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A review on mainstream deammonification of municipal wastewater: Novel dual step process

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Abstract

The conventional biological nitrogen removal process is receiving increasing pressure partially due to its energy-negative operation. To address this challenge, various mainstream deammonification processes have been explored for energy-neutral municipal wastewater treatment, whereas these processes appear challenging to be sustainably and stably achieved in conventional process configurations. Therefore, this review aimed to provide a comprehensive analysis of the state-of-the-art of mainstream deammonification, while highlighting the major technical challenges. It appeared that recently developed novel dual step process, i.e. A-B processes, could provide a feasible engineering option for mainstream deammonification, where A-stage is designed for COD capture with the aim to enhance energy recovery, and B-stage is tailored for nutrient removal/recovery. This indeed may lead to a promising integrated mainstream deammonification process towards energy-efficient and environmentally sustainable nitrogen removal. Meanwhile, this review also offered an opinion on future municipal wastewater treatment, aiming for concurrent water reclamation and energy recovery.

Keyword: Novel dual step process; Mainstream anammox; COD capture; Energy recovery; Process configurations

1 Introduction

The conventional activated sludge (CAS) process and its varieties have been most commonly adopted for municipal wastewater reclamation. In this process with an anoxic-oxic configuration, organic matters in terms of chemical oxygen demand (COD) and nitrogen compounds are concurrently removed. Ammonia is oxidized to nitrate with oxygen as the electron acceptor, which is subsequently reduced to nitrogen gas with wastewater COD as the electron donor. This biological nitrogen removal (BNR) process has been well known as full nitrification-denitrification (Rittmann & McCarty, 2001). It should be noted that most of influent COD is removed through bio-oxidation with carbon dioxide as the end product in the oxic phase along with nitrification. In the treatment of municipal wastewater with relatively low COD concentration, this BNR process seriously suffers from intensive aeration-associated energy consumption for COD oxidation and nitrification, insufficient COD to sustain full denitrification, and production of waste sludge, etc.

In fact, it had been reported that the average in-plant energy consumption already reached 0.47 kWh/m³ (Drechsel et al., 2015; Zhou et al., 2013), of which about 40-75% was attributed to aeration (Henderson, 2002; Mamais et al., 2015), while nitrification could lead to an increase in the energy consumption by 0.1 kWh/m³ (Goldstein & Smith, 2002). The global municipal wastewater production already stood at 330 km³/year (Sato et al., 2013), thus the total electrical energy consumed for treating municipal wastewater could be estimated as $0.47 \text{ kWh/m}^3 \times 330 \text{ km}^3/\text{year} = 155$ billion kWh/year at the global scale. Since electrical energy is mainly derived from the combustion of fossil fuels, high energy consumption in present municipal wastewater treatment plants (WWTPs) could be inevitably linked to increasing greenhouse gases (GHG) emission which has been believed to be responsible for

global climate change. On the other hand, it should also be realized that 0.86 g of volatile suspended solids (VSS) is produced in removing 1 gram of ammonium-N via nitrification-denitrification with acetate as carbon source (Ahn, 2006; Rittmann & McCarty, 2001), while 0.3-0.5 g VSS/g COD removed for the whole CAS process. These indicate that a substantial amount of waste activated sludge (WAS) is inevitably generated in WWTPs worldwide. In 2017, the WAS produced in China reached 10.5 million tons of dry mass (MOHURD, 2019), while the present WAS management is facing to pressing challenges due to stringent regulations imposed in many countries, e.g. sanitary landfilling is restricted by the limited land, while incineration might cause adverse impact on the environment due to the potential harmful gas emission and ashes production (Lu et al., 2019b). Nowadays, the volume reduction of WAS is becoming a global issue due to the rapid increase in WAS generation and lack of environmentally sustainable disposal approaches.

In addition, external COD is usually required for ensuring complete denitrification due to insufficient organic carbon source in municipal wastewater. It had been reported that 1.9-3.3 kg of methanol per kg of nitrate-N was needed for complete denitrification in full-scale municipal WWTPs (Peng et al., 2007; Theis & Hicks, 2012). As such, the addition of external organic carbon source should undoubtedly lead to additional operational cost of biological nitrogen removal.

