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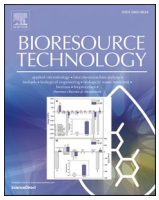

Circular economy-driven ammonium recovery from municipal wastewater: State of the art, challenges and solutions forward

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HIGHLIGHTS

- Biological nitrogen removal compromises ammonium recovery from municipal wastewater.
- Environmental and economic costs with wastewater ammonium are analyzed.
- Ammonium recovery technologies are critically reviewed with feasibility analysis.
- Integrated anaerobic membrane bioreactor and RO processes are proposed for ammonium recovery.
- Resource recovery is game-changing future municipal wastewater reclamation.

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ABSTRACT

In current biological nitrogen removal (BNR) processes, most of ammonium in municipal wastewater is biologically transformed to nitrogen gas, making ammonium recovery impossible. Thus, this article aims to provide a holistic review with in-depth discussions on (i) current BNR processes for municipal wastewater treatment, (ii) environmental and economic costs behind ammonium in municipal wastewater, (iii) state of the art of ammonium recovery from municipal wastewater including anaerobic membrane bioreactor turning municipal wastewater to a liquid fertilizer, capturing ammonium in phototrophic biomass, waste activated sludge for land application, bioelectrochemical systems, biological conversion of ammonium to nitrous oxide as a fuel oxidizer, and adsorption, (iv) feasibility and challenge of adsorption for ammonium recovery from municipal wastewater and (v) innovative municipal wastewater reclamation processes coupled with ammonium recovery. Moving forward, municipal wastewater reclamation and resource recovery should be addressed under the framework of circular economy.

1. Introduction

Ammonium removal from municipal wastewaters is essential to avoid undesirable environmental pollution, e.g. water eutrophication, potential toxicity to aquatic organisms (Feng et al., 2020; Sarvajith et al., 2020), and more and more countries have strictly regulated ammonium concentration in treated effluent. Various biological nitrogen removal (BNR) processes (e.g. nitrification–denitrification, partial nitrification–denitrification, autotrophic anaerobic ammonia oxidation (anammox) etc) (McCarty, 2018; Winkler & Straka, 2019), have been extensively explored, and some of which have been widely adopted for municipal wastewater treatment worldwide. However, it should be noted that the BNR processes are originally developed according to biological oxidation–reduction principle instead of resource recovery, in which most of ammonium is ultimately transformed to nitrogen gas, resulting in a significant loss of ammonium and an increasing energy consumption. For instance, in the conventional nitrification–denitrification process, ammonium in municipal wastewater is first oxidized to nitrate by nitrification in the presence of oxygen, leading to a high aeration-associated energy demand of 2.6–6.2 kWh/kg N removed (Cruz et al., 2019), while denitrification further reduces produced nitrate to nitrogen gas with external organic matter as the electron donor, e.g. 2.86–4.95 kg COD needed for denitrifying one kg of nitrate-nitrogen (Cruz et al., 2019; Theis & Hicks, 2012). To make BNR more energy

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effective and economically viable, anammox process recently has attracted increasing interest, in which anammox bacteria can use nitrite pre-generated from partial nitritation as the electron acceptor to oxidize ammonium to nitrogen gas under anaerobic condition. As the result, dissolved oxygen, energy and external organic carbon demands can be reduced substantially compared to the conventional nitrification and denitrification. However, similar to nitrification–denitrification, ammonium in municipal wastewater is also biologically transformed to nitrogen gas in anammox process, indicating impossible ammonium recovery. In addition, 0.01‰ to 6.6% of total ammonium removed could inevitably end up as N_2O in all BNR processes (Cruz et al., 2019), leading to an increased carbon footprint of municipal wastewater treatment plant as the greenhouse power of N_2O is about 300-time of carbon dioxide.

