

Combined anaerobic/aerobic (UASB + UBAF) system for organic matter and nitrogen removal from a high strength industrial wastewater

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Abstract The paper presents the experimental results collected from seven months of operation of a combined anaerobic/aerobic system treating an industrial effluent with averaged content of organic matter and nitrogen of 10.4 g COD/L and 790 mg NKT/L, respectively. The system was formed by an upflow anaerobic sludge blanket (UASB) and an upflow biological aerated filter (UBAF) connected in series, with a recycling line of the UBAF effluent into the UASB for its denitrification. The best results were obtained when operating the two reactors, UASB and UBAF, with hydraulic retention times (HRT) of 3.3 and 1.3 days, respectively, and a recycling ratio of 6.7. Under these conditions the system removed 98% of the organic matter and ammonia and 91% of the total nitrogen entering the system.

The activity of the different microorganisms was followed through activity assays consisting of measuring the consumption or production rate of any specific substrate or metabolism product. Thus when operating the system under the aforementioned conditions the reported values for the specific activity of methanogenic microorganisms and denitrifiers coexisting in the UASB were 1.05 g COD/g VS d and 32.08 mg NO₃-N/g VS d, respectively. While the activity of ammonia and nitrite oxidisers within the UBAF were 47.65 and 4.36 mg O₂/g VS h, respectively.

Keywords Combined systems; nitrification; denitrification; UASB; UBAF; industrial wastewater

Introduction

Traditionally the treatment of industrial effluents with high organic matter content has been pursued in anaerobic biological reactors due to the significant economic and technical advantages that can be achieved. The anaerobic treatment has been proved to show a high efficiency for suspended and soluble organic matter removal, however it is inherently inadequate for nutrient removal (Speece, 1996). The progressive application of more severe regulations limiting the discharge of total nitrogen and phosphorous to surface and groundwater streams is demanding the treatment of anaerobic reactor effluents to fulfil the discharge limits at the lowest cost. The technologies usually applied for total nutrient removal are based on a spatial and/or temporal sequence of different anaerobic, anoxic and aerobic steps. Its application for treating anaerobic reactor effluents can be an expensive solution due to the large operational and installation costs associated with it.

On the other hand, when only organic matter and nitrogen removal is required, the nitrification of effluents previously ammonified in an anaerobic reactor in a subsequent aerobic reactor it does not satisfy the total nitrogen discharge limit in many situations and consequently it is no longer a solution. There exists the possibility of integrating the downstream aerobic stage with the anaerobic reactor by recycling the nitrified effluent to the anaerobic reactor for its denitrification. This type of anaerobic/aerobic system has been proven to be successful for removing organic matter and solids from domestic sewage with 95 and 88 percent of suspended solids and COD removal respectively (Gonçalves *et al.* 1999). During the last decade the performance of different integrated or combined anaerobic/aerobic systems that have been operated for simultaneous removal of organic matter and total nitrogen (Acitores *et al.* 1991, Paul *et al.* 1998, Villaverde *et al.* 2000, Van Benthum *et al.*

1997, Osashi *et al.* 1999) has been reported. Some of these configurations operate with attached biomass in the nitrification phase in order to improve the retention of the slow growing nitrifying biomass. (Fdz-Polanco *et al.* 1996). Nevertheless this option of combining the anaerobic reactor with a subsequent aerobic stage is often refused due to the lack of information about the performance of combined anaerobic-aerobic systems that operate effectively for simultaneous removal of organic matter and nitrogen.

The aim of the experimental work presented here was improving the treatment of an industrial wastewater with a high COD and nitrogen content that was being treated in an anaerobic UASB reactor in order to fulfil the discharge limits for organic matter and total nitrogen. With this aim an anaerobic-aerobic system was designed, integrated and operated. The system was formed by the former UASB (similar to that employed at industrial scale) to which an UBAF was coupled. Initially this type of combined system was selected by its technical and economical potential advantages here summarised:

1. It produces effluents of very high quality accomplishing the discharge limits for total nitrogen.
2. The UBAF technology allows for a better usage and control of the oxygen, and for the recirculation of a nitrified effluent to the UASB with a low oxygen content.
3. The UBAF technology offers some advantages for nitrification such as very high biomass concentration (up to 30 g/L), high biomass retention time operating with low hydraulic retention time and an excellent behaviour overcoming volumetric or substrate overloads.
4. There is also the possibility of treating in the UASB the back-washing waters of the UBAF and thus mineralising the aerobic excess sludge.

