BIOLOGICAL SLUDGE LIQUOR TREATMENT AT MUNICIPAL WASTEWATER TREATMENT PLANTS – A REVIEW

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Abstract

Separate biological treatment of sludge liquor, produced when dewatering digested sludge at wastewater treatment plants, can be favourable in achieving sufficient nitrogen removal. The treatment has the potential to decrease volume requirements and electrical energy and external carbon consumption. The characteristics of sludge liquor (high temperature, high ammonium concentration, low COD:N ratio) support high autotrophic growth rates. These conditions also favour nitrite accumulation, which makes a short-cut nitrification-denitrification process, i.e. nitritation-denitritation, possible, e.g. in an SBR or in a chemostat. As there is no need for external carbon dosage to achieve around 89% nitrogen reduction in the nitritation-anammox process, it provides an interesting alternative to the nitritation-denitritation process. However, the very slow-growing anammox bacteria require long start-up periods, sufficient inoculum from other plants and extra knowledge for operators. The nitritation-anammox process can be configured in a one- or two-reactor system in floc-type suspended growth, granular or moving-bed biofilm systems. Today, the floc-based systems in SBRs are the most widely used nitritation-anammox systems in full-scale applications, possibly because most old sludge liquor treatment plants are SBRs. Furthermore, the floc-based system has the lowest electrical energy consumption and the start-up period can be very short because of the use of inoculation.

Key words - anammox; denitritation; full-scale; nitritation; sludge liquor

Introduction

Separate treatment of sludge liquor produced at municipal wastewater treatment plants (WWTP) when dewatering anaerobic digested sludge can be one solution for meeting higher load requirements or more stringent effluent standards regarding nitrogen removal. It was recognised early that physical/chemical methods for nitrogen removal are more expensive than the biological method of nitrification and subsequent denitrification (Siegrist, 1996). However, physical/chemical methods can be of interest if nitrogen or/and phosphorus are fixed and then used as a fertiliser.

Many sludge liquor treatment plants were built when upgrading WWTPs because of small investment cost brought about by small volume requirements. Other reasons could be to reduce costs for external carbon source dosage and possibly reduce aeration energy costs and the carbon footprint. Furthermore, a sludge liquor treatment plant is normally designed for removed load

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rather than good effluent quality since the effluent usually is led to the main WWTP.

This study is a review of literature pertaining to different biological sludge liquor treatment methods used for nitrogen removal in full-scale applications. The review will describe and compare different methods according to configuration, reduction rate, start-up, operation stability, energy consumption, costs and nitrous oxide emissions. Only methods for separate treatment of sludge liquor will be discussed, i.e. bioaugmentation methods (Parker & Wanner, 2007) will not be described. The study will also present the current status in Sweden regarding full-scale applications of sludge liquor treatment at municipal WWTPs.

Sludge liquor composition

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The sludge liquor normally contributes 15-20% to the total nitrogen load as a result of released assimilated ni-

trogen, but only 1% of the total wastewater flow at a municipal WWTP. The breakdown of proteins during digestion of the sludge produced at the WWTP dissolves ammonium ions (NH_4^+) , with bicarbonate (HCO_3^-) as counter-ion. Therefore, the molar ratio between bicarbonate and ammonium is generally observed to be around 1. Co-digestion with external substrates often increases the nitrogen load, but substrates very rich in carbohydrates or fats can reduce the ammonium content by increased biomass production in the digesters. The ammonium concentration in the sludge liquor is often between 500-1,500 mg NH4+-N/L, e.g. high sludge thickening prior to digestion increases the ammonium concentration. The sludge liquor is also characterised by high temperature (25-35°) because of digestion under mesophilic or thermophilic conditions. The digested sludge is normally not cooled down by heat exchangers to a temperature lower than the mesophilic temperature range, even if sludge cooling can prevent post-digestion, thereby reducing undesirable methane emissions and odour. A temperature decrease is also caused by dewatering, e.g. in centrifuges, or by storage of sludge liquor. Finally, the sludge liquor is characterised by low COD:N ratios (<1.0) because of degradation of COD and dissolution of ammonium in the digesters. Van Loosdrecht & Salem (2006) remarked that if the sludge liquor from thermal sludge drying is also led to the sludge liquor treatment plant, the counter-ion of ammonium can be acetate instead of bicarbonate.

Processes in nitrogen removal of sludge liquor

The composition and characteristics of the sludge liquor favour high growth rates, allowing low solid retention times (SRT) to retain slow-growing lithoautotrophic bacteria such as aerobic ammonia-oxidising bacteria (AOB), nitrite-oxidising bacteria (NOB), and anaerobic AOB (anammox) bacteria. Fast heterotrophic denitrification is possible if an external carbon source is added. Figure 1 shows the most important species in the nitrogen cycle.

Nitritation-denitritation

The high temperature favours NOB wash-out because aerobic AOB grow faster than NOB at temperatures above 20°C (Hellinga *et al.*, 1998). Ammonium oxidation to nitrite (i.e. nitritation) with subsequent reduction into dinitrogen gas (i.e. denitritation) theoretically saves up to 25% of the oxygen demand and up to 40% of the carbon source. Also, the sludge production decreases by 30% and the CO_2 emissions by 20% compared to a conventional nitrification-denitrification



Figure 1. The most important nitrogen conversion processes at wastewater treatment plants with the exception of nitrogen fixation (1). (2) is degradation of organic material, e.g. anaerobic digestion, (3) assimilation, (4+5) aerobic ammonium oxidation or nitritation, (6) nitrite oxidation, (7–10) denitrification while (8–10) is denitritation and (11) is anaerobic ammonium oxidation. Modified figure from Gustavsson (2011).

process. Furthermore, ammonia and free nitrous acid (FNA) inhibition (Anthonisen *et al.*, 1976), hydroxylamine inhibition (Yang and Alleman, 1992), low dissolved oxygen (DO) concentration (Hanaki *et al.*, 1990) due to the NOBs' higher oxygen half saturation concentration, and intermittent aeration (Turk and Mavinic, 1989) suppresses NOB and favours nitrite accumulation.

