New hybrid reactor concept incorporating a filter mesh for nitritation–anammox treatment of sludge return liquid

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ABSTRACT

A new approach to perform partial nitritation–anammox in a single tank was investigated. The tank incorporated a mesh (opening size 1.0 × 1.2 mm) as permeable barrier to create two distinct reaction zones (aerated and anoxic). The study reports on the operation and optimization of a 13 L laboratory scale reactor to treat sludge reject water with an NH4-N concentration of ~750 mg·L⁻¹. Performance throughout 250 days at increasing nitrogen loading rates is presented. The maximum loading rate applied was 1.5 kg NH4-N·m⁻³·d⁻¹ at a hydraulic retention time of 12 h. Typical composition of the effluent was ~50 mg·L⁻¹ NH4-N; <5–10 mg·L⁻¹ NO2-N and ~60 mg·L⁻¹ NO3-N. The corresponding average N removal rates were 85% for total nitrogen and 90% for NH4-N, respectively. Process control was very simple. It comprised only regulation of the aeration rate to maintain a pre-set pH (7.1) in the nitritation zone. Performance data clearly indicate that the investigated reactor configuration offers distinct advantages over currently installed processes. It demonstrated high robustness without the need for sophisticated process control. Apparently, the use of a permeable mesh to establish different reaction conditions in a single reactor provides new features of high potential.

Key words | anammox, mesh, partial nitritation, single tank hybrid reactor, sludge reject water

INTRODUCTION

Since its discovery in the early 1990s the anaerobic ammonia oxidation, or in short anammox process, has gained significant attention. It was instantly evident that it can beneficially combined with nitritation, i.e. the formation of nitrite out of ammonia, allowing efficient ammonia removal. In comparison to the standard approach, nitritation–denitrification, this process combination provides two major advantages: a reduced oxygen demand and the independence of availability of easily accessible biochemical oxygen demand (Jetten et al. 1997). These days the treatment of highly ammonia-loaded wastewater, in particular the reject water derived from dewatering of anaerobic sludge, via nitritation–anammox is a well-established procedure. Numerous full scale applications have been developed within the last years (Kuenen 2008). Generally the applied processes can be divided into two categories (Jaroszynski & Oleszkiewicz 2011; Lackner et al. 2014): two-step processes where nitritation and anammox occur in two separate reactors and single-stage processes working in a sequential mode, i.e. consecutive phases of aeration/non-aeration in a sequencing batch reactor (SBR). The latter has the advantage of being a single unit process of technical simplicity, however, at the disadvantage that reaction conditions periodically switch from being either favourable for nitritation (aeration phase) or for anammox. In consequence, the volumetric turnover rates are typically inferior to two-stage processes.

Nevertheless, there are certain other issues that militate in favour of the single-stage reactor. On important aspect is the converse impact of the two biological reactions on pH: nitritation leads to a drop in pH whereas the anammox process partially recovers the initial pH. A key factor in this context is the buffer capacity in the form of the bicarbonate ion. Each bicarbonate can absorb one proton. The molar stoichiometric relationship for ammonium oxidation to nitrite is 2 moles HCO₃⁻ per mole NH₄⁺ to prevent a pH drop to nonphysiological levels (Galí et al. 2007). On the other hand, sludge return liquor contains only about 1.2 times more HCO₃⁻ than NH₄⁺ on a molar basis (Fux et al. 2002). Fortunately, according to stoichiometry the optimum ratio of ammonia to nitrite is 1:1.32 for the anammox

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reaction. In other words only 57% of the ammonia needs to be converted to nitrite. Nevertheless, to provide sufficient buffer capacity it is necessary to feed the full amount of substrate into the nitritation step, at the disadvantage that careful control is required to maintain the appropriate NH₄⁻/NO₂⁻ ratio in the outlet (van Dongen et al. 2001). The sequential operation of the two processes in a single reactor leads to lower pH fluctuations resulting in a typical zigzag pattern of the pH course (Wett 2007).

