The SHARON process in the treatment of landfill leachate

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Abstract

The purpose of this paper was to study the partial nitrification of the nitrogen present in a landfill leachate applying the SHARON process in order to obtain a suitable effluent to the ANAMMOX process. As a first step, the SHARON reactor was fed anaerobically pre-treated leachate at an ammonium concentration of 2,000 mg N/L (1.1 kg N/m^3 d). In such conditions, the average ammonium and nitrite concentrations in the effluent were 775 mg N/L and 1,225 mg N/L, respectively. During this period the COD removal was very low since most of the biodegradable organic matter was removed in the anaerobic pre-treatment. Afterwards, the SHARON reactor was fed leachate without a previous treatment and the efficiency of the partial nitritation diminished. As well, the COD removal increased, achieving a percentage around 28%.

Keywords
Leachate; partial nitrification; SHARON process

Introduction

The increase in the lifestyle and the industrial and commercial growth has been accompanied by rapid increases in both the municipal and industrial solid waste production. Landfill disposal of solid wastes has been the most common destination for solid wastes throughout the world, particularly for municipal solid wastes (Renou et al.2008). Leachate is the aqueous effluent generated as a consequence of rainwater percolation through wastes and the inherent water content of wastes themselves. Therefore, landfill leachate is a complex wastewater whose composition depends on the age of the landfill, the type of wastes in the landfill, the seasonal variations, etc.

The selection of the best treatment for the landfill leachate depends on its composition (Zgajnar et al. 2009). Usually, application of biological treatment alone is not a good option due to the leachate characteristics: high COD and ammonium concentrations, toxic compounds, refractory organic matter, etc. Furthermore, neither biological nor chemical treatment separately achieves high treatment efficiencies. Consequently, many authors have investigated different treatment systems as biological, chemical or physical processes and different combinations between them. The most effective treatment is usually obtained with the combination of several treatment technologies in order to reach quality standards for discharge (Wiszniowski et al. 2006).

Biological systems offer good results in removing organic and nitrogenous matter from leachate when the biodegradability is high, at BOD/COD ratios higher than 0.5 (Renou et al. 2008). However, at BOD/COD ratios lower than 0.30, physical – chemical processes are usually more effective than biological treatments (Alvarez-Vazquez et al. 2004). In general, leachate generated from young landfills is characterised by high concentrations of both organic and nitrogen compounds. Conventional processes for the removal of organic and nitrogen compounds, such as nitrification and denitrification, can become expensive if an external carbon source is necessary to complete the denitrification, e.g. when the organic compounds present in the leachate are not biodegradable. Consequently, alterna- tive nitrogen removal systems are being developed.

Recently, some authors reported landfill leachate treatment by partial ammonium oxidation to nitrite. Spagni et al. (2008) and Spagni & Marsili-Libelli (2009) studied the nitrogen removal via nitrite of sanitary landfill leachate in a sequencing batch reactor. Nitrification and nitrogen removal were usually higher than 98 and 95%, respectively, whereas COD removal was approximately 20 – 30% due to the low biodegradability of organic matter in the leachate. Liang & Liu (2007) investigated the partial nitritation for landfill leachate treatment using a bench scale fixed bed bio-film reactor. Applying ammonium loads from 0.2 to 1.0 kg N/m3 d, the steady partial nitritation was achieved with an efficiency higher than 94% and obtaining a nitrite to ammonium ratio between 1.0 and 1.4. Ganigue´ et al. (2007, 2008) studied the nitrification via nitrite of the leachate using a sequencing batch reactor. Stable partial nitritation was reached treating high ammonium loads (1 – 1.5 kg N/m3 d), demonstrating the feasibility of this technology as a previous step of ANAMMOX process.

Other innovative alternatives for nitrogen removal are SHARON (Single reactor system for High activity Ammonium Removal Over Nitrite) and ANAMMOX (ANaerobic AMMonium OXidation) processes (van Dongen et al. 2001). In the SHARON process the partial nitrification takes place, working at high temperature (around 35ºC) and without retention sludge. In these conditions, nitrite oxidisers are selectively washed out. In the ANAMMOX process, the oxidation of ammonium to nitrogen gas iscarried out anaerobically using nitrite as electron acceptor, without external carbon source. In this process the conversion takes place in a molar $NH_4^+ : NO_2^-$ ratio of 1:1.32. Therefore, in order to combine SHARON and ANAMMOX systems, the SHARON effluent should contain ammonium and nitrite in this ratio, though studies about the use of the SHARON process in the treatment of landfill leachate were not found in the literature.