It appears that current BNR processes for municipal wastewater treatment are not designed for ammonium recovery and obviously depart from the philosophy of circular economy. Obviously, this is seriously challenging the environmental sustainability and economic viability of the BNR processes that have been practiced for decades, while illustrating the environmental relevance and economic necessity of recovering ammonium from municipal wastewater. In fact, ammonium has been known as an effective chemical fertilizer essential for sustaining global agricultural productivity. Currently, almost all ammonium fertilizers are produced through a highly energy-intensive industrial process, known as the Haber–Bosch reaction. It has been believed that ammonium recovery from municipal wastewater is game-changing the current landscape of municipal wastewater reclamation, and is perfectly aligned with the concept and practice of circular economy with the ultimate goal of transforming present wastewater treatment plants to a resource recovery factory (Batstone et al., 2015; Ye et al., 2018). Therefore, the main objectives of this article are to offer a systematic review and analysis of current BNR processes for municipal wastewater treatment, technological feasibility and status of ammonium recovery from municipal wastewater, innovative municipal wastewater reclamation processes coupled with ammonium recovery and future perspective. It appears that the development of future municipal wastewater reclamation technology should be driven by circular economy with a full consideration of resources recovery and reuse.

2. Environmental and economic costs behind ammonium in municipal wastewater

The quantity of global municipal wastewater with an average ammonium-N concentration of 40 mg/L has already hit 380 billion m^3/year, and will continue to increase due to rapid global urbanization and increasing population (Qadir et al., 2020). Currently, global municipal wastewater potentially contains 40 mg/L × 380 billion m^3/year = 15.2 million tons ammonium-N/year, equivalent to about 20 million tons ammonium/year or 18.5 million tons ammonia/year. It should be realized that ammonia supplied to the global agricultural industry is chemically synthesized via a highly energy-intensive Haber-Bosch process under high temperature of 400 to 600 °C and high pressure of 20 to 40 MPa (Bodirsky et al., 2014; Erisman et al., 2008):

$$N_2 + 3H_2 \rightarrow 2NH_3 \quad (1)$$

Generally, hydrogen gas required for ammonia synthesis (Eq. (1)) is primarily produced through steam reforming of natural gas with carbon dioxide as a byproduct. As such, the Haber-Bosch process consumes 3 to 5% of natural gas produced globally, while utilizing about 1–2% of the global energy in an annual basis. The energy consumed in the Haber-Bosch process had been reported to be 7.8, 10.6, and 11.7 MWh/kg ammonia with natural gas, coal, and fuel oil as the respective feedstocks (Giddey et al., 2017), i.e. roughly averaged about 10 kWh/kg ammonia. Meanwhile, about 1.6, 3.0, and 3.8 tons CO_2/ton ammonia, averaged at 2.8 tons CO_2/ton ammonia, could also be generated from each fossil feedstock correspondingly during hydrogen generation. It should be noted that carbon trading scheme is being adopted in more and more countries to control the emission of greenhouse gases according to the Paris Agreement on climate change. For example, Singapore has implemented a carbon tax of US$5 per ton of carbon dioxide emission since 2019 (NEA, 2019), and a carbon price of C$ 10/ton has been adopted in Canada since 2018, and it would eventually reach C$50 in 2022 (Liu et al., 2018a).

The information above indeed allows estimating the economic and environmental costs associated with ammonium in global municipal wastewater. Supposing that ammonium in municipal wastewater was originally produced by the Haber-Bosch method (Eq. (1)), the total electrical energy needed should hit 18.5 million tons/year × 10 MWh/kg = 185 billion kWh/year, together with a carbon dioxide emission of roughly 2.8 tons CO_2/ton ammonia × 18.5 million tons/year = 51.8 million tons CO_2/year. At a carbon price of €20/ton CO_2 equivalent (Bayer & Aklin, 2020) and an electricity price of €0.205 /kWh (Strom-Report, 2020), the total economic and environmental costs for chemically synthesizing ammonia in global municipal wastewater could be roughly estimated to be (€0.205 /kWh × 185 billion kWh/year)+ (€20/ton × (51.8 million tons CO_2/year)) = €39 billion/year. These calculations clearly reveal the economic necessity and environmental relevance of recovering ammonium from municipal wastewater. Unfortunately, all the BNR processes currently adopted for municipal wastewater treatment are based on oxidation–reduction principle by which most of ammonium chemically synthesized at an extremely high cost is all reduced to nitrogen gas. Obviously, this is seriously challenging the environmental sustainability and economic viability of the current BNR processes.

