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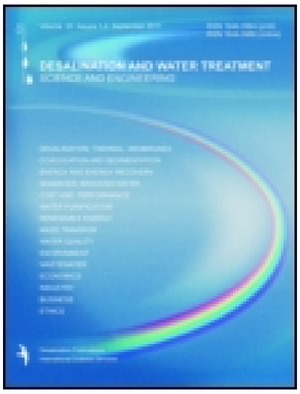

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Landfill-leachate treatment by simultaneous partial nitrification, anammox and denitrification (SNAD) process

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Landfill-leachate treatment by simultaneous partial nitrification, anammox and denitrification (SNAD) process

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ABSTRACT

Simultaneous partial nitrification, anammox and denitrification (SNAD) process was developed in a sequential batch reactor (SBR), and the application of SNAD process for landfill-leachate treatment was investigated. The SNAD process was developed from the biomass of a full-scale landfill-leachate treatment plant. After the adaptation of biomass, the SBR was operated in four stages (I–IV) with varying nitrogen loading rates (NLRs), i.e. 118–280 mg-N/L/d, and organic loading rates (OLRs), i.e. 100–200 mg/L/d. The increase in the NLR has proportionately reduced the NH_4^+-N removal. However, the NO_2^--N concentration was close to zero and the NO_3^--N concentration was less than 36 mg/L in all the stages. A consistent chemical oxygen demand (COD) removal was observed in stages I to IV, which indicate that the SBR was not affected by the increase or decrease of the OLR. The total nitrogen removal in the SBR was mainly achieved by partial nitrification and anammox (69–88%) that was evaluated by a stoichiometric model. Moreover, the influence of NLR and OLR on the SNAD process was determined based on sensitivity index (SI). The SI values indicate that the SNAD process was highly affected by the influent NH_4^+-N compared to COD.

Keywords: Nitrification; Denitrification; Anammox; Sequential batch reactor; Landfill-leachate

1. Introduction

In the recent years, anthropogenic processes have substantially altered the global nitrogen cycle by increasing both the availability and mobility of nitrogenous compounds in the environment including water systems. In order to prevent the pollution from nitrogenous compounds, more stringent wastewater discharge standards are framed. One of the major sources of nitrogenous pollution is from the landfill-leachate. A typical landfill-leachate comprises of high strength ammonium nitrogen and chemical oxygen demand (COD). Nitrogen compounds present in the wastewater can be removed by a variety of processes, out of which biological nitrogen removal has been widely adopted. Conventionally, biological nitrogen removal is achieved by nitrification-denitrification process, i.e. (i) aerobic nitrification of $NH4^+$ by chemolithoautotrophic bacteria to NO_2^- or NO_3^- with O_2 as the electron acceptor, and (ii) anoxic denitrification of NO_2^- or NO_3^- to gaseous N_2 by heterotrophic microorganisms using organic matter as carbon and energy source. However, the conventional nitrification-denitrification is not a cost-effective process for landfill-leachate treatment.

In the last decade, a novel microbial nitrogen removal process has been identified called anaerobic ammonium oxidation (anammox) [1], which is capable of oxidizing

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ammonium into nitrogen gas under the anaerobic condition with nitrite as the electron acceptor. Anammox process requires only 60% energy of nitrification-denitrification process [2]; however, the anammox bacteria are extremely slow growing organisms [3]. After the identification of anammox, many processes have been developed keeping anammox as the base including single reactor system for high ammonium removal over nitrite (SHARON) [4–7], completely autotrophic nitrogen-removal over nitrite (CANON) [8–10], oxygen-limited autotrophic nitrification-denitrification process (OLAND) [11–14], denitrifying ammonium oxidation (DEAMON) [15,16]. On the other hand, anammox removes only 90% of the incoming nitrogen as ammonium/nitrite and leaves 10% of nitrogen as nitrate in the effluent. The presence of oxygen and/or organic carbon can completely inhibit the anammox activity. Most of the wastewaters contain both organic carbon and nitrogen. Several wastewater treatment processes have been developed for the complete removal of organic carbon in the presence of nitrogen. Subsequently, the wastewater containing no or low organic carbon and nitrogen is treated via a variety of nitrogen removal processes. The direct application of anammox for wastewaters containing both organic carbon and nitrogen is questionable or else it requires an organic carbon removal process ahead.

