SBR as a relevant technology to combine anaerobic digestion and denitrification in a single reactor

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Abstract Two laboratory-scale sequencing batch reactors were used to study biological treatment of carbon and nitrogen in a new combined anaerobic-aerobic system. Piggery wastewater was used as a model effluent. The anaerobic reactor, fed with raw wastewater and nitrite/nitrate recycling from the aerobic reactor, carried out denitrification and anaerobic digestion of organic carbon. In the aerobic reactor, organic carbon removal and nitrification of ammonia to mainly nitrite occurred. Three recycle-to-influent ratios from 1 to 3 were tested. The higher the recycle-to-influent ratio, the lower the concentrations of nitrogen oxides in the final effluent. Carbon removal efficiency was quite stable, whatever the ratio. However, the effect of this ratio was attenuated because of denitrification in the aerobic reactor, which increased the performances of the process. The use of sequencing batch reactors was essential to apply the configuration proposed, in order to combine denitrification and methanogenesis that require antagonistic conditions.

Keywords Anaerobic digestion; carbon removal; denitrification; nitrification; nitrogen removal; piggery wastewater; sequencing batch reactor

Introduction

Anaerobic digestion is widely used to remove organic matter from high strength wastewaters because of its relatively low sludge production and energy needs, compared to aerobic treatments. Organic nitrogenous compounds present in the wastewater, such as proteins, amino-acids or urea, are mainly reduced to ammonia, which is not further degraded in anaerobic conditions. A post-treatment may be necessary to remove ammonia before discharge, biological nitrification-denitrification being the most widely used process (Odegaard, 1988). In the case of a wastewater with a low COD/TKN ratio, organic carbon content of the digested effluent may be insufficient to achieve complete denitrification and the addition of a costly external carbon source is then required.

Akunna *et al.* (1994b) proposed a process in which denitrification and anaerobic digestion take place in the same reactor. A following aerobic reactor completes carbon removal and achieves nitrification of the wastewater which is then recycled to the anaerobic unit. Few previous works reported this was possible with a preliminary enrichment of denitrifying bacteria (Hanaki and Polprasert, 1989; Kuroda *et al.*, 1988). Conditions for the feasibility of denitrification in an anaerobic digester using a synthetic wastewater and without any bacterial enrichment have been established to be strongly dependent on the COD/N–NO_x ratio and the nature of the carbon source (Akunna *et al.*, 1992, 1993, 1994a).

Attempts to carry out denitrification and methane production in a completely mixed reactor have not proved very effective (Akunna *et al.*, 1992, 1993, 1994a, 1998) because nitrogen oxides are known to inhibit (reversibly) methanogenic bacteria (Chen and Lin, 1993; Akunna *et al.*, 1998; Clarens *et al.*, 1998). Furthermore, dissimilatory nitrate reduction to ammonia can occur in the system depending on the type of carbon compounds (raw substrates or fermentation products) present in the system. Consequently, biofilters have firstly been used in the combined denitrification/anaerobic digestion system. Indeed, in the combined process developed by Akunna *et al.* (1994b), the anaerobic filter was shown to

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behave like a plug-flow reactor, with a denitrification activity at the bottom of the reactor and methane production in the upper part, where N-oxides had been totally removed. A spontaneous partition occurred between fixed denitrifying and methanogenic population, making easier the co-existence of the two processes in the same reactor. Another way to obtain a separation between the denitrification and anaerobic reaction is to use batch reactors in which the reactions should be separated, not spatially like in a plug-flow reactor, but temporally. It is why the SBR technology seems to be particularly suitable to this new configuration.

Due to its low COD/N ratio and according to previous results (Akunna *et al.*, 1992), piggery wastewater should be a good model to apply the system proposed by Akunna *et al.* (1994b). Indeed, a previous study showed that good denitrification performances could be obtained by a methanogenic sludge in piggery wastewater (Bernet *et al.*, 1996).

Materials and methods

Piggery wastewater

The first part of the work was carried out with wastewater sieved through 1 mm diameter mesh, and centrifuged at 11000 rpm for 15 minutes to remove most of the suspended solids (Table 1).

In the last part of the study, the raw wastewater was used without any preliminary treatment to check the effect of TSS on the performance of the system. TSS concentration in the wastewater was as high as 18 $g \cdot l^{-1}$.