Material and methods

Experimental set-up and experimentation planning

The experimental set-up was formed by two biological reactors, a UASB reactor and a UBAF of 10 and 3 L of volume, respectively, as shown in Figure 1. Both reactors were connected in series with a recycling line from the exit of the aerobic reactor to the entrance of the UASB reactor.

The recycling line had a buffer tank of 0.5 L from which the aerobic effluent was pumped to the UASB entrance and out of the system. A conventional liquid displacement device measured the biogas was generated in the anaerobic stage. Both reactors operated at a constant temperature of 33°C. The industrial wastewater was fed into the UASB reactor

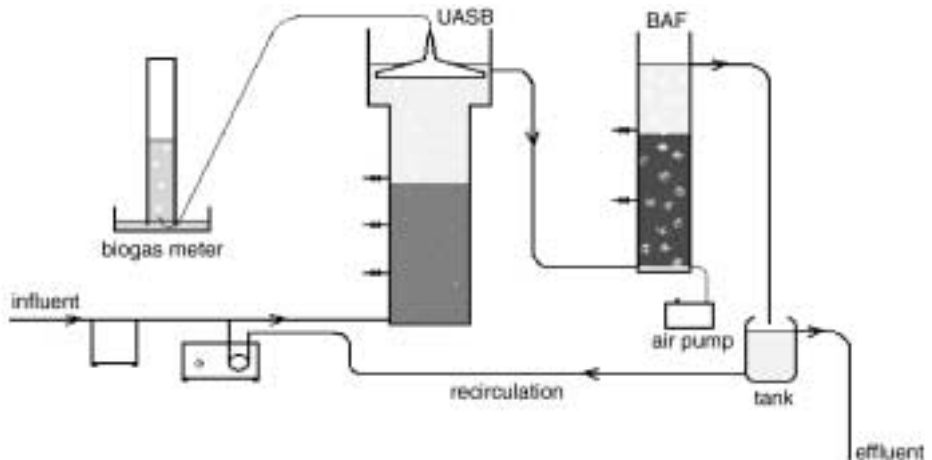


Figure 1 Experimental set-up

diluted in the recycling line coming from the UBAF. Different biological processes take place in the: anaerobic removal of organic matter, denitrification of nitrite and nitrate recycled from the UBAF, and hydrolysis to ammonia of the organic nitrogen that enters with the feed. The effluent of the UASB flowed into the UBAF by gravity, where is mixed with the air supplied by means of a diaphragm pump and the nitrification of the ammonia and aerobic oxidation of the organic matter that goes out of the UASB takes place. As a result of the growth of the aerobic microorganisms, both nitrifiers and heterotrophs, the filter is getting clogging and a periodic wash-out is necessary to remove the excess of biomass and let the biofilter operate in optimum conditions. The frequency of the wash-out depends on the applied load, but in this system varied between 7 and 14 days. The media of the biofilter consisted of 2 litres of particles of puzzolane with an average diameter of 4 mm. Before the start-up of the reactor, the media was maintained submerged in a nitrifying medium to improve the attachment of nitrifiers.

The UASB reactor was filled with 5 L of sludge from the industrial anaerobic reactor that is treating the same influent. Methanogenic and denitrifying activity was determined for this sludge, obtaining a value of the methanogenic activity at 30°C of 0.25 g COD/g VS d, and a denitrifying activity of 10.5 and 4.0 mg N/g VS d at 30 and 20°C respectively.

In order to check the influence over the global behaviour of the system of the ratio between reactors' volume, during the last two periods of operation the volume of the reactors were modified to 5 and 2 L for the UASB and the UBAF respectively. The media in the UBAF was reduced to 1 L.