Anammox

It is not only a nitritation-denitritation process that requires nitrite accumulation. The anammox bacteria oxidise ammonium with nitrite as electron acceptor (Strous *et al.*, 1998) according to an overall stoichiometry showing low biomass yield

NH₄⁺ + 1.32 NO₂⁻ + 0.066 HCO₃⁻ + 0.13 H⁺ → 1.02 N₂ + 0.26 NO₃⁻ + 0.066 CH₂O_{0.5}N_{0.15} + 2.03 H₂O.

The main product is dinitrogen gas, but 11% is converted to nitrate, which means that no more than 89% nitrogen removal is theoretically possible. Since only slightly more than half of the ammonium load has to be oxidised by nitritation into nitrite, the alkalinity in the sludge liquor is generally enough to attain the NH₄⁺:

 NO_2^- molar ratio needed. Compared to conventional nitrification-denitrification, the oxygen requirement in the nitritation-anammox process is 57% less and the carbon source needed through denitrification to fulfil 100% nitrogen removal is 86% less.

The main issue concerning the anammox bacteria is their slow growth rate. According to Strous et al. (1998), their estimated doubling time is 11 days at 32-33°C. Start-up of such a process takes several months and inoculation of sludge is preferred. The difference in growth rates between nitrifiers and anammox bacteria is significant. Typical maximum growth rates are at least ten times higher for the aerobic AOB and NOB (Sin et al., 2008). The dilemma in separating the nitrifiers and the anammox bacteria because of their difference in required SRT is described in Wett et al. (2010). In biofilm systems, e.g. moving-bed biofilm reactors (MBBRs), and granular systems, e.g. gas-lift or upflow reactors (van Dongen et al., 2001), there is a kind of natural SRT selection in a nitritation-anammox process because of stratification of the biofilm structure. The nitrifiers prefer the outer layers which are well supplied with oxygen and the anammox bacteria prefer the inner layers depending on the oxygen diffusion. Higher shear stresses and more erosion of the outer layer than the inner layer tend to give the anammox bacteria longer SRT (Wett et al., 2010).

In suspended growth systems, i.e. SBRs, the anammox bacteria form small granules with higher density in the sludge flocs (Wett et al., 2006). The anammox bacteria seem to have a natural ability to form granules (Trigo et al., 2006). Therefore, centrifugal forces can also be used to select the appropriate SRT for the different bacteria groups (Wett et al., 2010). It is important to separate the nitrifiers, or more precisely the NOB, because they compete with the anammox bacteria for the substrate nitrite. However, the NOB is also suppressed by other mechanisms already mentioned (e.g. low DO concentration and ammonia inhibition). Heterotrophic denitrifiers can compete with the anammox bacteria for nitrite but the availability of easily biodegradable COD in sludge liquor treatment is very limited, especially in anoxic micro-environments. However, a long SRT for the nitrifiers increases decay that can lead to available COD. When denitrifers are present nitrogen removal can be increased by denitrification of the residual nitrate, if the anammox bacteria do not oxidise the denitrification intermediate nitrite into nitrate again.

The anammox bacteria are also very sensitive to exposure of oxygen and too high nitrite concentrations (Strous *et al.*, 1999). The toxic nitrite level depends on size of biomass aggregates and acclimation periods (Wett *et al.*, 2010). In addition, methanol inactivates the anammox bacteria (Güven *et al.*, 2005).

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Nitrogen oxide emissions

Nitrogen removal in wastewater treatment plants is a distinct source of the prominent greenhouse gas nitrous oxide (N2O) (IPCC, 2006). However, reported emission values vary greatly (Kampschreur et al., 2009b) and few studies of measurement of nitrous oxide emission from full-scale sludge liquor treatment plants can be found. Both nitrifying and denitrifying bacteria can produce nitrous oxide as an intermediate product or as side- or end-products (Kampschreur et al., 2009b), while the anammox bacteria do not emit nitrous oxide (Kartal et al., 2010). Nitrous oxide may also be produced biologically or chemically by nitrite reduction to nitric oxide (NO) and nitrous oxide coupled to ferrous iron oxidation (Kampschreur et al., 2010). Generally, low DO concentrations, nitrite and rapidly changing conditions are the most common factors for nitrous oxide production in wastewater treatment plants (Kampschreur et al., 2009b). In addition to sludge liquor treatment often involving the conditions previously mentioned, the environment created is an extreme environment different to that of the main WWTP, e.g. with high reaction rates due to high temperature, high concentration of nitrogen species and different pH conditions. The savings in CO₂ emissions by decreasing the energy and external carbon dosage in the nitritationdenitritation process compared to a nitrification-denitrification process may be useless if the nitrous oxide emission is too high.

Process configurations Nitritation-denitritation

SHARON

The environment for nitrite accumulation and inhibition or wash out of NOB is good in sludge liquor treatment plants. The easiest way is to make use of the difference in growth rates between aerobic AOB and NOB as in the patented method SHARON®, Single reactor system for High activity Ammonia Removal Over Nitrite (Hellinga et al., 1998) (marketed by Grontmij). The SHARON process is a chemostat, which means that the reactor is operated without biomass retention, i.e. the hydraulic retention time (HRT) equals the SRT. Since effluent concentration is independent of influent concentration, the removal efficiency increases with higher influent concentrations. The HRT is chosen to prevent NOB growth, but also to be as short as possible without washing out the AOB, to minimise the tank volume. Laboratory studies showed that total nitrite accumulation was obtained at an aerobic HRT of 1 d at 35°C (Hellinga et al., 1998). However, in full-scale, the aerobic HRT should be between 1.3-1.8 d according to Mulder et al. (2006). The required anoxic HRT is recommended to be 0.5-0.75 d depending on type of carbon source. The volumetric nitrogen removal rates can be calculated to $0.2-0.8 \text{ kg N/(m^{3*}d)}$ with influent concentrations between 500-1,500 mg NH4+-N/L. A SHARON system can also consist of a two-tank system (one for nitritation and one for denitritation with possibilities for recirculation), which is more expensive to build, but the investment in aeration equipment will be less as the aeration can be continuous (van Loosdrecht, 2008). The first full-scale SHARON was installed 1997 in Utrecht in the Netherlands and there are now ten SHARONs in operation around the world, with two under construction (A. Schabbauer, personal communication).