Another issue is that both bacterial processes are subject to considerable substrate or product inhibition caused by ammonia and nitrite (Strous et al. 1999). In aqueous media both nitrogen species exist in the form of two conjugates. In dependence on pH and temperature the ionized forms, ammonium (NH₄⁺) and nitrite (NO₂⁻), are in equilibrium with their un-dissociated forms, free ammonia (NH₃) and nitric acid (HNO₃). It is widely accepted that the latter ones are actually responsible for the deterioration of microbial activity (de Prá et al. 2016). In particular high nitrite levels (or, more correctly, high nitric acid levels) are critical to the development of the control strategy are provided (Jin et al. 2012; Raudkivi et al. 2017).

Well balanced interaction of nitritation and anammox is necessary to avoid self-inhibition through accumulation of these metabolites. In single-stage systems concentrations of these inhibitory substances remain at relatively low level due to the sequential operation of the two microbial reactions (Lackner & Horn 2012), whereas in two-stage systems high nitrite concentrations occur in the outflow of the nitritation stage, which may easily cause process disturbances (van der Star et al. 2007).

The approach chosen here tries to combine the advantages of both types of reactors by the installation of a semi-permeable barrier made of mesh material. By such means two separate reaction zones, aerated and non-aerated, can be maintained. At the same time the permeable mesh allows passage of dissolved ions and such balancing of the different pH levels as well as the rapid transfer of the produced nitrite. Woven or non-woven filter cloths are already applied in wastewater treatment for a wide range of different purposes. This includes sludge separation in filter presses or belt filters, removal of fine suspended solids for effluent polishing or even for biomass retention as a substitute of the secondary clarifier (Grabbe et al. 1998; Fuchs et al. 2005). However, the idea of using mesh filters to establish two different reaction zones in a single tank has not been studied yet.

The present study describes the investigation and optimization of the described approach in a 13 L laboratory reactor. Performance throughout 250 days of operation and details on the development of the control strategy are provided.

MATERIALS AND METHODS

**Bioreactor**

The reactor was made of a rectangular 20 L plastic vessel (340 × 240 × 240 mm) and had a working volume of 15 L. It was dived into two equal compartments by an aluminium frame tightly squeezed into the reactor vessel. The frame was covered with a polyester mesh (opening size 1.0 × 1.2 mm). The whole reactor was immersed in a water bath to maintain a temperature of 35 ± 0.2 °C using an immersion thermostat (ED, Julabo, Germany). Two stirrers (RW 20, IKA, Germany) were implemented to obtain homogeneous mixing of both reaction zones. Initially the inflow was provided from a 60 L barrel placed on a balance, by means of a peristaltic pump (505 U Watson Marlow, USA); later on a 1.000 L bulk container served as substrate storage tank. Two automated pinch valves (SCH284B015 Asco, USA) allowed distribution of the inflow either to the nitritation or alternatively to the anammox compartment. The outlet was a simple overflow established within a small conical settling zone (upper/lower Ø 70/45 mm, height 140 mm). In its centre a small slow-moving rabble rake was mounted to remove gas bubbles. Aeration was done with a ceramic aerator (140 × 25 mm) immersed in the nitritation compartment using pressurized air. Air flow was controlled by a flow indicator regulator (Aera FC-PA7800c, Hitachi, Japan) and a variable area flow meter for visual control. The plant was equipped with an optical oxygen measurement device with two needle-type fibre-optical oxygen sensors (FireStingO2, Pyroscience, Germany) submerged in the two compartments. The device includes also a four-wire PT100 temperature sensor. pH in the nitritation compartment was measured by a standard pH meter (Mettler Toledo, USA). In the start-up phase pH was maintained within the two limits (pH 6.9, 7.8). For this purpose two pH-controlled peristaltic pumps (101 U/R Watson Marlow, USA) were installed dosing 1 M HCl or 1.5 M NaOH, respectively, into the nitritation zone. On-line monitoring, data storage and process control were conducted by means of a PLC (Melsec FX3G Mitsubishi). A scheme of the plant layout is presented in Figure 1.