The influent of the system consists of a wastewater from a food industry, whose composition is shown in Table 1.

Analysis and activity tests

All analysis followed standard methods (APHA, 1995). Total organic carbon (TOC) was determined using a SHIMADZU TOC Analyzer 5050. Temperature, pH, dissolved oxygen (DO), ammonia nitrogen and redox potential values were determined using selective electrodes and anions concentrations (NO_2^- , NO_3^- , and PO_4^{-3}) were determined by HPLC.

To follow the evolution of the different microorganisms groups involved in the system, substrate uptake rates (SUR) of methanogenics, heterotrophs, nitrifiers and denitrifiers were also determined as an indicator of their activity. These activity tests are based on measuring maximum rates of substrate utilization per biomass unit (μ_{max}/Y). Two different experimental set-ups were employed.

Methanogenic activity. The system consists of a 500 mL digester connected to a Mariotte flask filled with an NaOH solution similar to that described by Soto *et al.* (1993). Methane produced in the digester displaced an equivalent liquid volume of the Mariotte flask that is

Table 1 Wastewater composition

	Range	Average
PH	5.2–6.2	5.9
COD ₁ (mg/L)	8710–11725	10400
COD ₂ (mg/L)	7610–10415	9100
BOD ₅ (mg/L)	2665–6850	5250
TSS (mg/L)	815–2100	1275
VSS (mg/L)	705–1880	1040
NKT (mg/L)	568–919	790
N-NH ₄ ⁺ (mg/L)	77–290	210
Alcalinity (mg/L)	720–2225	1415

collected in a calibrated cylinder. The digester is placed in a shaker at 200 rpm and at constant temperature of 33°C.

Other activities. Specific activity of heterotrophs, ammonia and nitrite oxidizers were determined by closed respirometry. The respirometer consists of a half a litre thermostatic cell provided with inlets for introducing the biomass sample and the different substrates. An Oxymeter ORION 870 connected to a computer was used for recording data. A magnetic stirrer provided good mixing conditions inside the cell. The conditions under which these tests were carried out are described elsewhere (Villaverde *et al.* 1997). The activity of denitrifiers was determined with an experimental set-up similar to that used for respirometry assays and is shown in Figure 2. Prior to the assay oxygen was stripped off the liquid sample with nitrogen gas. Oxygen concentration in the sample was always under 0.05 mg/L. The initial substrate concentration was selected for each test in order to have zero order reaction rate conditions with respect to the substrate (Cech *et al.* 1984). The ratio between initial substrate and microorganisms concentrations was smaller than 3 to minimize biomass growth during the experiments (Chudoba *et al.* 1992). The results are expressed as milligrams of oxygen (O_2) consumed per gram of volatile solids (VS) and per hour ($mg\ O_2/g\ VS \cdot h$) for heterotrophs, ammonia and nitrite oxidizers; and as $mg\ N-NO_3^-_{red}/g\ VS \cdot d$ for nitrate reducers.

Results and discussion

Before both reactors were connected in series, they were operating for 25 days as single steps. The feed for each reactor was prepared by trying to simulate the operational condition once both reactors were connected. The anaerobic reactor was fed with the industrial wastewater diluted ten times (to simulate recirculation) and 100 mg $N-NO_3^-/L$ was added. The feed for the biofilter was composed by the effluent from the anaerobic reactor in which ammonia concentration was adjusted to 200 mg $N-NH_4^+/L$. From the results obtained in this period it can be indicated that:

Taking into account that the influents were diluted, COD reduction was considerable in both reactors, reaching values around 64% for the UASB and more than 50% for the biofilter.