3. State of the art of ammonium recovery from municipal wastewater

The current BNR processes for municipal wastewater treatment indeed are built on the traditional linear economy, i.e. a take-make-dispose model. It appears from the above that municipal wastewater should be regarded as a regenerative resource (e.g. water, energy and chemicals) which is an essential link in urban circular economy with the aims to maximize the resourceful uses of municipal wastewater. In fact, ammonium recovery from municipal wastewater is perfectly aligned with the concept and practice of circular economy, while may open a new horizon for future municipal wastewater reclamation. However, it should be realized that recovery of ammonium from municipal wastewater with a low concentration of around 40 mg/L is technologically and economically challenging. In fact, the existing recovery approaches for ammonium (e.g. air stripping, steam stripping, membrane contactor, electrochemical, electrodialysis, chemical precipitation etc) are only technically feasible and economically cost-effective for the concentrated streams with a high ammonium concentration of thousand milligrams per liter or even higher (e.g. landfill leachate, anaerobic digestate, urine etc) (Cruz et al., 2019). For example, air stripping, known as a process to strip out gaseous ammonia at a pH above 9, is practically feasible for wastewater with an ammonium concentration greater than 2000 mg/L (El-Bourawi et al., 2007). On the other hand, chemical precipitation of struvite through the reaction of $Mg^{2+} + NH_4^+ + PO_4^{3-} + 6H_2O \rightarrow NH_4MgPO_4{\cdot}6H_2O$ has been widely reported for ammonium recovery from highly concentrated anaerobic digester liquor instead of directly from municipal wastewater. In general, struvite can only form in an alkaline environment, while ammonium, magnesium and phosphate should be present at a molar ratio of 1:1:1. However, it is obvious that the conditions required for the formation of struvite are difficult to realize in municipal wastewater as external chemicals (e.g. magnesium and hydroxides) are needed. Several approaches with the promising applicability are thus discussed in the sections below.

3.1. Anaerobic MBR: Turning municipal wastewater to a liquid fertilizer

In addressing the energy challenge in the conventional WWTPs, increasing interest has been given to developing anaerobic membrane bioreactor (AnMBR) for municipal wastewater reclamation. AnMBR is a combination of anaerobic reaction and membrane separation, with the submerged mode being dominant. Compared to aerobic MBR, AnMBR has the advantages of high-efficiency COD capture with minimized generation of waste sludge, low energy demand because of no need for aeration, biogas generation and solids-free permeate rich in nutrients (e.g. N&P) (Liu et al., 2019).

AnMBR coupled with direct reuse of ammonium for irrigation can offer an economically viable and practically feasible engineering option for the cost-effective municipal wastewater reclamation. AnMBR had been demonstrated to be able to remove more than 90% of COD and nearly 100% of solids with the minimum consumption of soluble ammonium and phosphate (Gu et al., 2019; Kim et al., 2011; Liu et al., 2019; Wu et al., 2017), indicating a good quality permeate rich in nutrients. As such, the AnMBR permeate can be used as a nutritional fresh water for direct agricultural irrigation, while the delivery of AnMBR permeate to agricultural areas may be an issue that should be addressed in practical applications.