Analytical methods

Total Kjeldahl nitrogen and ammonium were determined using the titrimetric method after distillation with a Büchi apparatus (APHA, 1992). Nitrate and nitrite were analyzed by an ion chromatography system using conductivity detection (Dionex-100). Separation and elution of the anions were carried out on IonPac AS12A analytical column utilizing a carbonate/bicarbonate eluant and AutoSupression technology. Integration was done using a PC fitted with Peaknet Software. Total organic carbon (TOC) was determined by UV oxidation with a Dohrman DC 80 apparatus. Volatile fatty acids (VFA) analysis was done using a gas chromatograph fitted with a flame ionization detector (Chrompac CP 9000) and coupled with an integrator (Shimadzu CR 3A). Gas analysis by gas chromatography was carried out with a Shimadzu GC-8A apparatus with argon carrier using a katharometer detector. The chromatograph was coupled to a Shimadzu CR 3A integrator. Total suspended solids (TSS) and volatile suspended solids (VSS) were determined using *Standard Methods* (APHA, 1992).

The redox potential (E0) in the anaerobic reactor was monitored using an Ingold pH transmitter (2400) and a combination redox electrode (Ag/AgCl reference system, KCl 3M, $Eh_{ref}^{35°C}$= 199.8 mV). pH was monitored with an Ingold pH-meter (2301).

Table 1 Average composition of liquid swine manure after sieving and centrifugation

Parameter	Average value	Standard deviation
TOC ($g \cdot l^{-1}$)	5.86	1.33
Total VFA (% TOC)	57	
TKN ($g \cdot l^{-1}$)	3.69	0.74
NH_4–N ($g \cdot l^{-1}$)	2.94	0.40
TSS ($g \cdot l^{-1}$)	2.84–3.96	3.43
pH	7.68	0.14

TOC: Total Organic Carbon; VFA: Volatile Fatty Acids;
TKN: Total Kjeldahl Nitrogen; TSS: Total Suspended Solids

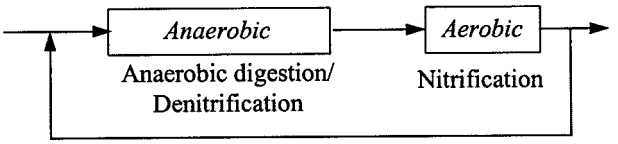

Figure 1 Configuration of the process proposed

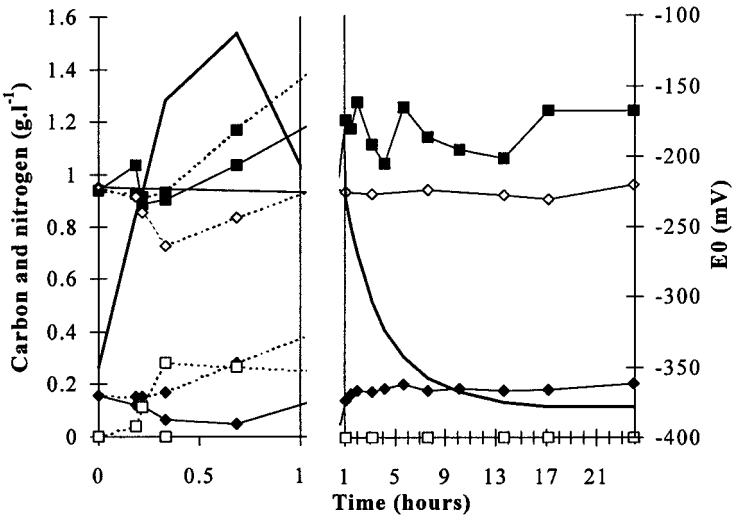

Figure 2 Variations of main parameters during a cycle in the anaerobic SBR at R=2. ■ : TOC, ◆ : VFA-C, □ : N–NO_x , ◇: NH_4–N, —: E0. Dash lines: concentrations introduced in the reactor; plain lines: measured concentrations

Experimental system

Figure 1 shows the process configuration. The cycle length was 24 hours for both SBR.

Anaerobic Reactor. The anaerobic reactor (AN) had an active liquid volume of 1.5 l. It was seeded with 0.75 l of anaerobic sludge obtained from a laboratory digester treating wine distillery wastewater. The volume was completed with 0.75 l of tap water. The temperature was kept constant at 35ºC by a water jacket and mixing obtained using a magnetic stirrer maintained at a constant speed of 400 rpm.