Alternatively, the development of anammox and denitrification in a single reactor can facilitate the simultaneous nitrogen and carbon removal. Recently, simultaneous partial nitrification, anammox and denitrification (SNAD) has been developed following the concepts of anammox and shortcut nitritation-denitritation (SND) [17,18]. The SNAD process has three mechanisms, i.e. partial nitrification, anammox and denitrification for removing nitrogen and COD simultaneously. However, it is difficult to develop a SNAD process in laboratory/full systems owing to the requirements of different environmental and operating conditions for the three processes. Previously, we have identified the occurrence of SNAD process in a full-scale landfill-leachate treatment plant in Taiwan [18]. The application of the SNAD species observed in the full-scale plant to other applications (laboratory/full scale) is still under research. In this study, the SNAD species from the full-scale plant was used to treat the landfill-leachate in a sequential batch reactor (SBR) under laboratory conditions. In addition, the effect of different nitrogen loading rate (NLR) and C/N ratio on the performance of SNAD species in the SBR was investigated. Finally, a theoretical model was applied to estimate the contribution of partial nitrification, anammox and denitrification in total nitrogen removal.

2. Materials and methods

2.1. Reactor system and operation strategy

The SNAD process was developed in a 2.5 L SBR using the biomass from a full-scale landfill-leachate treatment plant in Taiwan [18]. The SBR was operated in 24 h cycles with 12 h influent/reaction, 11.5 h reaction, 0.25 h settling and 0.25 h decanting. The hydraulic retention time (HRT) of the reactor was maintained at 2.5 d by feeding the reactor at 1 L influent per day. The SBR was operated in such a way to maintain a mixed liquid suspended solid (MLSS) concentration around 5,000 mg/L. The sludge retention time (SRT) in the SBR was maintained at infinite for retaining the slow growing anammox bacteria. In addition, a dissolved oxygen (DO) control system was installed in the SBR to supply/adjust the DO accurately. The DO control system composes of a DO meter, air flow valve and PID controller. The application of the DO control system is useful to maintain a DO level in the SBR as accurate as ~0.1 mg/L, which is helpful for preventing the rapid accumulation of nitrite and also to control the nitrite oxidation to nitrate. Throughout the study, the SBR was operated at 35°C and the reactor contents are mixed uniformly using an agitator.

The landfill-leachate samples were collected randomly from the full-scale landfill at four different periods in a calendar year without any predetermined-condition (named as stages I–IV). The collected samples were stored in a refrigerator and used for the SBR study without any pH adjustment. The SBR reactor was operated continuously using the landfill-leachate samples collected at various stages, i.e. stage I for 0–38 d, stage II for 39–54 d, stage III for 55–147 d, and stage IV for 148–191 d. The nature of the landfill-leachate at various stages (I–IV) is shown in Table 1. In the first stage of sampling, the influent NH_4^+-N concentration in the leachate was 295 mg/L with a C/N ratio of 0.85. However, the influent NH_4^+-N concentration increased gradually in the subsequent sampling periods with a maximum of 700 mg/L in stage IV. On the other hand, the COD/TN ratio was maintained as constant (0.85) in stage I and II, decreased gradually in stage III (0.55) and again increased in stage IV (0.71). This data shows that NH_4^+-N and COD concentrations have greatly varying in the sampling time. However, the NO_2^--N and NO_3^--N concentrations in the leachate were close to zero (0–4 mg/L) irrespective of the sampling time. From the influent data, the NLR and organic loading rate (OLR) are calculated using the Eqs. (1) and (2), respectively, and the values are shown in Table 1.

$$\mathrm{NLR} = \frac{\mathrm{Inf.}\left\{(\mathrm{NH})_4^+ - \mathrm{N}) + \left(\mathrm{NO}_2^- - \mathrm{N}\right) + \left(\mathrm{NO}_3^- - \mathrm{N}\right)\right\}}{\mathrm{HRT}} \quad (1)$$

$$\mathrm{OLR} = \frac{\mathrm{Inf.\ COD}}{\mathrm{HRT}} \quad (2)$$

2.2. Analytical techniques

The pH and ORP in the SBR were monitored online using the digital pH and ORP meters (Suntex PC320,