In the UASB almost a complete transformation of the organic nitrogen to ammonia (ammonification) took place and although the seed sludge was an anaerobic one, denitrification occurred from the first day. In the biofilter nitrification efficiencies higher than 90%

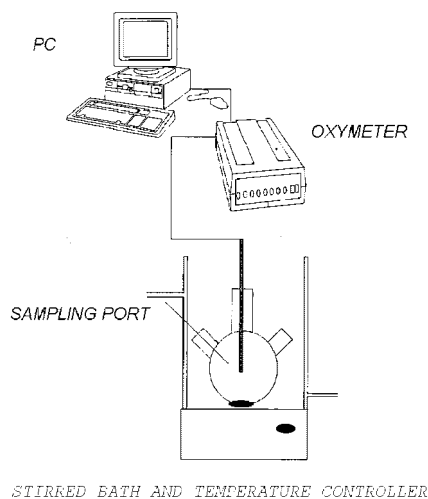


Figure 2 Experimental set-up for aerobic and denitrifiers activity test

were reached, and although during the first 10 days the final product was nitrite, after that the nitrification went to nitrate.

Once both reactors were operating according to the desired conditions they were connected. During the operation some conditions were changed and 10 different periods can be considered. The operational conditions, mainly organic loading rate (OLR), were similar to those applied in the industrial UASB. Table 2 shows these periods together with the removal of organic matter and nitrogen and the flow and composition of the gas generated in the UASB. Organic loading rate and nitrogen loading rate are shown in Table 3. From period I to period VIII, HRT reflected in Table 2 is referred to the total HRT in the whole system, being the HRT in the UASB 3.3 times the HRT in the biofilter. However for the periods IX and X both HRT are shown because the volumes of the reactors were slightly changed and thus the HRT ratio also changed. During the period VI, due to the lack of industrial wastewater, the system was fed with a mixture in which organic matter was supplied as beet sugar molasses and nitrogen as a mixture of NH_4Cl and $(\text{NH}_4)_2\text{SO}_4$ with concentrations similar to those of the industrial wastewater. After this period the reactor was stopped for 20 days.

In the experimental period HRT and recirculation/feed ratio (Q_R/Q_F) were changed. An increase in Q_R/Q_F ratio permits a higher nitrogen removal (more nitrate is recirculated to the UASB), however this parameter is limited by the hydrodynamics in the UASB. An increase in upflow velocity (increase in Q_R/Q_F) can increase the quantity of suspended solids in the effluent, and can also reduce the level of ammonification.

In order to get good conditions for the nitrification, the aerobic reactor operated with a dissolved oxygen concentration between 4.5 and 6.5 mg/L. In spite of the recirculation, dissolved oxygen level in the UASB was negligible. Although the pH of the feed increased during the operational period from 5.2 to 6.3, the effluent pH of the UASB was very stable around 7.5, except in the last two periods in which the pH increased to 8.2. A similar behaviour was observed in the biofilter with a pH around 8.5 except during the last two periods in which the pH increases to almost 9.0. Nitrification efficiency in those periods was not affected by the high pH. This pH variation can be due to the increase in denitrification percentage that occurred in those periods, together with the increase in recirculation flow.

Figure 3 shows the evolution of ammonia in influent and UASB and UBAF effluents. Ammonia was detected in the UBAF effluent during the 20 first days of operation, probably due to the acclimation of the microorganisms to the new operational conditions. The increase of ammonia concentration in the UBAF effluent during the period IV was due to a sharp increase in the influent flow. Once the flow was decreased to the previous values the reactor showed a favourable behaviour and nitrification started again. Figure 4 shows nitrite and nitrate concentrations in the effluents of UASB and UBAF; as can be observed

Table 2 Operational parameters and COD and Nitrogen removal for UASB and UBAF

Period	Day	HRT (d)		Q_R / Q_F	% Removal			Gas flow (L/d)	CH_4 (%)	N_2 (%)
		UASB	UBAF		COD	Nitrification	N_{total}			
I	1–27	5.20	2.40	2.40	97	71	51	13.72	86.25	1.31
II	27–34	5.20	4.24	4.24	97	98	70	12.76	71.5	5.09
III	34–41	4.51	3.68	3.68	96	97	59	14.73	–	–
IV	41–49	3.25	2.65	2.65	97	81	62	18.89	71.97	3.17
V	49–59	6.50	5.30	5.30	97	91	58	11.6	73.81	7.665
VI	59–87	13.00	10.60	10.60	Synthetic wastewater			5.34	–	–
VII	106–140	6.50	5.00	5.00	99	98	84	7.35	75.96	11.46
VIII	140–164	5.20	5.45	5.45	98	98	87	10.38	75.64	10.78
IX	164–192	6.67	2.67	10.4	98	98	89	2.81	76.01	16.37
X	192–205	3.33	1.33	6.67	98	98	91	4.07	75.29	18.93