So far, extensive effort has been devoted to exploring feasible engineering options for reducing the aeration-associated energy consumption, while minimizing WAS generation. In this regard, some traditional measures had been taken into consideration, e.g. use of more energy-efficient diffusers, development of sensor-aided on-line control for aeration, etc. For example, 10%-20% of aeration-associated energy savings could be achievable by replacing conventional diffusers with ultra-fine

bubble diffusers (Longo et al., 2016). However, in the nitrification-denitrification process, 4.67 mg/L of dissolved oxygen (DO) is theoretically needed for nitrifying 1 mg/L of ammonium-N through full nitrification. In fact, this seems to imply that further optimization of the process operation with upgrading of aeration equipment may not lead to substantially reduced aeration-associated energy consumption.

Currently, anaerobic digestion (AD) has been widely adopted for concurrent energy recovery and volume reduction of WAS. However, due to low hydrolysis efficiency of WAS, only about 30-50% of VSS could be converted to biogas (Cao & Pawłowski, 2012; Nges & Liu, 2010), which appeared to be insufficient to offset the in-plant energy consumption, whereas a substantial amount of digested solid residue still needed to be disposed of properly. Obviously, innovative biological processes should be developed to address the challenges encountered in the present WWTPs with the aims for achieving energy neutral or even positive biological nitrogen removal.

2 Anammox: an emerging technology for energy-efficient nitrogen removal

2.1 Working principle of anammox

In the conventional BNR process as discussed above, most of influent ammonia-N is removed through nitrification and denitrification, which indeed are highly energy-intensive. In this situation, anaerobic ammonia oxidation (anammox) process has gained increasing interest, and has been believed to serve as a potential game changer of BNR (Kartal et al., 2010; Lu et al., 2019a; Ma et al., 2016). In fact, anammox is an autotrophic biological process in which anammox bacteria oxidize ammonium to nitrogen gas with nitrite as the electron acceptor under anaerobic condition (Mulder et al., 1995). Since ammonia is readily available in municipal wastewater, anammox requires partial oxidation of ammonia to nitrite in a roughly equal molar ratio. Due to

the nature of anammox reaction, oxygen demand and sludge production both can be reduced substantially, and addition of external organic carbon is no longer necessary.

In anammox process, nitrite is a prerequisite for reaction with ammonium. As such, sustainable nitrite supply has been identified as a bottleneck towards stable anammox. So far, two approaches have been explored for nitrite production, i.e. partial oxidation of ammonia to nitrite by ammonia oxidizing bacteria (AOB) known as partial nitrification (PN), and partial nitrate reduction to nitrite by denitrifiers known as partial denitrification (PDN). The integration of PN or PDN with anammox has led to the development of novel biological nitrogen removal processes, i.e. PN-anammox and PDN-anammox for mainstream deammonification of municipal wastewater (Fig. 1). PN-anammox is a complete autotrophic process, while PDN-anammox is of a hybrid heterotrophic-autotrophic process. Compared to the full nitrification-denitrification process, nearly no COD is needed for PN-anammox, whereas less COD required for PDN-anammox. It had been reported that the theoretical COD consumption for complete nitrogen removal via PN-anammox and PDN-anammox were estimated as 0.31 and 1.31 g COD/g $\text{NH}_4^+\text{-N}$, indicating 89.2% and 54.2% reduced respectively in COD requirement compared to the conventional BNR process (Zhang et al., 2019b). The COD saved in both PN- and PDN-anammox can be further used for biogas production via AD which is helpful for improving the overall in-plant energy balance. Moreover, the oxygen required for removing one gram of ammonia-N also decreased from 4.57 g O_2 to 1.95 and 2.39 g O_2 for PN-anammox and PDN-anammox respectively, indicating 57.3% and 47.7% of cut in the aeration-associated energy consumption (Zhang et al., 2019b). Consequently, the mainstream deammonification processes may offer a feasible solution towards energy-efficient

BNR, while several technical barriers as identified in Fig. 1 should need to be resolved before bringing it to full-scale municipal wastewater reclamation.