Sequencing Batch Reactor (SBR)

The most common configuration for full-scale sludge liquor treatment plants is sequencing batch reactors (SBRs) with floc-type suspended biomass (Jardin *et al.*, 2006). Most of the plants have been operated according to the nitrification-denitrification process. However, several of these plants observed nitrite accumulation during periods and applied the nitritation-denitritation process (Fux *et al.*, 2006; Gustavsson *et al.*, 2010) even before the SHARON concept was introduced. Consequently, several full-scale nitritation-denitritation applications in the world are operated as SBRs.

An SBR cycle consists typically of fill and reaction phases, a settling phase and a withdrawal phase. Normally the cycle starts with a rapid filling phase, but continuous feeding has been observed to increase process stability and accelerate reaction rates (Fux et al., 2006). Filling into the sludge blanket during the settling and the withdrawal is also an option to avoid the need for an equalisation tank for the influent sludge liquor. An equalisation tank for the effluent might be required to balance the load to the main plant of the sludge liquor, which otherwise can affect the removal efficiency in the main plant. A nitritation-denitritation process in an SBR is normally not gained by washing out NOB by low SRT. The SRT is often very high, resulting in significant decay of bacteria, which increases the internal carbon source for the denitritation. In addition, the SRT is not easy to determine in an SBR due to the varying contribution of suspended solids from the sludge liquor. Consequently, the reaction rates are shown as volumetric rates rather than rates per SS or VSS in the literature. Nitrite accumulation in a nitritation-denitritation process thrives with high ammonia and FNA concentrations, low DO, hydroxylamine inhibition and intermittent aeration, but which of these factors is the most important one in a full-scale reactor is often un-known.

The required size of an SBR depends on the nitrogen load, reaction rates and settling properties. The nitritation rates are normally around 1.2-1.4 kg N/(m^{3*}d) (Wett et al., 1998; Fux et al., 2003, Gustavsson et al., 2010) and are especially affected by the DO concentration (Gustavsson et al., 2008) and nitrous acid concentration and availability of inorganic carbon source (Vadivelu et al., 2007). The denitritation rates vary even more depending on factors such as type of carbon source, carbon limitation, FNA and/or nitric oxide concentrations (Fux et al., 2006; Gustavsson et al., 2010; Yuan and Pijuan, 2009) with normal full-scale rates between 1.4-2.2 kg N/(m³*d) (Fux et al., 2003; Gustavsson et al., 2010). The settling properties are often very good in an SBR. The necessary HRT for settling and withdrawal shown by Gustavsson et al. (2010) was only 0.16 days, even if the settling was not subjected to any optimisation. The volumetric nitrogen removal rates are calculated to be between $0.5-0.8 \text{ kg/(m^{3*}d)}$.

Gustavsson et al. (2010) tried to optimise the alkalinity production needed in an SBR nitritation-denitritation system, which resulted in inhibition of the denitrifiers because of recurring periods with high nitrite concentration and simultaneous low pH, i.e. high FNA concentrations. The inhibition mechanism was not found but the same pattern was found in Fux et al. (2006), where accumulation of nitric oxide, an intermediate in the denitritation process, reduced the denitritation rates. The risk of nitric oxide accumulation increases in acidic conditions (Murray & Knowles, 2001) at high nitrite concentration due to direct inhibition of nitric oxide reductase by nitrite (von Schulthess et al., 1995) and in intermittent aeration conditions due to sequential induction of enzymes in anoxic conditions (Casey et al., 1999). However, it is not known whether nitric oxide or FNA (Yuan and Pijuan, 2009) acts as main inhibitor. Furthermore, shortage of carbon source for the denitrifiers leads to non-complete denitrification and an accumulation of undesirable intermediates, i.e. nitric oxide and nitrous oxide (Casey et al., 1999).

SBR vs. SHARON

Figure 2 compares the HRT needed for a 95% reduction of ammonium in the effluent with a SHARON and an SBR configuration. The design and possible HRT in a SHARON are shown to be of great importance, as well as the influent ammonium concentration. The reactor size of a recommended SHARON design has to be almost twice as big as the SBR reactor when comparing to the full-scale experiences at Bern WWTP with the influent concentration of 1000 mg NH₄⁺-N/L.

The normally smaller reactor volume shown in Figure

Figure 2. Comparison of HRT requirements according to SHARON laboratory studies (1.0 aerobic HRT and 0.5 d anoxic HRT) (Hellinga et al., 1998), fullscale design value for SHARON (1.5 d aerobic HRT and 0.75 d anoxic HRT) (Mulder et al., 2006) and for full-scale SBRs at Sjölunda WWTP (Gustavsson et al., 2010) and at Bern WWTP (Fux et al., 2003). The flow is set to be 700 m³/d and the nitrogen removal is assumed to be 95 %.



1 favours the choice of an SBR configuration since the other economic aspects are shown to be almost the same (Fux et al., 2003). The SHARON is said to be simpler in operation by simply controlling the HRT, while an SBR is normally dependent on other parameters (i.e. low DO concentration) to maintain nitrogen removal over nitrite. The SHARON reactor does not accumulate undesirable suspended solids in the sludge liquor unlike in an SBR, which decreases the aeration requirements in the sludge liquor treatment plant. The internal hydrolysis of these solids and the long SRT in an SBR decreases the amount of external carbon source needed. The stochiometric value of the required COD:Neliminated mass ratio is 2.86 for denitrification and 1.72 for denitritation. Including sludge production, the ratios are about 4 and 2.4 respectively (Mulder et al., 2006) depending on carbon source. In the full-scale SHARON applications in the Netherlands, the COD:N ratios are below 2.4 g COD_{dosed}/g NO₂⁻-N_{eliminated} (Mulder et al., 2006), which should be good evidence of a denitrification of mostly nitrite, i.e. denitritation. Gustavsson et al. (2010) and Fux et al. (2003) operated SBRs with COD consumption at 2.0 g COD_{dosed}/g $N_{eliminated}$ and 2.2 g COD_{dosed}/g N_{eliminated} respectively. Lower ratios than those theoretically required can be caused by internal hydrolysis, but too low ratios increase the risk of inhibition by nitric oxide accumulation and nitrous oxide emissions.