**Inoculum and substrate source**

Inoculum was obtained from the anammox process (SBR) established at the Strass wastewater treatment plant (WWTP) in Lower Austria. Details on the operation of the plant can be found in Wett (2007). The substrate (feed)
was sludge reject water deriving from the anaerobic digester of the Klosterneuburg WWTP, Lower Austria (55,000 population equivalent). Dewatering of digested sludge is carried out by flocculation and a decanter centrifuge. To reduce variations in NH₄-N concentration, the reject water was artificially spiked with NH₄HCO₃ to achieve an NH₄-N level of around 750 mg·L⁻¹. The composition is provided in Table 1.

Analytical procedures

Routine analysis was performed with photometric cuvette tests (LCK tests + DR 2800 UV/VIS photometer, Hach Lange, Germany). NH₄-N and total Kjeldahl nitrogen (TKN) values were periodically crosschecked according to standard methods (ISO 5664 (ISO 1984a), EN 25663 (CEN 1993)) using a Büchi distillation/titration unit (K370, Büchi, Switzerland). For TKN determination samples were previously digested with sulfuric acid (Digestion Automat K-438, Büchi). pH was measured with a standard laboratory pH meter (WTW 340i + Sentix 41 pH probe, WTW, Germany). Bicarbonate alkalinity was analysed according to ISO 9963-1 (ISO 1984b) using an auto-titration unit (Titrino 721 NET, Metrohm, Switzerland) through titration with 0.1 M HCl to the end point pH 5.0 following a protocol provided by the manufacturer. Dry matter was determined using an automatic moisture analyser with infrared heating system (MA35, Sartorius, Germany). To correct for dissolved dry matter, samples were filtered and analysed in the same way. The difference between the two values obtained corresponds to the suspended solids concentration.

RESULTS AND DISCUSSION

Initial test phase/first experiences

In a first test phase the reactor was operated with artificial wastewater (1,000 mg·L⁻¹ NH₄-N, in the form of
Nitrogen carbonate \( \text{NH}_4\text{HCO}_3 \) at a loading rate of \( 0.2 \text{ kg N·m}^{-3}\cdot\text{d}^{-1} \) for 30 days. In this period implementation and calibration of the different sensors was conducted and basic behaviour of the system was checked. Subsequently the substrate was replaced by real sludge reject liquor. Based on increasing experience several modifications of the set-up were made. At changing loading rates it turned out to be relatively difficult to maintain constant pH levels due to imbalances of the two reaction steps. Sometimes overshooting nitritation occurred but also accumulation of ammonia was observed as indicated by very low or high pH values, respectively. Besides the direct impact of pH itself, attention must be paid to its influence on free ammonia and undissociated nitric acid concentration. Data on the optimum pH range for nitritation/anammox provided in literature are not fully consistent and vary depending on the investigated system (Jaroszynski et al. 2011; Jin et al. 2012). However, generally the neutral to light alkaline range is considered to provide optimum conditions. To avoid nonphysiological pH conditions and to minimize the potential ammonia or nitrate inhibition two dosing pumps for acid and base were installed keeping the pH within the limits of 6.9 and 7.8. As well, an option for diversion of the incoming substrate was implemented and substrate dosage was divided at a ratio of 60:40 (nitritation:anammox compartment) following the stoichiometry of the process. Moreover, due to clogging problems the initially installed relatively tight mesh (0.12 x 0.12 mm openings) was replaced by a mesh with much wider mesh size (1.0 x 1.2 mm, as described in the ‘Materials and methods’ section). With these improvements the reactor performed fairly well, however, with a certain susceptibility to process instabilities. After around 120 days this initial test period had to be stopped when high nitrite accumulation occurred which further on led to inhibition of the anammox bacteria.