Insert Figure 1

2.2 Is mainstream anammox ready for full-scale application?

It appears from the above discussion that the mainstream anammox is an emerging technology for energy-efficient nitrogen removal. So far, PN-anammox has been successfully applied at pilot- and full-scale for treating various types of high-strength wastewaters, e.g. sludge digester liquor, landfill leachate, etc. (Nsenga Kumwimba et al., 2020; Sun et al., 2016), while its application for municipal wastewater treatment has hardly been reported so far. On the other hand, PDN-anammox seems still at its early developmental stage, and related research has been mainly conducted with synthetic wastewater in lab-scale units. It should be noted that anammox bacteria had been found in the conventional BNR process treating municipal wastewater (Shu et al., 2015), however, their abundance appeared to be too low to actively contribute to BNR. So far, the technical bottlenecks towards sustainable mainstream anammox have been identified for municipal wastewater treatment, e.g. high COD/N ratio, stable nitrite production, and temperature, etc. (Xu et al., 2015).

As illustrated in Fig. 1, the presence of COD can trigger the competition between anammox bacteria and heterotrophic denitrifiers on nitrite in both PN-anammox and PDN-anammox processes. Due to extremely low growth rate of anammox bacteria (e.g. 0.065 d^{-1}), they are unable to compete with heterotrophic denitrifiers with a typical growth rate of 1.2 d^{-1} on nitrite in the presence of COD (Mokhayeri et al., 2006; Strous et al., 1998), likely leading to the prevailing of denitrifiers and eventual

156 washout of anammox bacteria. It was found that the COD/NO₂⁻-N ratio higher than
157 2.92 favored denitrification of nitrite by denitrifiers, while the growth and activity of
158 anammox bacteria were seriously repressed (Tang et al., 2010). To promote the
159 growth of anammox bacteria against denitrifiers, a lowered COD/N ratio is essential,
160 i.e. a COD/N ratio of 0.5 as suggested by Daigger (2014) for PN-anammox. In PDN-
161 anammox process, part of COD is needed for heterotrophic reduction of nitrate to
162 nitrite. However, even in this case, a higher COD/N ratio should also be avoided in
163 order to prevent further reduction of nitrite to nitrogen gas. Hence, a COD/N ratio of
164 2.0-3.0 had been proposed for PDN (Cao et al., 2016; Ge et al., 2012; Si et al., 2018).
165 In practice, after primary clarification, municipal wastewater generally has a COD/N
166 ratio of 7.0-10.0, which is obviously too high for both PN- and PDN-anammox. These
167 clearly suggest that prior COD removal is necessary for creating a suitable COD/N
168 ratio required for mainstream deammonification of municipal wastewater.

169 In addition to COD/N ratio, a stable supply of nitrite is a prerequisite for
170 mainstream anammox. As can be seen in Fig. 1, the growth and activity of NOB
171 should be suppressed against AOB, while denitrifying bacteria should be controlled to
172 prevent heterotrophic denitrification of nitrite in the presence of wastewater COD in
173 PN-anammox process. In PDN process, the nitrate reduction should be limited to the
174 nitrite stage through enzymatic regulation by suppressing the activity of nitrite
175 reductase against nitrate reductase, or using nitrate respiring bacteria which can only
176 reduce nitrate to nitrite due to lack of dissimilatory reductases of nitrite (Zhang et al.,
177 2019b). However, it should be realized that these approaches for nitrite supply have
178 not yet been fully demonstrated at large scale. Additionally, anammox bacteria had
179 been reported to be sensitive to variation in temperature, e.g. the specific activity of
180 anammox bacteria was lowered by a factor of 10 when temperature decreased from

30°C to 10°C (Lotti et al., 2015a). Obviously, low temperature would negatively affect the stability of anammox process (Laurenzi et al., 2016; Guo et al., 2015). Thus, it had been proposed that the effect of low temperature might be alleviated by adopting anammox biofilms or granules (Lotti et al., 2015a). Consequently, mainstream deammonification still remains a great challenge for low-strength municipal wastewater treatment.