Furthermore, the SHARON is more sensitive to inhibition as a wash out occurs very fast, while the sludge is retained in an SBR. An SBR can be started up within three weeks, by inoculation of activated sludge from the main plant and with the strategy of reaching normal working temperature quickly without inhibiting the ammonia oxidisers. This is achieved by avoiding too high ammonia concentrations and by continuous aeration at high DO to avoid oxygen limitation (Gustavsson

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et al., 2007 & 2010). The start-up strategy and time should be approximately the same for the SHARON process, but van Kempen *et al.*, (2001) reported a full-scale start-up period of seven weeks for a SHARON process. The required instruments for controlling the two process options, i.e. pH and DO meters, are the same. The length of the anoxic phase can be favourably controlled by online measurement of the oxidation-reduction potential (ORP) (Gustavsson *et al.*, 2010). If lower ammonium oxidation than the possible one is desirable monitoring the ammonium concentration online can help in an SBR. It has been observed that the conductivity correlates very well with the ammonium concentration (Levlin & Hultman, 2008).

Problems with foam have been reported in SBRs and SHARONs (Fux *et al.*, 2003; Mulder *et al.*, 2006; Gustavsson *et al.*, 2010). Reactors have been equipped with sprinklers (Fux *et al.*, 2003), but anti-foam agents have also been used. However, these decrease the oxygen transfer efficiency (Gustavsson *et al.*, 2010).

Nitritation alone as an option

Complete oxidation of the ammonium in the sludge liquor treatment plant is not always required for fulfilling the effluent requirements for the main plant depending on the reason for separate treatment of the sludge liquor stream. The need for separate treatment can also vary during the year. Sufficient nitrification capacity in the main plant during parts of the year may be a reason to reduce the oxidation in the sludge liquor plant to avoid external carbon source addition. Often denitritation is only required because of the need for alkalinity addition rather than because of denitrification capacity problems in the main plant. Addition of alkalinity, in the form of sodium hydroxide, is more expensive than addition of an external carbon source (Gustavsson *et al.*, 2008). Since nitritation produces two moles of H^+ and the sludge liquor often contains one mole alkalinity per ammonium ion, almost one mole extra alkalinity is needed to promote a pH reduction. A pH below around 6.5 suppresses the AOB (van Kempen *et al.*, 2001) by low substrate availability (bicarbonate and ammonia) or by the pH itself. The required alkalinity can be retained by denitritation since it produces one mole alkalinity when denitrifying one mole nitrite. However, if only half of the ammonium content in the sludge liquor is to be oxidised, then no alkalinity addition is needed if it is assumed that there is no lack of denitrification capacity in the main plant. Therefore, one possible process chosen could be nitritation alone, with 50% ammonium oxidation.

Even if denitritation is economically preferable to direct alkalinity dosage, the operation with nitritation alone is simple and more flexible than a nitritation-denitritation process (Gustavsson *et al.*, in preparation) and is practised in full-scale at Sjölunda WWTP in Sweden (Gustavsson *et al.*, 2008).

Nitrous oxide emissions

Only three studies of nitrous oxide emissions in nitrification-denitrification systems in sludge liquor treatment at municipal WWTP have been found in literature. Björlenius (1994) observed that 33% of the eliminated nitrogen in an SBR with a nitrification-denitrification process working with DO concentrations just below 1.0 mg O_2/L was emitted as nitrous oxide. These emissions corresponded to an increase in the total nitrous oxide emission from the WWTP of 1.5-4.5%. Kampschreur et al. (2009) studied a chemostat nitritation reactor with around 50% ammonium oxidation and found that 1.7% of the nitrogen load was emitted as nitrous oxide. Gustavsson & la Cour Jansen (2010) measured the nitrous oxide emissions in an SBR with nitritation alone (90% ammonium oxidation received by sodium hydroxide dosing) to be 3.8 % of the ammonium nitrogen load. No full-scale studies of nitritation-denitritation plants have been found. Björlenius (1994) shows the potential for extremely high emissions.

Nitritation-anammox

The nitritation-denitritation process is mostly performed on the basis of suspended activated sludge. On the other hand, the nitritation-anammox (also called deammonification) process is also implemented as biofilm and granular sludge systems in full-scale. The process can be accomplished in a one- or two-reactor system.

Full-scale plants

The first full-scale nitritation-anammox plant was a onereactor biofilm system, a cascaded MBBR (40% filling with Kaldnes carriers K1 (effective area $500 \text{ m}^2/\text{m}^3$)), called DeAmmon[®] (marketed by Purac), started in Hattingen WWTP (53,000 population equivalents [P.E.]) in Germany early in 2001 (Rosenwinkel *et al.*, 2005). The plant was operated with nitritation-denitritation for the first six months, before switching operation to a nitritation-anammox process (the reactor was originally designed for a nitritation-denitritation process). One and a half year later the nitrogen removal reached 70-80% at a load between 100-160 kg N/d. The original design was based on 120 kg N/d, 200 m^3 sludge liquor/d and 80% reduction.

In 2007, a second DeAmmon plant (design load 670 kg N/d, 32 % filling) was started at Himmerfjärden WWTP (260,000 P.E.) in Sweden (Ling, 2009). Two existing pre-sedimentation tanks were converted into MBBR tanks. The experiences in Hattingen led to a process based on intermittent aeration. The start-up times for the two lines at Himmerfjärden were 9 and 12 months, but Ling (2009) noted that the effective start-up time was 6–7 months (excluding problems during start-up). Today, there are three DeAmmon plants in operation. The new one, in Dalian, China, is in start-up phase (design load is 2200 kg N/d) (L. Kanders, personal communication).