This work is integrated in a project about the global treatment of a leachate from a landfill of urban solid wastes. In this project the organic matter removal was undertaken by biological and advanced oxidation processes, while the combination of the SHARON and ANAMMOX processes was proposed for the nitrogen removal. The aim of this work was to study the partial nitrification of the nitrogen present in the landfill leachate applying the SHARON process in order to obtain a suitable effluent to the ANAMMOX process. As a first step, the SHARON reactor was fed anaerobically pre- treated leachate and afterwards, raw leachate was used.

Materials and methods

SHARON reactor

A lab-scale continuous stirred tank reactor (CSTR) with an effective volume of 1.9 L was used. The reactor was inoculated with sludge from a partial nitrification reactor treating wastewater from an aminoplastic resin producing factory. The influent was supplied to the reactor with a peristaltic pump at 1.06 L/d, maintaining the hydraulic retention time around 1.8 d. The temperature was kept at 36ºC using a water jacket and the pH was controlled around 7. An air diffuser located at the bottom of the vessel supplied oxygen from an air pump, maintaining the dissolved oxygen concentration around 2 mg/L throughout the reactor operation.

Synthetic medium

The start up was carried out feeding a synthetic solution with an ammonium concentration of 1,000mgN/L and a molar $HCO_3^- : NH_4^+$ ratio of 4:3. The solution was supplemented with 1mL/L of micronutrients (Eiroa et al. 2004) and 250 mL/L of a nutrient solution which contained (g/L): $MgSO_4 \cdot 7H_2O$ 0.24, KH_2PO_4 0.50, NaCl 2.00 and $CaCl_2 \cdot 2H_2O$ 0.16.

Landfill leachate

The used leachate was collected from a landfill of urban solid wastes in the province of A Coruña (Spain). The leachate was characterised before evaluating the possible configurations for its global treatment. The biodegradability is one of the parameters commonly used in order to select the best process for its treatment. In this study, the BOD/COD ratio was around 0.37; in such a way a biological system can be effective in order to remove the biodegradable organic matter. Consequently, the organic matter present in the leachate was reduced by anaerobic treatment (Vilar et al. 2008), which was carried out in a lab-scale Upflow Anaerobic Sludge Blanket Reactor (UASB). The anaerobically pre-treated leachate used in the period I of this study was obtained feeding the anaerobic reactor with raw leachate diluted 1/5. In the period II, the raw leachate was diluted 1/5 and fed to the SHARON reactor without a previous anaerobic treatment. In both periods, ammonium and bicarbonate concentrations were adjusted to a molar HCO_3^- : NH_4^+ ratio of 4:3 before feeding the SHARON reactor.

Analytical methods

Nitrite and nitrate anions were analysed by capillary electrophoresis using a ^{3D}CE system Hewlett Packard with a micro capillary tube of fused silica. A sodium phosphate solution was employed as the electrolyte and UV detection was undertaken at a wavelength of 214nm and 450nm as reference. Ammonium concentration was determined by a colorimetric method based on the reaction of ammonium anion with hypochlorite and phenol. The absorbance of the compound obtained was determined at 635nm using a UV/VIS spectophotometer (Lambda 11, Perkin Elmer). Volatile suspended solids (VSS), total suspended solids (TSS), total Kjeldahl nitrogen (TKN), dissolved oxygen (DO), alkalinity and pH were evaluated according to Standard Methods (1998). Chemical oxygen demand (COD) was also analysed and recalculated according to Standard Methods (1998), taking into account the interference due to the presence of nitrite (nitrite exerts a COD of 1.1mgO_2/mg N-NO_2^-). Biological oxygen demand (BOD) was determined using BOD systems (Velp Scientifica) in which the internal pressure is translated by a microprocessor directly into BOD. In this study, the BOD was measured after 20 days since an initial period was necessary for acclimatising the sludge to the leachate.

Results

Initially, the characterisation of the landfill leachate was undertaken in order to propose the appropriate configuration to remove organic and nitrogen compounds using different combinations of biological and chemical processes. The composition of the leachate is presented in Table 1. It is characterised by high organic matter and nitrogen concentrations. Therefore, the main goal in the treatment of this leachate is the organic matter and nitrogen removal. The organic matter removal was undertaken by anaerobic treatment and advanced oxidation processes (Vilar et al. 2006, 2008), while the combination of the SHARON and ANAMMOX processes was proposed for the nitrogen removal. In this paper, the results obtained in the SHARON process are presented.

Table 1. Characterisation of the landfill leachate (all parameters in mg/L, except pH)

pH	8.1–8.6
CODtotal	8,760–12,110
CODsoluble	7,770–10,900
BOD	2,300–4,590
N- NH_4^+	3,260–5,910
TKN	3,400–5,700
Alkalinity	16,380–18,760
TSS	47–166
VSS	46–124

The SHARON reactor was inoculated with sludge from a partial nitrification reactor and the start up was carried out feeding with synthetic solution. During the start up, ammonium loading rate was around 0.56 kgN/m^3 d and the obtained nitrite percentages were above 50% (data not shown). After the start up, the SHARON reactor was fed anaerobically pre-treated leachate (period I) and afterwards, raw leachate was used (period II).