3 Novel dual step process: a promising option for mainstream deammonification of municipal wastewater

3.1 Working philosophy of novel dual step process

As discussed above, mainstream deammonification has not yet been demonstrated in full-scale municipal WWTPs due to high COD/N ratio, inefficient repression of NOB, ineffective control of denitrifiers, etc. It seems that soluble COD should be captured prior to the mainstream deammonification of municipal wastewater, i.e. the removal of organic carbon and nitrogen should be separated into two stages. With this idea in mind, Wan et al. (2016) proposed a novel dual step process, i.e. A-B process, where A-stage is designed to capture organic matters in municipal wastewater for energy recovery, whereas B-stage is tailored for energy-efficient nitrogen removal/recovery. Compared to the conventional BNR process, this A-B process has advantages of (i) enhanced energy recovery from captured COD; (ii) reduced energy consumption associated with bio-oxidation of COD; (iii) creation of a lower COD/N environment for deammonification at B-stage and (iv) substantially reduced sludge production. On the basis of this idea, different A-B process configurations have been proposed, and they appeared highly possible for achieving mainstream deammonification of municipal wastewater.

3.2 Typical **dual step** processes for mainstream deammonification of municipal wastewater

3.2.1 Integrated HRAS and mainstream deammonification process

The high rate activated sludge (HRAS) process has been used for capturing particulate, colloidal and soluble COD from municipal wastewater through biosorption and bioaccumulation. To minimize COD bio-oxidization, HRAS process is generally operated at a short sludge retention time (SRT) and hydraulic retention time (HRT) together with a low DO concentration (Rahman et al., 2016). It had been reported that 55-65% of COD could be removed by HRAS (Wett et al., 2007), which in turn created a low COD/N environment for subsequent deammonification. In this sense, HRAS should be considered as a suitable A-stage prior to mainstream deammonification of municipal wastewater as shown in **Fig. 2**.

Insert Figure 2

As can be seen in **Fig. 2**, municipal wastewater is firstly treated by HRAS for COD capture via biosorption and bioaccumulation, followed by clarification for liquid and solids separation. Subsequently, the supernatant with a low COD/N ratio is fed to the mainstream deammonification unit for energy-efficient BNR, which can be PN-anammox or PDN-anammox. The generated sludge in the whole process is then subjected to anaerobic digestion (AD) for biogas production. It had been reported that about 44% of influent COD could be captured by A-stage HRAS, in which 32% ended up in sludge (Miller et al., 2015). Overall, about 33% of influent COD could be recovered as biogas via AD.

In the HRAS-deammonification process, a longer SRT is preferable for achieving a low effluent COD which can help to minimize the competition between anammox

bacteria and denitrifiers on nitrite. However, the longer SRT, on the other hand, favored COD mineralization, leading to compromised energy recovery through AD. For example, when SRT was prolonged from 0.1 to 2 days, the COD concentration in HRAS effluent was reduced from 55% to 10%, whereas the COD mineralization was almost increased by 5 times, i.e. from 14% to 67% (Jimenez et al., 2015). In fact, the COD/N ratio in the HRAS effluent was found to vary over a wide range of 0.67-8.0 (Table 1), likely due to different SRT control strategies and wastewater characteristics. So far, evidence suggests that HRAS process can be considered as a suitable A-stage for producing the effluent with a suitable COD/N ratio for subsequent deammonification of municipal wastewater.

As elucidated above, influent COD should be captured as much as possible to create a lower COD/N ratio for PN-anammox, while the removal of COD should be controlled in order to create a COD/N ratio of 2.0-3.0 for PDN-anammox. High COD/N ratio had been demonstrated to be harmful for anammox, and it could also promote denitrification at B-stage. For example, at a COD/N ratio of 8.0 in the HRAS effluent, only about 13.4% of nitrogen was removed by anammox against 56% via heterotrophic denitrification (Miller et al., 2015). Moreover, the energy consumption associated with aeration for COD oxidation at B-stage was increased accordingly. On the contrary, for HRAS effluent with a low COD/N ratio, about 75% of total nitrogen was removed through anammox (Table 1). It should also be noted that lower total nitrogen removal was observed at a low COD/N ratio, which was likely due to the failure of NOB repression as indicated by a relatively high $\text{NO}_3^- \text{-N}_{\text{produced}} / \text{NH}_4^+ \text{-N}_{\text{removed}}$ ratio (Lotti et al., 2015a). It thus appears that effective control strategies for NOB repression are still needed towards sustainable deammonification of municipal wastewater, e.g. the combined control of SRT, DO and sludge return ratio (Wang et

al., 2019). In PDN-anammox process, when the COD/N ratio was controlled in the range of 2.0-3.0, the conversion efficiency of nitrate to nitrite via PDN could stably stand at over 90%, and 95% of total nitrogen removal was achievable in treating synthetic municipal wastewater (Du et al., 2019).

