\cdot L^{-1}$	$TN/mg\cdot L^{-1}$	NH_4^+-$N/mg\cdot L^{-1}$	NO_3^--$N/mg\cdot L^{-1}$	NO_2^--$N/mg\cdot L^{-1}$
Range	7.8–8.9	5872–7630	1960–2444	1748–2300	0.8–3.2	0.2–1.3
Average	8.3	6830	2140	2037	1.5	0.9