**Continuous test run**

In the following it was decided to restart the system applying a different start-up procedure. Instead of using the pure substrate it was diluted with tap water. Maintaining a fixed hydraulic retention time (HRT) (36 h) the dilution factor was gradually decreased to raise the \( \text{NH}_4\text{N} \) loading rate. With this new strategy, start-up and operation turned out to be relatively simple. Within a period of 21 days a loading rate of \( 0.5 \text{ kg N·m}^{-3}\cdot\text{d}^{-1} \) was achieved without notable system imbalances. No major difficulties were observed with respect to high pH fluctuations. Indeed the pH control was not active any more because pH always stayed within the given limits. This remained so until the end of the experiment, with the exception of two phases where the process was heavily disturbed due to operation failure (day 188 and day 289). Later on the pH control was completely turned off. The loading rate of \( 0.5 \text{ kg N·m}^{-3}\cdot\text{d}^{-1} \) was kept for around 50 days. In this period the control strategy to achieve partial nitritation was gradually modified. Initially the aeration of the nitritation compartment was operated in an on-off mode (30 min aeration, 15 min off). The maximum \( \text{O}_2 \) concentration in the oxygenation period was limited to \( 0.2 \text{ mg·L}^{-1} \). To a certain extent this approach was adopted from the full scale SBR that served as the inoculum source. With increasing loading rate the oxygenation interval was manually adjusted to keep the pH within the desired range. Achieving an aeration interval of 30 min on/5 min off, it was decided to proceed with permanent aeration at a constant aeration rate. Finally a flow controller was implemented which varied the aeration rate to maintain a pre-set pH value of 7.1. Again, the choice of the pH set point was based on experience from the full scale SBR (Wett 2007). Variations of the pH set point confirmed that this setting provided best results in terms of total nitrogen (TN) removal efficiency.

Subsequently another increase of the loading rate to \( 0.8 \text{ kg N·m}^{-3}\cdot\text{d}^{-1} \) was made (day 225). In the following period the distribution of the substrate to the two reactor zones was varied. The initial idea was – based on first experience – to avoid overshooting nitrite formation in the first compartment. Several modifications of the dosing ratio (from 50:50% to 100:0% nitritation:anammox) were tested. The applied ratios are illustrated in Figure 2(a). A comparison with results presented in Figure 2(b) and 2(c) shows that little impact on reactor performance or distribution of nitrogen species in the effluent was found. This observation can be attributed to the high permeability of these ions through the mesh barrier. With a look at simplicity of operation, from then on all substrate was delivered into the nitritation compartment. This period ended due to a failure of the substrate pump control. A massive overloading of the reactor occurred which led to the accumulation of ammonia (up to \( 500 \text{ mg NH}_4\text{N·L}^{-1} \), day 289). In response, the substrate supply was stopped until ammonia levels dropped. Subsequently the loading rate was raised again quite quickly to 1.0, subsequently to 1.2 and finally to \( 1.5 \text{ kg NH}_4\text{N·m}^{-3}\cdot\text{d}^{-1} \). Despite the rapid increase in nitrogen loading the process ran very stably and highly reasonable nitrogen elimination rates were observed. An overview of the reactor efficiency and the concentration of nitrogen species in feed and reactor outflow throughout the whole period is provided in Figure 2(a)–2(c).
The achieved NH₄-N conversion rates were typically higher than 90% and the overall TN removal was about 80–85%. NH₄-N concentration in the effluent was 40 mg·L⁻¹ on average and very low nitrite concentrations (typically <5 mg·L⁻¹) were observed. Nitrate concentration in the outflow was around 86 (±35) mg·L⁻¹ at loading rate <1 kg N·m⁻³·d⁻¹ and 63 (±16) mg·L⁻¹ at higher loading rates. This corresponds to a specific nitrate production per N removed of 0.13 (±0.5) and 0.09 (±0.3) g NO₃⁻·g⁻¹ N, respectively. The measured values are well in line with the proposed stoichiometry according to which 11% of the ammonia removed occurs in the form of nitrate. Certain studies on successful nitritation–anammox in laboratory and full scale report lower nitrate production rates in the range of 5–6% (Wett 2007; Zhang et al. 2010). Generally it is assumed that simultaneous denitrification under anoxic conditions is responsible for that. However, other plants exhibit nitrate levels higher than anammox stoichiometry due to
formation of nitrate by nitrite oxidizing bacteria (NOB) (Lackner & Horn 2012). To what extent NOB were present in the current reactor configuration was not examined. Direct measurement of NO$_3^-$ formed in the nitritation zone was not possible due to the exchange of ions between the two compartments. The generally applied approach to exclude NOB is through control of the HRT. At elevated temperatures (>30 °C) the growth rate of ammonium oxidizing bacteria are usually higher than those of NOB. Therefore in suspended biomass systems selective wash-out of NOB is not possible due to parallel nitrite consumption. Nevertheless, this value can be estimated based on stoichiometry of the anammox reaction. According to that, at highest loading the nitrite production rate was 1.5–1.6 kg N-m$^{-3}$·d$^{-1}$ at HRT of 6 h. For the anammox part the average nitrogen consumption rate reported by Fux et al. was 0.6 kg N-m$^{-3}$·d$^{-1}$. They mention that this rate was limited by nitrite availability due to the SBR mode and presume that a nitrogen elimination rate of 1.0 kg N-m$^{-3}$·d$^{-1}$ at HRT of 16–24 h in the anammox zone is feasible. In the reactor configuration presented here the maximum nitrogen consumption rate was 2.7–2.8 kg N-m$^{-3}$·d$^{-1}$ at HRT of 6 h.