Insert Table 1

3.2.2 Integrated anaerobic and mainstream deammonification process

Anaerobic process has gained increasing interest for municipal wastewater treatment due to the obvious advantages of aeration-free, much less sludge production, direct biogas production, etc. (McCarty et al., 2011; Wu et al., 2017). Unlike HRAS, influent COD is directly converted to biogas during anaerobic treatment of municipal wastewater. So far, different anaerobic processes, e.g. up-flow anaerobic sludge blanket (UASB), expanded granular sludge bed (EGSB), anaerobic fixed bed reactor (AFBR) and the anaerobic membrane bioreactor (AnMBR) processes, have been developed for mainstream municipal wastewater treatment, with a COD removal of 60-92% (Dai et al., 2011; Florencio et al., 2001; Kim et al., 2011). It should be pointed out that dissolved methane is an issue that cannot be neglected in anaerobic treatment of municipal wastewater, which indeed may compromise the energy recovery potential of wastewater (Liu et al., 2014). It had been reported that about 20-50% of total methane was dissolved in treated effluent (Gao et al., 2014; Smith et al., 2013). To this end, some techniques, e.g. down-flow hanging sponge reactors, degassing membranes, a flash chamber, etc., have been developed for dissolved methane recovery or removal (Cookney et al., 2016; Gu et al., 2017; Kubota et al., 2014; Matsuura et al., 2015). It should be noted that soluble nutrients (e.g. ammonium) nearly remained unremoved in anaerobic processes. As a result, low COD/N ratios

below 2.5 were generally obtainable in anaerobic effluent (Miao et al., 2018), implying that anaerobic unit could be an ideal lead A-stage for mainstream PN-anammox towards energy-efficient BNR (Table 1).

An integrated anaerobic and PN-anammox process had been successfully demonstrated in the A-2B configuration consisting of an anaerobic fixed-bed biofilm reactor (AFBR) as A-stage, followed by a two-stage PN-anammox process, where a sequencing batch reactor (SBR) as B1-stage and a moving bed biofilm reactor (MBBR) as B2-stage for nitrification and anammox, respectively (Gu et al., 2018). A portion of influent was directed to blend with A-stage effluent via bypass 1, while a portion of A1-stage effluent mainly containing ammonium was directed to B2-stage via bypass 2. The mass balance showed that in the A-stage, 58% of influent COD was directly converted to methane with about 75% of less sludge production as compared to that in the CAS process. The B1-stage SBR stably produced 15 mg N/L of nitrite for subsequent anammox at B2-stage, accounting for 28% of total nitrogen, while another 33% of nitrite-N was denitrified to nitrogen gas with the carbon source from bypass 1. The effective NOB repression could be attributed to the alternating anoxic/oxic conditions coupled with SRT control as previously demonstrated (Xu et al., 2017). The effluent of B1-stage rich in nitrite was blended with the bypassed anaerobic effluent with a low COD/N ratio to make up a proper $\text{NH}_4^+\text{-N}/\text{NO}_2^-\text{-N}$ ratio of 1.1 suitable for anammox at B2-stage.

Recently, a step-feed reactor had been explored for single-stage deammonification of municipal wastewater, in which an upflow anaerobic fixed-bed biofilm reactor (UAFBR) served as the A-stage for COD removal and direct biogas production (Gu et al., 2019b). The step-feed reactor at B-stage consisted of a series of alternating anoxic and oxic chambers packed with anammox biocarriers in anoxic chambers. The

influent was fed to the A-stage UAFBR where 79% of COD was converted to methane, while generating an effluent with the COD/N ratio of 0.44 on average. Subsequently, the A-stage effluent was uniformly directed to the anoxic chambers of the step-feed reactor for anammox, with nitrite produced from oxic chambers. The sludge production in this integrated process was found to be as low as 0.05 g VSS/g COD removed which was about 89% less than that in the CAS process. The mass balance further revealed that about 80% of nitrogen was removed in the step-feed reactor, of which 87% was removed via anammox pathway in anoxic chambers. In addition, the AOB activity was found to be 6 times higher than that of NOB, indicating successful NOB repression which could be attributed to (i) anoxic disturbances caused by alternating anoxic and oxic shift in spatial (Kornaros et al., 2008); (ii) timely nitrite removal in anoxic chambers to exert substrate limitation on NOB (Gu et al., 2019b). Bio-carriers used helped to retain anammox bacteria in anoxic chambers, while avoiding exposure to oxygen in oxic chambers. The aforementioned multi-parameters control strategy could be easily adopted in the step-feed reactor, allowing the easy implementation of the integrated anaerobic and PN-anammox process for mainstream deammonification of municipal wastewater.