Table 1
Characteristics of landfill-leachate before and after treatment

Stages	Time of operation (d)	Parameters														
		NH_4^+-N (mg/L)		NO_2^--N (mg/L)		NO_3^--N (mg/L)		Inf. BOD (mg/L)	COD (mg/L)		*Inf. COD/TN (BOD/TN) ratio	OLR (mg/L/d)	NLR (mg-N/L/d)	*Eff. TN (mg/L)	NH_4^+-N removal (%)	COD removal (%)
		Inf.	Eff.	Inf.	Eff.	Inf.	Eff.		Inf.	Eff.						
I	38	295	53	2	1.54	0	36.5	88	250	198	0.85 (0.35)	100	118	91	82	21
II	16	590	37	4	2.23	0	26.5	164	500	329	0.85 (0.33)	200	236	66	94	34
III	93	660	111	0	0.68	0	28.9	155	365	252	0.55 (0.42)	146	264	140	83	31
IV	44	700	126	0.5	0.61	1.8	26.5	178	500	279	0.71 (0.36)	200	280	153	82	45

*TN is the sum of NH_4^+-N, NO_2^--N and NO_3^--N

Taiwan), respectively. All chemical analyses were performed according to the Standard Methods [19]. The concentrations of ammonium and nitrite were determined by colorimetric methods, whereas nitrate was measured spectrophotometrically. The organic matter content of the wastewater was analyzed according to the Standard Methods and expressed as COD. Moreover, the solids in the SBR including suspended solid (SS), volatile suspended solid (VSS), mixed liquid suspended solid (MLSS) and mixed liquid volatile suspended solid (MLVSS) were determined by drying in the oven and muffle furnace as per the Standard Methods [19].

3. Results and discussion

3.1. Performance of SNAD system

Table 1 shows the influent characteristics of the landfill-leachate at various stages of the SBR operation. It can be seen in Table 1 that the NLR was gradually increasing over stages I–IV. On the other hand, the OLR was fluctuating greatly due to the random fluctuation of COD in the landfill-leachate samples. The BOD levels in the landfill-leachate also varying significantly between stages I–IV. However, the BOD/TN value of the landfill-leachate remains almost the same (0.33–0.42) in all stages due to the elevated NH_4^+-N concentration. This information reveals that (i) maintaining consistent influent characteristics to any landfill-leachate treatment system is highly impossible, and (ii) the treatment system could be designed in such a way to withstand varying influent concentration and/or shock loading. At the same time, constructing a balancing tank prior to the bioreactor could be useful for adjusting the flow rate to the bioreactor.

The effluent concentrations of TN, COD and their removal efficiencies are shown in Table 1. In SNAD system, a part of NH_4^+-N is converted to NO_2^--N by conventional chemolithoautotrophic ammonium oxidizing bacteria (AOB). The successive oxidation of NO_2^--N to NO_3^--N is not possible owing to the low DO concentration in the aeration tank (~0.1 mg/L). Subsequently, the NO_2^--N produced is utilized along with the remaining NH_4^+-N by the anammox bacteria to nitrogen gas. Finally, the NO_3^--N produced in anammox process is utilized by the heterotrophic denitrifiers. The detailed methodology for assessing the contribution of each process in nitrogen removal is based on our earlier report [18]. The profiles of nitrogen species under all the stages are shown in Fig. 1. The NH_4^+-N removal in the reactor was unstable in the beginning period of stage I; however, complete NH_4^+-N removal was observed in stage II. This indicates that doubling the NLR (118–236 mg-N/L/d) and OLR (100–200 mg/L/d) has no significant effect on the SBR system. However, the subsequent increase in the NLR decreased the performance of the SBR, which is evident from the higher effluent NH_4^+-N concentrations. This could be due to the limitation of partial nitrification process under low DO concentration in the SBR. A decrease in DO concentration of as low as 0.04 mg/L was observed in an SBR performing the CANON process [9]. The DO in this study was controlled precisely around 0.1 mg/L through PID controller. Therefore, it is envisaged that the major rate-limiting step in the SBR is probably the transfer of oxygen from the gas-phase to the liquid phase [10].