In 2002, the first full-scale anammox reactor in the world was started in Rotterdam (Dokhaven WWTP, 620,400 P.E.) in the Netherlands (van der Star et al., 2007). At the WWTP, a SHARON reactor was already installed (Mulder et al., 2001) and this reactor is used as a nitritation reactor with chemostat-mode with a controlled aerobic HRT of 1.4 days (Kampschreur et al., 2008). The partly nitritated effluent from the nitritation reactor passes a tilted plate settler before reaching the anammox reactor. The anammox reactor is a gas lift-reactor, with granular sludge, where the biomass retention is obtained by a three-phase separator. Lemaire et al. (2008) defines granules as compact and dense aggregates of microbial origin with an approximately spherical external appearance that do not coagulate under reduced hydrodynamic shear and settle significantly faster than conventional activated sludge flocs. The two-reactor concept is called SHARON®-ANAMMOX® (van Dongen et al., 2001). Since the nitritation-anammox concept in Rotterdam was scaled-up directly from laboratory-scale to full-scale, based on van Dongen et al. (2001), and because of various problems (van der Star et al., 2007), the design load of 500 kg/d was not reached until 3.7 years. The full-scale SHARON-ANAMMOX process was developed through cooperation between the companies Paques and Grontmij together with Delft University of Technology.

The next development by Paques was a one-reactor ANAMMOX[®]. The anammox granules from Rotterdam were used as inoculum to start-up a granular onestage nitritation-anammox plant treating sludge liquor from a municipal WWTP in Olburgen in the Netherlands and pre-treated wastewater from a potato processing plant with a capacity of converting 1200 kg N/d (Abma et al., 2007 & 2010). This time, the start-up period was around 5 months reaching the actual load of 700 kg N/d (Abma et al., 2007). The reactor is a chemostat of 600 m³ and the granules are retained by a separator on the top of the reactor (Abma et al., 2010). Thorough mixing is ensured by several riser pipes in the reactor (Kampschreur et al., 2009a). The reactor is operated with continuous aeration. The aeration flow is adjusted in response to online measurements of the ammonium and nitrite concentrations in the effluent to attain the desired treatment results.

Paques has installed a total of eight full-scale nitritation-anammox plants in the world, of which three are placed at WWTPs.

The first full-scale one-reactor system for nitritationanammox with floc-type suspended biomass in an SBR was started at Strass WWTP (200,000 P.E.) in Austria in 2004 (Wett, 2006). However, the actual start-up of the full-scale reactor (500 m³) of six months was preceded by an enrichment period of two years, starting with four litres inoculum from a pilot plant operated by EAWAG in Zürich to a 300 l reactor gradually increased to a volume of 2.4 m³. The process is operated with low DO (0.3 mg/l) and intermittent aeration. The aeration is pH-controlled and is activated at the upper pH set-point within a very tight pH interval of 0.01. During aeration, nitritation soon reduces the pH, while during anaerobic conditions the nitrite is consumed by the anammox bacteria. The anammox reaction produces some alkalinity but the pH increases mostly because of the continuous filling of sludge liquor. The low DO set-point is chosen in order to prevent rapid nitrite accumulation and to maintain a continuous repression of nitrite oxidation by NOB. The importance of a narrow pH interval and low DO set-point to minimise nitrite accumulation that will inhibit both aerobic AOB and anammox bacteria was further investigated by Wett et al. (2007). This pH-controlled deammonification system is called DEMON® (Wett, 2007) and is marketed by the companies Grontmij and Cyklar-Stulz. The DEMON process also includes a patented hydrocyclone which is fed with the waste activated sludge from the SBR (Wett et al., 2010). The centrifugal forces in the cyclone divide the sludge into two fractions and help the system select the appropriate SRT for the two desirable bacteria groups. The overflow,

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which includes brownish flocs with, for example, the aerobic AOB (and other fast-growing bacteria), is wasted, and the underflow, which includes most of the red anammox granules, is recycled to the SBR. Model simulations in Wett *et al.* (2010) showed that the SRT was six times greater for the anammox bacteria compared to the AOB with the use of the cyclone.

Today, a total of eleven DEMON-plants are in fullscale operation (all in Europe) but seven more are under construction (A. Schabbauer, personal communication). The capacity of the plants varies between 50–2400 kg N/d. Most of the plants are located at WWTP.

There are also other floc-type suspended growth systems in SBRs in full-scale (five) with a one-reactor nitritation-anammox process that is not pH-controlled (Joss et al., 2009). At Zürich WWTP, there are two SBRs working parallel treating sludge liquor with nitritationanammox, each with a target load of 625 kg N/d. The first of the reactors was started by an inoculum from an 8 m³ pilot plant in October 2007 and the start-up time was reported to be 180 days, while the next reactor received its sludge from the first reactor and was loaded with the targeted load directly in June 2008. The DO set-point is below 1 mg O_2/L and the reaction phase is stopped at an ammonium stop-value. The operation of parallel SBRs in Zürich has been a great opportunity to study different operation modes in parallel reactors. Comparisons between continuous and intermittent aeration during the reaction phase shows that except for more accurate monitoring of the reactor performance by several online signals at unchanged aeration regimes and less on/off switching of the aerators, higher nitrogen reduction rates could be achieved by continuous aeration. In addition, the emissions of nitrous oxide were lower, 0.4% of the removed nitrogen load for continuous aeration and 0.6% for intermittent aeration (Joss et al, 2009).

Comparison between the nitritation-anammox configurations

One-reactor or two-reactor system?

In a two-reactor system the nitritation and anammox processes are physically separated, in offering a wider range of optimal process conditions than in a onereactor system (Veys *et al.*, 2010). This allows flexibility, but it is important to have a stable composition of the nitritated effluent to avoid nitrite intoxication of the anammox bacteria in the second reactor. In addition, the very high nitrite concentrations (several hundred mg NO₂⁻-N/L) in the influent to the anammox reactor must be continuously removed to avoid toxic concentrations and thorough mixing is required. One option in the case of too high nitrite concentrations is always to partly by-pass the anammox reactor or to dilute by recirculating the anammox effluent. Joss *et al.* (2009) also pointed out that they measured lower nitrous oxide emissions in a one-reactor system than those measured by Kampschreur *et al.* (2008) in a two-reactor system in Rotterdam, probably because of higher nitrogen species concentration during nitritation. Furthermore, Joss *et al.* (2009) stated that a one-reactor system is considerable simpler to operate due to less need for control.