3.2.3 Environmental and economical sustainability

With the increasing public concerns over energy crisis and climate change, the conventional BNR with considerable energy demand, high operational cost and large carbon footprint is no longer economically viable and environmentally friendly. The A-B processes discussed above could offer a promising option for mainstream deammonification of municipal wastewater.

Currently, AD for waste sludge treatment has a lower energy efficiency of 31%-61% (Cao, 2011; Gans et al., 2007; Mizuta & Shimada, 2010), i.e. current WWTPs

are operating in an energy-negative fashion (Table 2). The novel A-B processes with decoupled removal of organic and nitrogen matters enable enhancing energy recovery directly from wastewater COD in A-stage, while reducing the energy consumption associated with BNR via mainstream deammonification in B-stage. For example, in the integrated HRAS and PN-anammox process, HRAS unit was able to capture more COD for AD, and harvested sludge from HRAS unit had higher anaerobic digestibility (Bolzonella et al., 2005), indicating a higher methane production potential. For municipal wastewater with 592 mg /L of COD, the recoverable electrical energy in the integrated HRAS and PN-anammox process was estimated to be 0.38 kWh/m³, which was 1.53 times of that in the CAS process (Solon et al., 2019). Meanwhile, the aeration associated energy consumption was reduced by 25%, i.e. from 0.2 to 0.15 kWh/m³ due to reduced COD concentration and PN-anammox for BNR.

Insert Table 2

Different from HRAS, influent COD was directly converted to biogas in integrated anaerobic and PN-anammox process, leading to an energy recovery of 0.37 kWh per cubic meter of municipal wastewater treated, which was about 2.5 times higher than that achieved in the CAS process (Gu et al., 2019b). Moreover, the aeration-associated energy consumption could be cut to 0.03 kWh/m³ because of reduced energy demands for COD bio-oxidation as well as for PN-anammox (Gu et al., 2019b). Consequently, A-B process showed great potential in achieving energy self-sufficient municipal wastewater treatment (Table 2).

Carbon footprint is another serious concern for the environmental sustainability of municipal wastewater treatment. Electrical energy consumption, methane and nitrous

oxide production during BNR are primarily responsible for the large carbon footprint of current WWTPs. Nowadays, electrical energy is mainly produced from the combustion of non-renewable fossil fuels with GHG emission. It had been reported that the annual carbon dioxide emission associated with the generation of electricity used for municipal wastewater treatment in China had already hit 1.14×10^8 tons (Hao et al., 2015). Moreover, it should also be realized that carbon dioxide emission may be no longer free of charge with the implementation of carbon tax (Zhang et al., 2019c). Since the energy consumption could be significantly reduced in the novel A-B processes, the smaller in-plant carbon footprint could be reasonably expected. According to the analysis by Cogert et al. (2019), when taking Modified Ludzack-Ettinger (MLE) process as benchmarking, the GHG emission could be reduced to $0.512 \text{ kg CO}_2\text{e/d/m}^3$ at the COD/N ratio lower than 5 in the integrated HRAS and PN-anammox process. By contrast, $1.72 \text{ kg CO}_2\text{e/d/m}^3$ was achievable in the integrated AnMBR and PN-anammox, even without dissolved methane recovery when treating wastewater with median to high COD concentration. This is largely due to the excellent energy recovery achieved in AnMBR, which could be used to offset the in-plant energy consumption. The GHG emission was $0.371 \text{ kg CO}_2\text{e/d/m}^3$ less when treating wastewater with low influent COD by anaerobic process, due to reduced dissolved methane emission and energy-efficient nitrogen removal via PN-AMX (Cogert et al., 2019).