Aerobic Reactor. Two aerobic reactors were used successively in this study. The first one (N1) with an active volume of 1.5 l was used at low flow rate (0.1 l · d^{-1}). When the organic carbon load of the system was doubled, a 4 l reactor (N_2) containing 3 l of mixed liquor was used. These reactors were inoculated with sludge from another nitrifying reactor treating piggery wastewater at room temperature. Aeration was provided by compressors connected to plastic tubes placed at the bottom of the reactors. The reactors were fed with digested effluent from the anaerobic reactor. They were kept at ambient temperature (20–22ºC) and mixing was carried out using a magnetic stirrer moving at 700 rpm.

Sequencing batch reactors operations

The reactors were operated in a sequencing batch mode. The cycle length was 24 hours for both aerobic and anaerobic reactors with a minimum reaction time of 22 hours. The filling time depended on the recycle-to-influent ratio (R) applied. The different conditions applied are detailed in Table 2. Note that $R=Q_2/Q1$, where Q_1 and Q_2 are the raw wastewater and the recycled nitrified effluent flow rates respectively.

Table 2 Hydraulic conditions at the different flow-rates (Q_1) and recycle-to-influent ratios (R) used

Period No.	Period (days)	Q_1 ($l \cdot d^{-1}$)	R ($l \cdot l^{-1}$)	Anaerobic reactor volume (l)	Aerobic reactor volume (l)	Process HRT (days)
I	1–64	0.1	1	1.5	1.5	30
II	65–155	0.1	2	1.5	1.5	30
III	156–361	0.2	2	1.5	3	22.5
IV	361–429	0.2	3	1.5	3	22.5
V	429–704	0.2	3	1.5	3	22.5

Results and discussion

We mainly focus on the anaerobic reactor performances; more detailed results on the process can be obtained in Bernet *et al.* (2000).

Anaerobic reactor

Complete denitrification was observed in the anaerobic reactor at all loading rate and recycle-to-influent ratios studied. Figure 2 shows the results obtained during a cycle at R=2 and an influent flow rate of $0.2\,l \cdot d^{-1}$. Filling time for raw wastewater and nitrified effluent (from aerobic reactor) were 65 and 20 minutes respectively.

The amount of nitrogen oxides (97% nitrites) fed into the anaerobic reactor at R=2 corresponded to a concentration of about 250 mg NO_x–$N \cdot l^{-1}$ in the reactor. All these nitrogen oxides were eliminated during the 20-minute filling period, together with a corresponding reduction of VFA. Neither nitrite, nor nitrate could be detected in the reactor. Denitrification was the only reduction pathway of the nitrogen oxides in the anaerobic reactor. In all experimental conditions studied, dissimilatory reduction to ammonia (DNRA) was not observed. This could be due to the low C/N ratio of the wastewater and the high VFA fraction of the organic matter, both parameters being favoured by the batch feeding, compared to continuous reactors. These two parameters have been shown to favour denitrification over dissimilatory reduction to ammonia (Akunna *et al.*, 1992, 1993).

A significant increase in the redox potential (from –350 mV to –112 mV) was observed during filling with nitrified effluent. It has been shown earlier that such an increase would not be due to the presence of oxidized nitrogenous compounds but rather to denitrifying activity (Percheron *et al.*, 1999). It took almost three hours for the redox potential to go down to a value of about –300 mV, which was a much more conducive environment for methanogenesis. No significant changes in TOC concentration could be observed before and after filling (even though TOC consumption should have taken place during the reduction of nitrogen oxides), probably due to the hydrolysis of the VSS in the reactor which might have replenished consumed soluble TOC.

Figure 3 shows that gas composition in the anaerobic SBR, during a period of 325 days, is closely linked to the nitrogen oxides loading rate. The reactor activity was very sensitive to the composition of the influent. In the presence of high nitrite and/or nitrate concentrations, organic matter was first used as electron donor for denitrification. During this period a high nitrogen concentration was detected in the biogas. When the nitrogen oxides were absent or in a low concentration in the influent, organic matter was then mainly converted to biogas and an increase in methane concentration was observed in the gas produced, together with a decrease in the proportion of nitrogen gas produced. Therefore, this system ensures an optimization of the utilization of available carbon, with a priority for denitrification. The low CO_2 concentration of the biogas was due to the high pH value in the reactor, causing an important conversion to HCO_3^-.