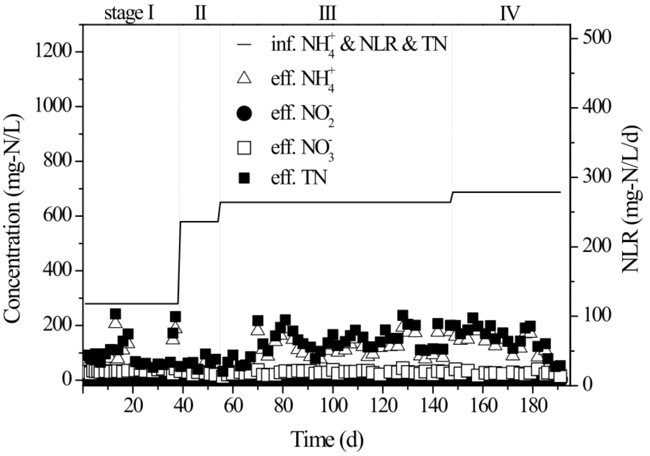

Fig. 1. Profiles of influent and effluent nitrogen compounds in the landfill-leachate.

The removal efficiencies of NH_4^+-N and the profiles of TN concentration are shown in Fig. 1. On the other hand, the NO_2^--N concentration was close to zero and the NO_3^--N concentration was less than 36 mg/L (Fig. 1) in all the stages indicating that the anammox bacteria are highly efficient in the SNAD process. The COD present in the SBR is used by heterotrophic bacteria as carbon and energy sources during denitrification; the consistent removal of COD (Fig. 2) proves the activity of the denitrifers in the SBR. The COD removal profiles under stages I–IV indicate that the SBR was not affected by the increase or decrease in the OLR. Theoretically, the molar ratio of NH_4^+-N: NO_2^--N consumed in anammox is 1:1.32 and produces 0.26 mole of NO_3^--N, and subsequently that is utilized in denitrification. In our previous study, it was observed that 1 g of COD is consumed for 0.38 g of NO_3^--N removal [18]. The increase in NLR could increase the stoichiometric production of NO_3^--N, which is directly related to the COD consumption in the SBR. Therefore, the COD consumption in the SBR is based on the performance of partial nitrification and anammox. However, the excess COD or the presence of non-biodegradable organic matter in the landfill-leachate has no significant effect on the performance of the SBR. On the other hand, the successful autotrophic nitrogen removal processes, for example anammox, is not stable in the presence of complex COD. A recent study indicated that anammox bacteria were successful in the oxidation of propionate

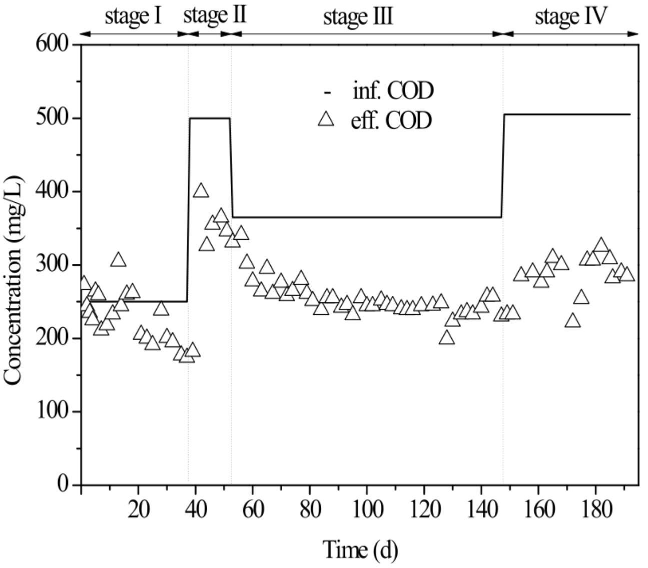

Fig. 2. Profiles of influent and effluent COD.

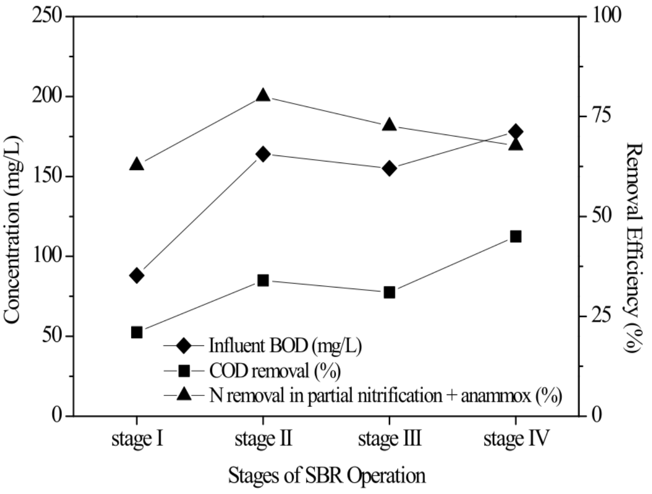

Fig. 3. Performance of SNAD process at various stages of SBR operation.