Nitrous oxide is another significant contributor to the carbon footprint of WWTPs, accounting for about 30% of total in-plant GHG emissions (Liu et al., 2019). Nitrous oxide is a common intermediate of biological nitrogen removal processes, which can be generated via three pathways, i.e. hydroxylamine oxidation, nitrifier denitrification and denitrification (Schreiber et al., 2012; Zhang et al., 2019a). In the conventional

BNR process, 0.001-2.59% of influent nitrogen could be converted to nitrous oxide (Parravicini et al., 2016; Yoshida et al., 2014). It had been well accepted that anammox reaction is not the main source of the nitrous oxide emission (Ali et al., 2016; Okabe et al., 2011), indicating that the anammox-based deammonification process could significantly reduce the potential GHG emission. Consequently, the novel A-B process provides a promising option for mainstream deammonification towards economically viable and environmentally friendly municipal wastewater treatment.

4 Is BNR unique for future municipal wastewater reclamation?

So far, BNR with many various configurations has been extensively employed for municipal wastewater treatment. Mainstream deammonification as an emerging BNR technology is still at its developmental stage with several major challenges pending for solutions. It should be pointed out that municipal wastewater is a source of fresh water, energy and nutrients rather than a “waste” (Liu et al., 2018). The ever-growing global water demand makes the water scarcity increasingly severe. For example, the global water demand by 2030 has been projected to be 6,906 km³, of which about 40% will be in short (Boccaletti et al., 2010). In fact, municipal wastewater reclamation has been considered as an economically viable approach for augmenting traditional water resources (Vincent et al., 2014). One of such examples is Singapore’s NEWater, a reclaimed high-grade ultrapure water.

The integrated aerobic membrane bioreactor (AeMBR) and reverse osmosis (RO) process had been gradually applied for wastewater reclamation to high-grade product water for industrial and in-direct potable use (Daigger et al., 2016). However, AeMBR with aerobic biological treatment as the core, to some extent, shares the same challenges with the CAS-based processes, e.g. intensive energy consumption,

excessive sludge production, low energy recovery potential, etc. (Fenu et al., 2010). As illustrated above, these challenges could be readily addressed by adopting anaerobic treatment. Moreover, anaerobic process integrating with membrane filtration, i.e. anaerobic membrane bioreactor (AnMBR), could offer extra benefits, such as rapid process start-up, high-quality permeate production, etc. (Wu et al., 2017). To this end, integrated AnMBR and RO processes have recently been developed for energy-efficient reclamation of municipal wastewater to high-grade product water (Gu et al., 2019a; Liu et al., 2020). For example, in the study by Liu et al. (2020), an anaerobic fixed-biofilm membrane bioreactor (AnfMBR) was employed as the lead for direct biogas production with minimized sludge generation, while the solid-free permeate from AnfMBR was further treated by RO for high-grade water production.

In the AnfMBR, it was found that the total organic carbon (TOC) in permeate was reduced to 6.58 mg/L, corresponding to a removal efficiency of 95%, while the methane production rate reached 83.35 L/m³ wastewater treated. The observed sludge yield in the AnfMBR was 0.03 g VSS/g COD_{removed}, which only accounted for about 6.7% of that in CAS-based processes (Liu et al., 2020). Due to the nature of anaerobic reactions, most of influent ammonium and phosphate still remained in the AnfMBR effluent, i.e. 34.4 to 32.5 mg N/L and 5.0 to 4.4 mg P/L, respectively. In the subsequent treatment by RO unit, significant removal of TOC, ammonium and phosphate were observed, e.g. above 97% for TOC and ammonium and 99% for phosphate. Meanwhile, the removal efficiencies of other major ions, e.g. sodium, potassium, calcium, chloride, etc., were all above 96%. In some extreme conditions (e.g. high influent nitrogen concentration), an ion-exchanger could be integrated after RO unit for the purpose of polishing (Gu et al., 2019a). As such, for the parameters

investigated, the product water could meet or even better than the typical NEWater quality in Singapore (PUB, 2017) and the quality standards for high-grade reclaimed water in USA, Europe, and China (MEP, 2002; U.S. EPA, 2012).