Table 3 Organic loading rate (OLR) and nitrogen loading rate (NLR) for the different experimental periods

Period	I	II	III	IV	V	VII	VIII	IX	X
OLR									
Kg COD/m³d	2.11	2.25	2.44	2.76	1.52	1.34	1.59	1.27	2.64
NLR									
Kg TKN/m³d	0.12	0.13	0.17	0.20	0.09	0.10	0.12	0.08	0.16

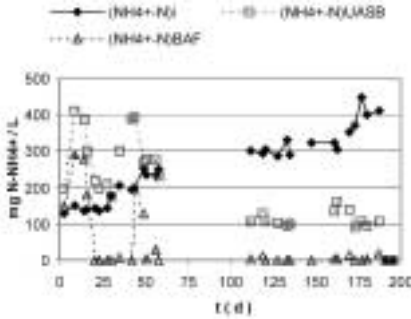


Figure 3 Ammonia evolution

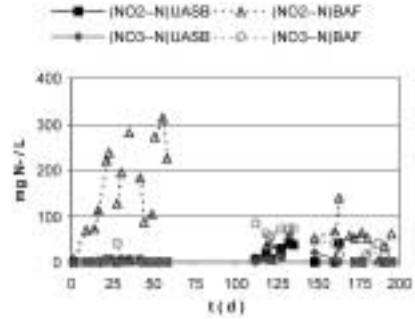


Figure 4 Nitrite and nitrate evolution

oxidised nitrogen concentration in UBAF effluent is quite dependent on the recycle ratio. For the last period in which the recycle ratio was between 5 and 10, oxidised nitrogen concentration in UBAF effluent is almost always below 100 mg/L, but this value was even higher than 200 mg/L for lower recycle ratios.

After a 20 day stop, some changes were detected in the behaviour of the system. Nitrification percentage was not affected, but denitrification increased from 60% to more than 80%, reaching an average value of 91% at the end of the experimental period (Figure 5). It must be mentioned that before the stop, ammonia was transformed in the UBAF mainly to nitrite, but during the first days after the restart-up nitrate was the major oxidised nitrogen compound. This situation evolved to the former one in a few days, obtaining nitrite in the effluent. This situation is in accordance with the results of the activity test shown in Table 4 in which can be seen that the activity of the nitrite producing microorganisms is ten times the activity of the nitrate producers. Heterotrophic activity in the biofilter is very low because organic matter entering the biofilter is also very low.

Methanogenic and denitrifying activity in the UASB (Table 4) increased 4 and 3 times respectively from the initial to the final ones, obtaining a very high value for methanogenic activity of the anaerobic sludge (1.05 g COD/g VS d) at the end of the experimental period.

Suspended solids concentration in the final effluent were very low, usually around 10 mg TSS/L, instead of the concentration of suspended solid in the UASB effluent, which

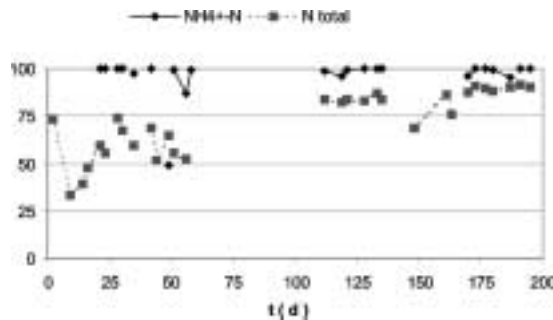


Figure 5 Total nitrogen and ammonia removal in the experimental period

Table 4 Initial and final activity of the anaerobic and aerobic sludges

	Methanogenic activity (g COD/g VS d)	Denitrifying activity (mg N/g VS d)	Ammonia oxid. to nitrite activity (mg O ₂ /g VS h)	Nitrite oxid. to nitrate (mg O ₂ /g VS h)	Heterotrophic activity
Initial	0.25	10.5			
Final	1.05	32.8	47.6	4.4	0.8