**Comparison with performance of established nitritation/anammox processes**

Lackner et al. (2014) made a survey on full scale partial nitritation/anammox processes. A comparison of the data on treatment of sludge return water provided there with reactor performance in the present study is given in Table 2.

Being aware that a comparison of a laboratory plant under well controlled conditions with a practical operation is not fully justified, Table 2 shows that the investigated type of reactor outperforms current full scale applications. The overall treatment capacity is even better than advanced immobilized or two-stage systems. Only the second (anammox) stage in Rotterdam, a high rate tower reactor with suspended carriers (van der Star et al. 2007), has a higher volumetric loading rate (7.05 kg N-m$^{-3}$·d$^{-1}$). Still the overall volumetric turnover of the two stages in combination is lower than the one achieved in the present experiments. It should be mentioned that in other investigations at laboratory scale even much higher conversion rates were achieved. Nitrite formation rates as high as 4.7 kg N-m$^{-3}$·d$^{-1}$ (Wang et al. 2016) and nitrogen removal rates through anammox of up to ~75 kg N-m$^{-3}$·d$^{-1}$ (Tang et al. 2011) are reported. However, most studies were conducted with artificial wastewater using complex reactor configurations. Moreover, they addressed only one of the two partial reactions.

The most convincing argument for the investigated reactor type was the simplicity of plant control. According to
Table 2 | Performance data of full scale plants treating sludge return liquor (data from Lackner et al. 2014) in comparison of with results obtained in the present study