Furthermore, it was found that a net energy consumption of 0.17-0.21 kWh/m³ product water was achievable in the integrated AnfMBR and RO process at a water recovery rate of 75%, while the energy consumption in the current practice for high-grade water production (e.g. the CAS-based process equipped with AD followed by microfiltration and RO), was calculated to be around 1.04 kWh/m³ product water at the same water recovery rate. Moreover, the integrated AnMBR and RO process is expected to be more stable in operation than the other BNR processes, simply because of the better controllability and predictability of physical separation against biological processes. Looking forward, it is expected that the concept of the integrated AnMBR and RO process might shed light on the future direction of municipal wastewater reclamation towards water, energy and environmental sustainability.

5 Concluding remarks

Mainstream deammonification offers a viable option for energy-efficient municipal wastewater treatment, while it is still difficult for implementation in conventional processes. To this end, the novel **dual step** process segregating COD capture and nitrogen removal into two stages may provide a feasible engineering solution. Analysis showed that novel **dual step** processes integrating mainstream deammonification with HRAS or anaerobic treatment as the lead could provide great opportunities for achieving energy-efficient nitrogen removal. Synergistic integration of biological and chemo-physical process should deserve further systematic investigation towards concurrent energy recovery and water reclamation, and this indeed may represent the future direction of municipal wastewater reclamation.

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738 **Figure caption**

739 Figure 1. Working principles of PN-anammox (pathway ①) and PDN-anammox
740 (pathway ②). AOB: ammonia oxidizing bacteria, NOB: nitrite oxidizing bacteria, DB:
741 denitrifying bacteria, AnAOB: anaerobic ammonia oxidizing bacteria. The yellow,
742 red and dark lines indicate nitrification, anammox and denitrification pathways,
743 respectively.

744 Figure 2. Illustration of an integrated HRAS and mainstream deammonification
745 process.

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749 Figure 1. Working principles of PN-anammox (pathway ①) and PDN-anammox
 750 (pathway ②). AOB: ammonia oxidizing bacteria, NOB: nitrite oxidizing bacteria, DB:
 751 denitrifying bacteria, AnAOB: anaerobic ammonia oxidizing bacteria. The yellow,
 752 red and dark lines indicate nitrification, anammox and denitrification pathways,
 753 respectively.

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Figure 2. Illustration of an integrated HRAS and mainstream deammonification process.

A-stage	B-stage	COD/N ratio after A-stage	N concentration after A-stage, mg/L	TN removal by PN- anammox	References
HRAS	Single-stage plug-flow PN-anammox	0.67	26.8	33–46%	Lotti et al. (2015a)
	Single-stage MBBR PN-anammox	3	21	73%	Laureni et al. (2016)
	Single-stage IFAS PN-anammox	2.8	40–60	50%	Lemaire et al. (2014)
	Single-stage MBBR PN-anammox	1	49	70–75%	Lackner et al. (2015)
	Two-stage AVN PN and MBBR anammox	8	38	13.4%	Miller et al. (2015)
Anaerobic treatment	Single-stage upflow PN-anammox	1-3.5	28	65.7%	Li et al. (2017)
	Single-stage MBBR PN-anammox	0.95	47.4	52%	Kouba et al. (2016)
	Single-stage MBBR PN-anammox	1.2-2.3	29-41	19-40%	Malovanyy et al. (2015)
	Two-stage PN and MBBR anammox	0.7	41	34%	Gu et al. (2018)
	Single-stage step-feed PN-anammox	0.44	43	80%	Gu et al. (2019b)

Note: MBBR: Moving bed biofilm reactor; IFAS: Integrated fixed-film and activated sludge; AVN: Ammonia versus NO_x-N.

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763 Table 2 Energy situation in conventional processes and integrated mainstream deammonification processes.

Process	Sludge production (COD _{sludge} /COD _{inf})	CH ₄ production (COD _{CH4} /COD _{inf})	Energy recovery, kWh/m ³	Energy consumption, kWh/m ³	References
CAS coupled with AD	62.5% ^a	17.9%	0.15	0.44	Cao (2011)
CAS coupled with AD	--	--	0.081	0.258	Gans et al. (2007)
Integrated anaerobic treatment and PN-anammox	7%	48.8%	0.37	0.18	Gu et al. (2019b)
Integrated HRAS and PN-anammox	23.7% ^b	50.8%	0.48 ^c	0.27 ^c	Jia et al. (2020)

764 Note: a: including sludge from primary settling tank; b: after anaerobic digestion of produced bio-sludge; c: calculated value.