was around 170 mg/L. Gas composition was determined throughout the experimental period, obtaining a very constant percentage of methane around 75% for the whole period, however nitrogen percentage in the gas increased as denitrification did, reaching a value around 16%. Carbon dioxide percentage decreased as nitrogen percentage increased, obtaining final values around 7%. Logically, gas production was very dependent on influent flow. According to these values it can be seen that, for this wastewater, recirculation of the nitrified effluent and denitrification in the anaerobic reactor did not considerably decrease the gas quality, and can be used as energy source.

Mass balance

From the data obtained in the period X, COD and nitrogen mass balances can be done in order to corroborate the material flux. In the mass balance for the nitrogen it has to be taken into account that the nitrogen present in the influent and not in the effluent, will be denitrified or assimilated for microbial growth.

Influent nitrogen = effluent nitrogen + nitrogen in the gas + nitrogen assimilated

$$N_{inf} = 726 \text{ mg/L (mainly as organic nitrogen)}$$

$$N_{effl} = 10 \text{ mg NKT/L} + 36 \text{ mg N-NO}_2^-/\text{L} + 15 \text{ mg N-NO}_3^-/\text{L} = 61 \text{ mg N/L}$$

$$N_{gas} = 520 \text{ mL N}_2/\text{L}_{feed} \Rightarrow 20.4 \text{ mmol N}_2/\text{L}_{feed} \Rightarrow 573 \text{ mg N/L}$$

According to these data, 92 mg N/L are employed in biomass growth. Considering a nitrogen content in the biomass of 8.5%, the biomass yield will be 1080 mg VSS/L, equivalent to 1535 mg COD/L. This value for biomass yield is in accordance with the expected values for this system.

For the COD, the soluble COD in the influent will be transformed in CH₄, employed as a carbon source in denitrification, oxidised in the UBAF, will leave the system with the effluent or will be transformed into biomass.

Influent COD = effluent COD + COD for denitrification + COD methanised + COD oxidised + biomass

$$\text{COD}_{inf} = 10000 \text{ mg/l}$$

$$\text{COD}_{effl} = 190 \text{ mg/L}$$

$$\text{COD}_{CH4} = 2035 \text{ mL CH}_4/\text{L}_{feed} \Rightarrow 5815 \text{ mg COD/L}$$

$$\text{COD}_{denit.} = 520 \text{ mL N}_2/\text{L}_{feed} \Rightarrow 20.4 \text{ mmol N}_2/\text{L}_{feed} \Rightarrow 1450 \text{ mg COD/L}$$

$$\text{COD}_{biomass} = 1535 \text{ mg/L}$$

The COD balance agree within a 10% that can be due to gas losses with the effluent. According to the balance the biomass yield is 0.1 g VS/g COD which is an acceptable value for a system in which an anaerobic degradation of organic matter and denitrification takes place.

Conclusions

Effective treatment of an industrial wastewater with a high COD and organic nitrogen content can be achieved with a combined system formed by a UASB and a UBAF. Organic matter removal was maintained above 95% for the whole operational period, with COD effluents as low as 190 mg/L. Almost complete hydrolysis of organic nitrogen to ammonia was achieved, and total nitrogen in the effluent was below 70 mg N/L (13 mg NKT/L and 56 mg NO_x^- /L).

As expected the recycle ratio from the aerobic reactor has a great influence over nitrogen removal in the system because a higher recycle ratio reduces oxidised nitrogen concentration in the effluent. For this system operating with a recycle ratio of 6.7 more than 90% of nitrogen removal was achieved.

Methane content of the gas produced in the UASB was very constant with an average value of 75%, the percentage of nitrogen varied with denitrification rate, reaching a value of 16%. The other measurable gas was CO_2 . Due to the high content of methane, the biogas can be used as energy source, and its quality was not appreciably modified by denitrification process.

Acknowledgements

This work has been supported by the Spanish Commission of Science and Technology (CICYT projects AMB98 0833 and 1FD97-2175).

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