Period I: anaerobically pre-treated leachate

In this period, the SHARON reactor was fed anaerobically pre-treated leachate and the ammonium and bicarbonate concentrations were adjusted. Initially, the ammonium concentration in the influent was maintained at 1,000mgN/L (Figure 1A), the loading rate being 0.56 kgN/m3 d. The variation in the obtained ammonium and nitrite concentrations in the effluent was fairly high, the average values being 430 and 580 mgN/L, respectively. Afterwards, the ammonium in the influent was increased to 1,500 and 2,000mgN/L (0.83 and 1.11 kgN/m^3 d). In the last conditions, the average concentrations in the effluent were 775 mgN/L for ammonium and 1,225mgN/L for nitrite. The nitrate concentration in the effluent was very low during all the operation time.

The free ammonia and free nitrous acid concentrations in the SHARON reactor were evaluated (Figures 1B and C). During this period, the values of free ammonia were around 9.68mgNH_3/L; below the range estimated by Anthonisen et al. (1976) for the inhibition of ammonium oxidiser bacteria (10–150mg NH_3/L). On the other hand, the average free nitrous acid concentration was 0.51mg HNO_2/L; within the range estimated by Anthonisen et al. (1976) for the inhibition of nitrifying organisms (0.22–2.88mg HNO_2/L). However, partial nitritation took place, thus free nitrous acid did not cause inhibition in our study.

With regard to organic matter, during this period the COD removal was very low (Figure 2). This fact showed that most of the biodegradable organic matter was removed in the anaerobic pre-treatment and the organic compounds present in the influent of the SHARON reactor were refractory. Therefore, the recalcitrant compounds of the landfill leachate did not affect the efficiency of partial nitritation.

Period II: raw leachate

Afterwards, the SHARON reactor was fed leachate with the same dilution as in the previous period but without a previous treatment. The ammonium and bicarbonate concentrations were also adjusted, maintaining an ammonium concentration in the influent of 2,000mgN/L (1.11 kgN/m^3 d). As it is shown in Figure 1A, the efficiency of the partial nitritation diminished when the raw leachate was used. The ammonium concentration in the effluent increased while the nitrite concentration decreased.

The contribution of free ammonia and free nitrous acid to the inhibition of the partial nitritation during this period was evaluated (Figures 1B and C). During this period, as it was expected, the free ammonia concentration in the reactor increased while the free nitrous acid concentration decreased. The values of free ammonia were from 9.98 to 42.35mg NH_3/L and the free nitrous acid concentration decreased from

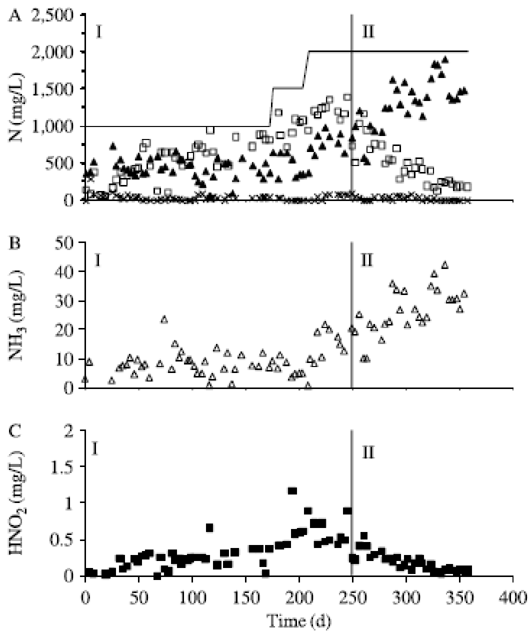

Figure 1 | Evolution of nitrogen compounds. A: Ammonium in the influent (—) and ammonium (▲), nitrite (□) and nitrate (×) in the effluent. B: Free ammonia in the reactor (△). C: Free nitrous acid in the reactor (■). Period I: anaerobically pre-treated leachate, period II: raw leachate.

0.55 to 0.05mg HNO_2/L. According to these data, it is unlikely that free nitrous acid caused inhibition, since its concentration was lower than in period I in which there was no inhibition. With regard to free ammonia, its concentration in the reactor 25 days before and after the change of the feed was very similar, around 16.23 and 17.79mg NH_3/L, respectively. However, the partial nitritation started to decrease in the first days of period II, reaching values of 31% after 25 days of the new conditions. The decrease of the partial nitritation took place before the free ammonia concentration increased slightly. Therefore, the presence of free ammonia could favour the low partial nitritation percentages that was obtained, but was not the main reason for its decrease.