Start-up

The slow growth rate of the anammox bacteria results in long start-up periods if no inoculum is provided. The effective start-up of the biofilm process DeAmmon at Himmerfjärden WWTP was 6–7 months (Ling, 2009). Rosenwinkel et al. (2005) reported that the anammox bacteria do not seem to be able to form a biofilm structure and need an existing biofilm in which to enrich. Consequently, aerobic operation with high substrate load, i.e. pre-settled wastewater (Ling, 2009) will provide a biofilm on the carriers. A start-up with a nitritation-denitritation process has also been tested (Rosenwinkel et al., 2005; Ling, 2009), but methanol should be avoided because of its toxicity on anammox bacteria. Later, a gradually increased load of sludge liquor with adequate aeration seems to be the concept for start-up. Start-up of granular and suspended growth systems has taken several years without sufficient inoculum (van der Star et al., 2007; Wett, 2006). However, today there are several full-scale plants providing large amounts of seeding sludge that, naturally dependent on design load, minimise the start-up periods to a few weeks down to immediate required capacity. A requirement for this is that there is sufficient inoculum available on the market. One solution could be to have certain inoculum plants at large WWTPs where reduced capacity of the nitrogen removal in the sludge liquor treatment plant does not affect the treatment results for the main WWTP. Inoculation with carriers with an already existing biofilm containing anammox bacteria during start-up of an MBBR has not been reported in full-scale, but the necessity of seeding sludge in an MBBR system has been questioned in literature (Schneider et al., 2009). However, very recently Lemaire et al. (2010) reported that seeding with colonised carriers with pre-existing deammonification biofilm in lab-scale MBBRs was successful, and reduced the start-up time. A start-up should favour the anammox bacteria with low nitrite and DO concentrations, which also suppress NOB. Since the aeration required during start-up is lower than the blower capacity needed later, intermittent aeration is usually applied (Joss et al., 2009). It is also important to avoid ammonia inhibition of the ammonia oxidisers which can lead to higher DO concentrations in the reactor. Recirculation of the effluent decreases the concentration of the nitrogen species.

Installing facilities to heat the reactor will decrease the start-up period but is very costly. Microbial characterisation using, for example FISH analysis, is normally performed in order to monitor the anammox growth (Ling, 2009). In biofilm reactors, study of the dry solids and the volatile fraction is recommended (Ling, 2009).

Reduction capacities

Van der Star et al. (2007) estimated the theoretical maximum volumetric conversion rates of different reactor types since data from full-scale applications do not show the potential conversion rates because they are often substrate-limited. The limiting process for maximum conversion rate was the oxygen transfer for the airlift and the SBR processes and the oxygen penetration for the MBBR process. The volumetric conversion rates were much higher for the airlift and the SBR (8 kg $N/(m^{3*}d)$ – both reactors) than for the MBBR (1.2 kg $N/m^3/d$ or 5 g $N/m^2/d$). In a granular reactor with only the anammox process, the volumetric conversion was estimated at 12 kg N/(m³*d), limited by the hydrodynamics. The effective surface area in an MBBR is 200 m^2/m^3 , with a filling degree of 40% with the Kaldnes carrier K1 (500 m²/m³). The volumetric conversion rates can be increased with "chip" type carriers due to their greater specific protected surface area (Lemaire et al., 2010). In a granular sludge reactor the effective surface area is at least 3,000 m²/m³ (Abma et al., 2007). The total volumetric conversion rate in a SHARON-Anammox depends mostly on the ammonium concentration, 500-1,500 mg/l gives reaction rates between 0.31-0.88 kg N/(m³*d) calculating with 89% nitrogen removal. A possible two-reactor system is an SBR-Anammox system, probably giving reaction rates above 1 kg N/(m³*d) (Wyffels et al., 2004). Reported full-scale volumetric rates are found in Table 1.

Operational stability

The most important factor for a stable nitritation-anammox process is control of aeration, e.g. to avoid overaeration, which inhibits the anammox bacteria because of high DO and high nitrite concentrations. Anammox in biofilms and granules are less susceptible to environmental stressors such as high DO and nitrite than in floc-type suspended growth systems. For example, nitrite concentrations up to 42 mg NO₂^{-/L} did not inhibit the anammox bacteria in a full-scale ANAMMOX reactor (Abma et al., 2010), while nitrite concentrations are kept well below 5 mg/L in the DEMON process to avoid inhibition (Wett, 2007). Insufficient aeration can lead to inhibition of the aerobic AOB by too high ammonia concentrations, which is the biggest problem experienced at Himmerfjärden WWTP (L. Kanders, personal communication).

Table 1. Reported full-scale volumetric rates for nitritationanammox systems.

| Process | Volumetric rate (kg N _{eliminated} /(m ^{3*} d)) |
|--|--|
| SHARON [®] -ANAMMOX [®] One-stage ANAMMOX [®] DeAmmon [®] DEMON [®] SBR | $\begin{array}{c} 0.6^{\rm a)} \\ 1.1^{\rm b)} \\ 0.3\text{-}0.4^{\rm c)} \\ 0.6^{\rm d)} \\ 0.5^{\rm c)} \end{array}$ |

^{a)} Only the aerobic HRT is included in the rate calculations (van der Star *et al.*, 2007; Kampschreur *et al.*, 2008).

^{b)} Abma *et al.* (2010).

^{c)} Rosenwinkel et al. (2005) and Ling (2009).

^{d)} Wett (2007).

e) Joss et al. (2009).

At Himmerfjärden WWTP the DeAmmon process has been subjected to calcium precipitation on the carrier material, which strongly decreased the removal capacity (S. Stridh, personal communication). The problem was probably caused by high pH due to malfunctional problems with a pH-meter (L. Kanders, personal communication). Rosenwinkel et al. (2005) pointed out that 28% of the initial running costs were spent on dosage of chemicals preventing this precipitation in Hattingen. However, it was also stated that the dosage of acid could be gradually reduced. The Himmerfjärden plant was not built with capability for acid dosage because of the high costs of adjusting pH (Ling, 2009) and because several years of pilot tests at Himmerfjärden did not experience this problem (Trela et al., 2006).