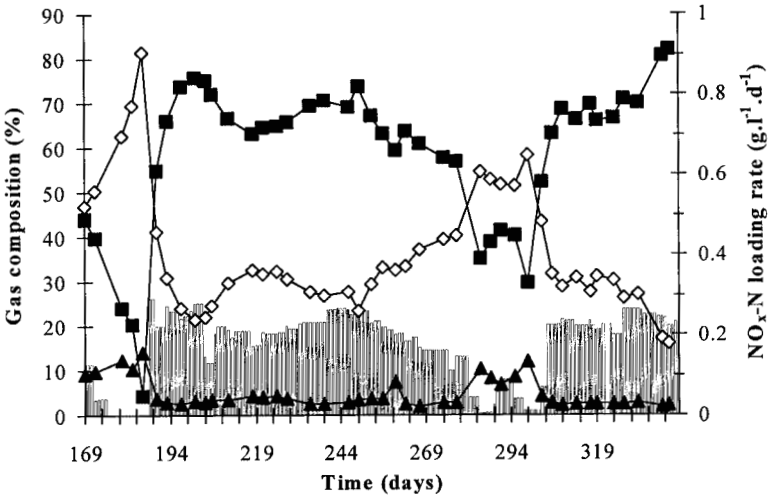

Figure 3 Gas composition and N-oxides loading rate in the anaerobic SBR at R=2 (period III). □ : NO_x–N loading rate, ■: N_2, ◇: CH_4, ▲: CO_2

Table 3 Average performances of the process in the different conditions tested.

Waste water	Q_1 ($l \cdot d^{-1}$)	R ($l \cdot l^{-1}$)	TOC removal (%)	TKN removal (%)	Total N removal (%)
Centrif	0.1	1	84	87	59
Centrif	0.1	2	92	91	76
Centrif	0.2	2	89	89	72
Centrif	0.2	3	82	90	69
Sieved	0.2	3	81	85	76

Aerobic reactor

DO concentration decreased quickly to a low value of about 0.6–0.8 mg $O_2 \cdot l^{-1}$ during the filling period, because of the high oxygen demand caused by the introduction of anaerobic effluent (not shown). It is more than probable that at such a low DO concentration, denitrification occurred in the reactor especially at the very beginning of the cycle when biodegradable carbon from the anaerobic SBR was available. The DO concentration began to rise only after a period of up to 11 hours. The increase in DO seemed to signal the end of nitrification. Such information is available because SBR is a dynamic process compared to continuous processes in which steady states are generally obtained.

Nitrite was generally the main end-product of nitrification. This could be due to insufficient dissolved oxygen availability and to the lower affinity for oxygen of nitrite oxidizers, compared to ammonia oxidizers (Hanaki *et al.*, 1990; Laanbroek and Gerards, 1993) or/and to free ammonia inhibition (Anthonisen *et al.*, 1976).

Overall process

Table 3 presents the average performance of the process under the different experimental conditions. Good results were obtained for soluble TOC and TKN removal.

The difference between TKN and total N removal is due to nitrogen oxides discharged in the final effluent. The amount of nitrogen oxides in the final effluent depended only, theoretically, on the recyling ratio, R. High values of R should result in high total nitrogen removal and subsequently low concentrations of nitrogen oxides in the final effluent. However, as mentioned above, denitrification in the aerobic reactor increased the performances of the system. This phenomenon could be enhanced to optimize the process.

Conclusion

A system coupling two sequencing batch reactors for the biological treatment of carbon and nitrogen was carried out, using piggery wastewater as a model effluent. It was possible to carry out denitrification in the methanogenic reactor. Methane production followed denitrification. In the aerobic reactor, nitrification and carbon removal were observed. The efficiency of nitrogen removal was dependent on the applied recycle-to-influent ratio and was enhanced by partial denitrification in the aerobic reactor. Overall performances of TOC and TKN removal of respectively 81 to 91% and 85 to 91% were achieved, without any optimization of the cycle length.

The process ensured an optimization of carbon use in the anaerobic reactor since methane production started after carbon use for denitrification. This configuration induced a dilution of the raw wastewater in the digester, reducing the concentrations of inhibitory compounds like ammonia. Good nitrification performances were obtained with nitrite as the main product.

The use of sequencing batch reactors for the application of this new configuration is essential for the success of the process. Indeed, the separation of denitrification and methanogenesis in the digester is the key point of the system which is possible either by using plug flow reactors, or batch reactors.

This process could be applied to the treatment of high strength agro-industrial wastewaters.

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