and the presence of glucose, formate and alanine had no effect on the anammox process. However, the presence of methanol is found to have irreversible inhibition at concentration as low as 0.5 mM. The application of SNAD system could completely remove nitrogen and a part of COD. As a whole, the SNAD system is more suitable for the treatment of low C/N wastewater/landfill-leachate.

3.2. Evaluation of the performances of different processes in the SNAD

In our previous study, a simple stoichiometric model was applied to evaluate the performance of each process, i.e. partial nitrification, anammox and denitrification, in nitrogen removal [18]. Similarly, the model was applied in this study to evaluate the performance of each process at various stages of SBR operation, and the outcomes are shown in Table 2. The results reveal that partial nitrification and anammox are the main nitrogen removal processes in the SNAD. On the other hand, heterotrophic denitrification is responsible for the removal of NO_2^--N, NO_3^--N and COD, and resulted in a TN removal of around 6–9% (Table 2). The profiles of average influent BOD, COD removal and the TN removal in partial nitrification and anammox at various stages of treatment are shown in Fig. 3. In stages I–III, the trends of all three profiles are similar indicating that nitrogen removal is in good correlation with organic matter removal irrespective of the variations in OLR and NLR. However, a decrease in the efficiency of TN removal by partial nitrification and anammox was observed at the highest NLR (stage IV); whereas, the COD removal continues to increase due to the production of higher NO_3^--N in stage IV. These facts indicate that the SNAD system is more sensitive to NLR than the OLR under the conditions investigated in this study. Subsequently, the response of the SNAD system under various OLR and NLR was evaluated based on the sensitivity index as shown in Eq. (3) [20].

$$SI = \frac{O_{\max} - O_s}{O_s} \tag{3}$$

where Omax is the maximum concentration of substrate in the effluent at stages II–IV (mg/L), and Os is the normal concentration of substrate in the effluent at stage I (mg/L). The values of SI for all nitrogen species and COD are shown in Table 3. The SI values indicate that the SNAD process is affected more by the influent NH_4^+-N compared to COD. Moreover, the negative values for NO_2^--N and NO_3^--N reflects that the conversion of NH_4^+-N to NO_2^--N

Table 2
Nitrogen consumption in the various processes of SNAD system

Item	Stage I	Stage II	Stage III	Stage IV
TN removal by partial nitrification + anammox (%)	62.7	80	72.6	67.7
TN removal by denitrification (%)	6.2	8.9	6	7.6
Overall TN removal (%)	68.9	88.9	78.6	75.3

Table 3
Sensitivity of the SNAD system to various influent C/N ratios

Stages	Sensitivity index (SI) for various parameters*			
	COD	NH_4^+-N	NO_2^--N	NO_3^--N
Stage II	0.66	–0.30	0.45	–0.27
Stage III	0.27	1.09	–0.56	–0.21
Stage IV	0.41	1.38	–0.6	–0.27

*Effluent quality of stage I is kept as the reference for SI calculation

and NO_3^--N (since no NO_2^--N and NO_3^--N in the influent) have no significant effect on the performance of the SBR.

4. Conclusion

The SNAD process was successfully developed in a SBR. The influence of OLR and NLR on the performance of the SBR was investigated. The experimental outcomes indicate that the performance of the SBR is affected more by the NLR compared to the OLR. The influent BOD has good correlation with the TN removal in the system. Partial nitrification and anammox were responsible for majority of total nitrogen removal in the SBR. The landfill-leachate contains less bioavailable COD; therefore, the effluent from the SNAD process still needs some treatment with regard to organics. The maximum NH_4^+-N removal in the SBR was close to 94%, however, it can be improved by increasing the oxygen-transfer from gas to liquid phase. As a whole, the experimental findings indicate that the SNAD process is more suitable for landfill-leachate treatment with high nitrogen and low COD content.

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