During this period, the organic matter concentration in the influent was about 2,526mgCOD/L and the COD removal achieved percentages around 28% (Figure 2). As it was expected, the organic matter removal increased with regard to period I since the BOD/COD ratio of the raw leachate was about 0.37. It seems that the presence of biodegradable organic matter was the reason of the low efficiency of the SHARON process during period II since the recalcitrant compounds of the landfill leachate did not

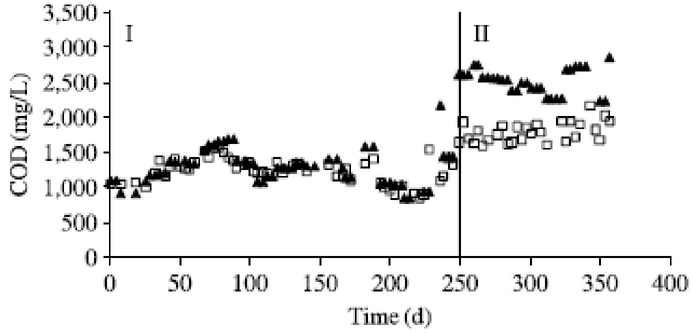

Figure 2 | COD in the influent (▲) and in the effluent (□) of the SHARON reactor. Period I: anaerobically pre-treated leachate, period II: raw leachate.

affect its efficiency (period I). A competition between the ammonium oxidiser bacteria and the heterotrophic bacteria could take place due to the organic matter removal. This is corroborated by the increase in the sludge concentration in the reactor during period II (Figure 3). When the reactor was fed anaerobically pre-treated leachate the VSS concentration was around 400 mg/L. However, when raw leachate was added the VSS concentration increased up to 2,500 mg/L.

Comparing the obtained results with the literature, Ganigué et al. (2007) also studied the nitrification via nitrite of an urban landfill leachate. The leachate was characterised by a low biodegradability (BOD/COD ratio around 0.15). An organic matter concentration between 3,500 and 4,500mgCOD/L was fed, obtaining a COD removal between 11 and

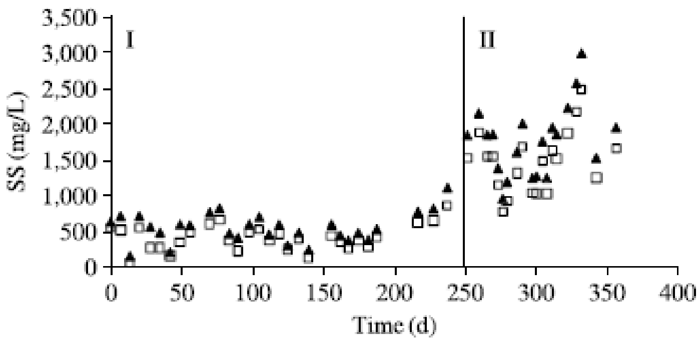

Figure 3 | TSS (▲) and VSS (□) in the SHARON reactor. Period I: anaerobically pre-treated leachate, period II: raw leachate.

14%. Stable partial nitritation was reached treating high ammonium loads (between 1 and 1.5 kgN/m^3 d) in spite of the organic matter removal. The difference with regard to our study could be due to the different reactor configuration. Ganigué et al. used a sequencing batch reactor with an average sludge retention time of 5 days. In our study, the SHARON reactor without biomass retention could cause the washout of the ammonium oxidiser bacteria due to the competition with the heterotrophic bacteria.

Conclusions

When the SHARON reactor was fed anaerobically pretreated leachate at an ammonium concentration of 2,000mgN/L (1.11 kgN/m^3 d), the average concentrations in the effluent were 775mgN/L for ammonium and 1,225mgN/L for nitrite. With regard to organic matter, during this period the COD removal was very low. The recalcitrant compounds of the landfill leachate did not affect the efficiency of partial nitritation.
When the SHARON reactor was fed raw leachate maintaining the ammonium concentration, the efficiency of the partial nitritation diminished. The presence of free ammonia could favour the low partial nitritation percentages, but was not the main reason for its decrease. As it was expected, the organic matter removal increased since the BOD/COD ratio of the raw leachate was about 0.37. It seems that the presence of biodegradable organic matter was the reason of the low efficiency of the SHARON process.
According to the obtained results, in order to remove the nitrogen from the leachate by the SHARON process it is necessary to remove previously the biodegradable organic matter. The low biodegradability seems to be one of the key factors to reach the good development of the system.

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