<table>
<thead>
<tr>
<th>Process</th>
<th>Amersfort</th>
<th>Apeldoorn</th>
<th>Balingen</th>
<th>Heidelberg</th>
<th>Ingolstadt</th>
<th>nieuwegine</th>
<th>plettenberg</th>
<th>Zürich</th>
<th>Malmö</th>
<th>Landshut</th>
<th>rotterdam</th>
<th>Present study</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>DEMON</td>
<td>DEMON</td>
<td>DEMON</td>
<td>DEMON</td>
<td>SBR</td>
<td>DEMON</td>
<td>SBR</td>
<td>ANITAMox</td>
<td>Terrana</td>
<td>suspended carriers</td>
<td>suspended sludge</td>
<td>suspended sludge</td>
</tr>
<tr>
<td>No. of stages</td>
<td>1-stage</td>
<td>1-stage</td>
<td>1-stage</td>
<td>1-stage</td>
<td>1-stage</td>
<td>1-stage</td>
<td>1-stage</td>
<td>suspended carriers</td>
<td>2-stage</td>
<td>suspended carriers</td>
<td>(2nd stage)</td>
<td>suspended sludge</td>
</tr>
<tr>
<td>Reactor volume</td>
<td>m³</td>
<td>780</td>
<td>2,400</td>
<td>705</td>
<td>2 × 570</td>
<td>2 × 560</td>
<td>450</td>
<td>134</td>
<td>2 × 1,400</td>
<td>4 × 50</td>
<td>288 + 495</td>
<td>1,800 + 70</td>
</tr>
<tr>
<td>TSS g L⁻¹</td>
<td>4.5</td>
<td>3.5–4</td>
<td>1.2</td>
<td>1.0–2.5</td>
<td>2.0–4.0</td>
<td>–</td>
<td>3</td>
<td>3.5–4.5</td>
<td>16ᵃ</td>
<td>10–12, 5–6</td>
<td>0.27, 7–10ᵃ</td>
<td>3–4</td>
</tr>
<tr>
<td>HRT h</td>
<td>26</td>
<td>58</td>
<td>94</td>
<td>114</td>
<td>75</td>
<td>42</td>
<td>40</td>
<td>45</td>
<td>24</td>
<td>(25, 42)</td>
<td>(36, 6) 42ᵇ</td>
<td>(6, 6) 12</td>
</tr>
<tr>
<td>Vol. loading kg N m⁻³ d⁻¹</td>
<td>0.65</td>
<td>0.54</td>
<td>0.04–0.11</td>
<td>0.20</td>
<td>0.18–0.20</td>
<td>0.61</td>
<td>0.45</td>
<td>0.4</td>
<td>1.0–1.2</td>
<td>(1.11, 0.65) 0.41ᵇ</td>
<td>(0.27, 7.03) 0.26ᵇ</td>
<td>(3.0, 3.0) 1.5</td>
</tr>
<tr>
<td>Sludge loading g N-kg TSS⁻¹ d⁻¹</td>
<td>145</td>
<td>155</td>
<td>35–95</td>
<td>119</td>
<td>71</td>
<td>–</td>
<td>149</td>
<td>134</td>
<td>64</td>
<td>101, 108</td>
<td>–, 238</td>
<td>790, 640</td>
</tr>
</tbody>
</table>

ᵃEstimated data as given by Lackner et al. (2014).
ᵇValues for the overall plant, calculated from the data provided (data for the individual stages are listed in brackets).
SSR: suspended sludge reactor; TSS: total suspended solids.
Lackner et al. (2014) online monitoring and control are important aspects in the operation of any type of partial nitritation/anammox system. Due to the rather complex combination of microbial processes compared to conventional nitrification/denitrification processes, there is a high demand on online sensors. In the current experiments several control strategies were tested. However, the simplest one turned out to be the one providing best results. Process control relied basically on a single pH sensor in the nitritation compartment which regulated the aeration rate. The two oxygen sensors served merely as a monitoring tool. Despite the low detection limit of the sensor (0.02 mg L⁻¹, according to specification) measurable levels of O₂ in the nitritation zone occurred only at low to medium loading rates. At the highest loading rate, O₂ was below the detection limit. In the anammox compartment no O₂ was measured except for rare cases with significant process disturbances. At the test conditions the two-phase reaction turned out to be a self-regulating process and no additional measures to balance nitritation and anammox were necessary. Moreover, even after process failure, recovery of the system was very quick.

Despite the obvious advantages, also potential hurdles for further scale-up should be noted. One issue is the inappropriate function of the settling device. However, it is expected that this difficulty can be relatively easily overcome at a larger scale. Three-phase settlers are a standard tool in, for example, anaerobic wastewater treatment processes (van Lier et al. 2010). Last but not least the application of a permeable mesh is a completely new approach and no experience is available on practical implementation of such a reactor configuration. Nevertheless, the tested set-up has demonstrated high potential. It seems therefore well worth further investigation.

CONCLUSIONS

A new approach to perform partial nitritation–anammox in a single tank was tested incorporating a mesh barrier to achieve two distinct reaction zones. In a successful long-term test run, proof-of-concept was demonstrated. Performance data obtained clearly indicate that the investigated reactor configuration provides distinct advantages over other currently installed processes. In particular it combines high robustness with simple process control. Moreover, the achieved loading rates were significantly higher than performance data reported for other reactor configurations. It is obvious that these first results need further confirmation at a larger scale. Nevertheless, the use of a mesh to establish different reaction conditions provides interesting new features which are well worth further research. Finally, it should be noted that the basic principle of the reactor design might be also transferred to other two-stage processes such as nitrification–denitrification.

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REFERENCES


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