Biomass retention is critical for the slow-growing anammox bacteria. In SBR floc-type systems settling sometimes can be a problem. Joss *et al.* (2009) added a nanostructured flocculant during one week to improve the settling when the MLSS concentration was below 2 g/L. However, during regular operation (3.5–4 g MLSS/L) settling was good. In an MBBR process it is important to minimise the shear stress exerted by the mixing devices in order to build up a biofilm with good performance (Jardin *et al.*, 2006).

No major problems with NOB and/or heterotrophic growth have been reported. Abma *et al.* (2010) remarked that, in a granular system, the incoming solids and floctype biomass growth resulting from the incoming COD are washed out of the reactor. In an SBR system, accumulation of incoming SS could be a problem affecting the settling. The hydrocyclone in a DEMON reduces this problem. At Himmerfjärden WWTP the operation

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of the DeAmmon includes automatic by-pass if the incoming SS concentration (measured online) is >1,500 mg/L (S. Stridh, personal communication). In some installations, presettling of SS in lamellas or a sedimentation tank is also implemented (van der Star *et al.*, 2007; Ling, 2009). A small contribution of heterotrophic denitrification of the nitrate produced, without affecting the anammox bacteria, could be welcomed in order to increase the nitrogen reduction.

Monitoring

Several online instruments have been used at the various full-scale nitritation-anammox configurations. Measurement of DO concentration and pH is required in all systems to avoid over-aeration and ammonia inhibition. Online measurements of ammonium are used in most reactors, also for controlling the process, but the conductivity can be used instead (Levlin & Hultman, 2008) and measurement of the conductivity is cheaper and implemented at Himmerfjärden (S. Stridh, personal communication). Some installations also include online measurement of nitrite (van der Star et al., 2007; Abma et al., 2010). The temperature is measured but normally not adjusted. Covered reactors are preferable in order to minimise heat losses. The air flow and the electrical energy consumption are also measured for the separate sludge liquor treatment plant in order to calculate the actual savings of the separate treatment. The SS content can be measured online in order to by-pass in high SS content situations. In addition to online measurements, supplementary flow-proportional or grab samples for laboratory analyses are also normally taken to monitor the reactor performance. Monitoring the biomass cultivated by measuring the volatile SS content and by microscopic analyses are also reliable indicators for efficient performance of a nitritation-anammox plant (Ling et al., 2009).

Costs

Investments costs are very site-specific, because existing reactors and machinery of the WWTP can sometimes be used (Jardin *et al.*, 2006). The most compact solution is a granular system, because of the greater surface area available. An MBBR process includes costs of carriers, which are probably greater than the costs for a settling system for granules (Abma *et al.*, 2007).

Electrical energy consumption at the DEMON in Strass WWTP was reported to be 1.16 kWh/kg $N_{eliminated}$ (Wett, 2007) and the SBRs in Zürich showed similar figures, 1.0 kWh/kg $N_{eliminated}$ (Joss *et al.*, 2009). The designed electric energy consumption for the DeAmmon in Himmerfjärden WWTP was 2.3 kWh/kg $N_{eliminated}$ (L. Kanders, personal communication), which is close to the consumption for an SBR with nitritationdenitritation at Sjölunda WWTP in Sweden, 2.9 kWh/ kg N_{eliminated} (Gustavsson et al., in prep.). It should be noted that the SBR at Sjölunda WWTP had problems with worn fine bubble membrane disc diffusers. The electrical energy consumption in the DeAmmon in Hattingen was reported to be as high as 5.6 kWh/kg Neliminated (Jardin et al., 2006) and was explained by low ammonium load (L. Kanders, personal communication). In conventional nitrification-denitrification in the main line of WWTPs, the electrical energy consumption is normally 3.5-5.7 kWh/Neliminated (Beier et al., 2008). Since the penetration of oxygen into the biofilm in MBBRs limits the reduction rates, higher DO concentration is needed in such systems. In floc-type suspended biomass systems, the DO set-point is below 0.5 mg O₂/L (Wett, 2006; Joss et al., 2009), while, for example, the MBBR at Himmerfjärden WWTP has a DO set-point between 3-4 mg O₂/L (J. Örnmark, personal communication) and large bubble air diffusers. No energy consumption values have been published in literature for granular systems, but Kampschreur et al. (2009a) showed that the DO concentration during regular operation was 5 mg O2/L in the one-stage ANAMMOX at Olburgen WWTP, which gives higher energy consumption than in floc-type suspended growth systems.

Personnel requirements were estimated to be 0.25 man/year for the DeAmmon in Hattingen (Jardin *et al.*, 2006). In all the marketing nitritation-anammox solutions some licence or royalties costs are required. There should also be certain agreements on support, particularly in the event of failure.

Nitrous oxide emissions

There are very few full-scale studies on nitrous oxide emissions from nitritation-anammox systems. As mentioned earlier, Joss et al. (2009) measured smaller emissions in a suspended growth SBR with continuous aeration (0.4% of the nitrogen load) than the two-reactor SHARON-ANAMMOX in Kampschreur et al. (2008) (2.3% of the nitrogen load). However, the two-reactor system in Kampschreur et al. (2008) emitted less nitrous oxide than the main WWTP in relation to the nitrogen load. Joss et al. (2009) found that continuous aeration is preferable, not only because of higher reduction rates, but also because the nitrous oxide emissions decreased (0.4% and 0.6% respectively). Consequently, a DE-MON plant, with its intermittent aeration, is expected to have higher nitrous oxide emissions, which is the case for the plant in Strass (1.5% of the gaseous nitrogen turnover) according to Weissenbacher et al. (2010). Kampschreur et al. (2009b) measured emissions from a one-reactor granular system and found that 1.2% of the nitrogen load was emitted as nitrous oxide.

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A comparison between emissions of CO_2 equivalents from the nitritation-anammox process in an SBR (Joss *et al.*, 2009) with continuous aeration during reaction phase and the conventional nitrification/denitrification at Zürich WWTP revealed a decrease of 3.0 kg CO₂/kg N_{eliminated} with this specific sludge liquor treatment (Joss *et al.*, 2009). The comparison included aeration energy, external carbon dosage and 0.1% and 0.4% of nitrogen eliminated emitted as nitrous oxide in the main line and the sludge liquor treatment respectively. Almost the same decrease was obtained when no external carbon dosage was applied because of decreased biogas formation due to the use of organic substances in the wastewater for denitrification.

Foaming

Foam formation is observed in suspended growth systems, and sprinkling systems have therefore been installed (Wett, 2006; Joss *et al.*, 2009).

Carbon source or anammox?

Decision matrices for selection of process for sludge liquor treatment can be found in van Loosdrecht & Salem (2006) and van Loosdrecht (2008). However, it is important to point out that decision criteria can be very site-specific. Anyway, some rules of thumb have been suggested. Capacity problems in nitrification can be caused by too low aerobic SRT (select bioaugmentation) or lack of aeration capacity (select separate treatment). If the counter-ion to ammonium is acetate, select the nitritation-denitritation process, and if it is bicarbonate select nitritation-anammox. Capacity problems in denitrification can be caused by lack of carbon source (select nitritation-anammox) or space for denitrification (select bioaugmentation).

First of all, the choice of separate treatment process, including denitrifers or anammox bacteria, depends on the level of available carbon source for denitrification in the sludge liquor. Heterotrophic activity outcompetes the anammox bacteria and the amount of added external carbon source may be minimal. However, the COD:N ratio is normally very low in the sludge liquor. Consequently, a nitritation-anammox system is preferable due to lower electrical energy costs (i.e. in floc-type suspended growth systems), no need for external carbon addition, and lower nitrous oxide emissions. A nitritationanammox process in an MBBR may need chemicals for pH control in order to avoid precipitation. The construction cost is very much the same, except when choosing a two-reactor concept. Other aspects to consider, whether comparing the choice of carbon source addition or not, are the required reduction, start-up, operation stability and flexibility and need for knowledge.

The nitrogen removal capacity is theoretically lower in a nitritation-anammox process than a nitritation-denitritation process because of the nitrate production (< 89%). The actual removal in full-scale nitritation-anammox plants shown in literature differs between 70– 86%, with higher reduction in floc-type systems. The nitrogen removal in nitritation-denitritation systems is potentially very high and 90–95% removal is generally received. The design load to the main WWTP may, however, be able to accommodate the difference in reduction capacity since the nitrogen load from the sludge liquor treatment plant is normally only 15–20%. In addition, if the available carbon source in the influent of the main WWTP is sufficient, the remaining nitrate from the anammox process can be removed easily.

The process stability of the two different choices should be equal with sufficient supervision by operators. The difference is that complete inhibition of the anammox bacteria will have a much more severe effect than inhibition of the nitrifiers and the denitrifiers, because of the very long doubling time for the anammox bacteria. With an inoculum of nitrifying activated sludge a nitritation-denitritation process can be started within three weeks, while a nitritation-anammox process needs much longer or sufficient quantities of inoculum from another WWTP, currently from a very long-distance, in order to fulfil the same reduction as before the inhibition. Therefore, supervision of the nitritation-anammox system should be more extensive, perhaps always including online measurements of ammonium and nitrite and more laboratory analyses. The nitritation-denitritation system could be said to be more flexible since the process can be shut down and started up in matter of weeks without involving any external WWTP or company.

Knowledge about the anammox bacteria also has to be incorporated at the WWTP, while nitritation and denitritation already occurs in the main lines of the WWTPs. However, the total number of personnel required at the WWTP should be the same in the both cases. Since the sludge production is much higher in a nitritation-denitrition system because of the heterotrophic growth of the denitrifiers, more nutrients (e.g. P) and trace elements (e.g. Cu) are needed than in a totally autotrophic grown system, i.e. the nitritation-anammox process. At Nykvarn WWTP in Linköping, Sweden, preliminary results show that P- and Cu-deficiencies are the case and these elements are added in a newly started SHARON process (H. Tengliden, personal communication). Heat production is significant when heterotrophic growth is present, while the contribution of heat from the totally autotrophic process is low. Therefore, coverage of nitritation-anammox plants is recommended to minimise the required volume, which makes supervision more difficult.

Status in Sweden

In Sweden, a total of 23 municipal WWTPs have sludge liquor treatment (Figure 3a). The most common configuration is SBR followed by bioaugmentation plants. The bioaugmentation plants are ScanDeNi[®] processes (Rosén & Huijbregsen, 2003) or modified ones where the sludge liquor is treated mixed with the return activated sludge from the main line. Separate sludge liquor treatment plants are implemented at 10 WWTP (Figure 3b). The most dominant process choice is nitrification-denitrification, but nitritation-denitritation processes are found (one SHARON at Nykvarn WWTP, Linköping and one SBR at Sundet WWTP, Växjö), as well as nitritation-anammox (DeAmmon at Himmerfjärden WWTP, Södertälje) and nitritation (SBR at Sjölunda WWTP, Malmö). It is noteworthy that the ammonia stripping plant for sludge liquor treatment at Eslöv WWTP (Thorndahl, 1993) is not in operation and a ScanDeNi process is now installed. In 2007, a network for process engineers and operators at wastewater treatment plants with sludge liquor treatment in Sweden and Denmark was initiated for exchange of experiences.



Figure 3. a) Configurations of sludge liquor treatment plants in Sweden. b) Processes in separate sludge liquor treatment plants in Sweden.

Conclusion

Today, several different full-scale sludge liquor treatment plants are in operation around the world. The nitritation-denitritation process is performed with activated sludge in SBRs or in chemostats. The chemostat often requires a larger volume than the SBR. The nitritationanammox full-scale applications consist of floc-type suspended growth, granular or moving-bed biofilm systems. The floc-based systems in SBRs dominate, probably because most old sludge liquor treatment plants are SBRs, the system has the lowest electrical energy consumption and the start-up period is very short because of inoculation. Surprisingly, the literature about nitritation-denitritation and nitritation-anammox systems still lacks sufficient operational experiences and data from full-scale plants.

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