It should be aware that urban agriculture also relies on freshwater and fertilizer supplies. If AnMBR is applied for handling global municipal wastewater, about 20 million tons ammonium/year in AnMBR permeate could be directly used for irrigation without further processing, while reducing the demand on chemical fertilizers. Meanwhile 380 billion tons of fresh water could also be harvested. The total global water supply has been estimated to be as much as 3300 billion m^3/year (Stenzel et al., 2019), of which 70% is used for sustaining irrigated agriculture (Caldera & Breyer, 2019; Steduto et al., 2018). These in turn indicate that direct reuse of AnMBR permeate would be able to substitute 16% of the current water demand by global agriculture, making the agricultural industry more environmentally sustainable. It should also be noted that biomethane produced from AnMBR in treating municipal wastewater is eventually enough to offset the energy required to drive the operation of AnMBR. For example, about 0.327 kWh of electrical energy/m^3 could also be produced via biogas in AnMBR treating municipal wastewater with a COD concentration of 400 mg/L (Liu et al., 2020b), i.e. 124.26 billion kWh/year would be harvestable at a global scale. In addition, urban horticulture and greening is another big consumer of freshwater and fertilizers. For example, the fertilizer needed for urban horticulture and greening had hit 6092 thousand tons per year by 2019 in China (Hanzhou-Xianlue-Investment-Company, 2020), while China produces about 49.238 billion cubic meters of municipal wastewater per year (MOHURD, 2019), with an ammonium-N concentration of 20 to 30 mg/L averaged at 25 mg/L, i.e. a total ammonium-N of 1.23 million tons which is more than enough for satisfying its nitrogen-fertilizer needs for urban horticulture and greening. These in turn suggest that AnMBR permeate with soluble nutrients should be considered as a potential freshwater and nutrients sources for environmental greening in highly urbanized cities, leading to a significant cut in the usages of chemical fertilizers and natural freshwater. This approach indeed is closely aligned with the urban circular economy towards a complete recovery and reuse of ammonium in municipal wastewater.

3.2. Capturing ammonium in phototrophic biomass

In general, ammonium in municipal wastewater is removed through microbial anabolism (i.e. assimilation) and catabolism (i.e. dissassimilation). In the conventional BNR processes, most of ammonium in municipal wastewater is dissimilated to nitrogen gas (Fux & Siegrist, 2004), while the remaining is assimilated into biomass (Rittmann & McCarty, 2001). Under the circular economy framework, nutrients recovery is to game-change present municipal wastewater reclamation practice (Liu et al., 2018b). Given such a situation, phototrophs (e.g. phototrophic bacteria, microalgae etc) have been extensively explored for concurrent nutrient recovery and wastewater treatment (Abinandan and Shanthakumar, 2015; Hülsen et al., 2016; Wen et al., 2016). These phototrophs able to survive in different conditions (e.g. photo-autotrophic, photo-heterotrophic and chemo-heterotrophic) have been tested for treating various kinds of wastewaters (Arashiro et al., 2018; Arbib et al., 2013; Craggs et al., 2014), while their applicability for handling municipal wastewater with a relatively high COD/ammonium-N ration of around 10 on average has not yet been fully demonstrated.

Hülsen et al. (2016) reported the use of purple phototrophic bacteria (PPB) in a continuous photo-anaerobic membrane bioreactor for concurrent removal of organics and nutrients from municipal wastewater. It was found that about 6.4 ± 1.3 g NH_4^+-N and 1.1 ± 0.2 g PO_4^{3-}-P could be assimilated with 100 g of soluble COD consumed at a hydraulic retention time of 8–24 h, with the effluent COD concentration below 50 mg/L, nitrogen and phosphorus less than 5 mg/L and 1.0 mg/L, respectively. Different from other types of bioreactors, it should be noted that the performance of a photobioreactor is largely determined by the ratio of surface area receiving sunlight to working volume. Generally, PPB are able to assimilate one gram of nitrogen by consuming 16 g of COD (Hülsen et al., 2016; Lu et al., 2018). As discussed above, for a typical municipal wastewater with an average COD concentration of 400 mg/L and ammonium-N of 40 mg/L, it is obvious that COD in municipal wastewater is insufficient for PPB to assimilate all ammonium. Thus, external carbon source is inevitably required to achieve reasonable nutrients assimilation and acceptable effluent concentrations for discharge. For example, a minimum of 200 mg/L of ethanol was added to a photo-MBR (Hülsen et al., 2016). In addition, the pretreatment of wastewater may also be needed (e.g. pre-fermentation) as PPB could only use organic acids, alcohols, sugars etc as the substrates. Obviously, addition of external organics is not economically nor environmentally justifiable. In fact, these make the application of PPB for concurrent municipal wastewater treatment and nutrients recovery highly restrictive, especially at a large scale.

Different from phototrophic bacteria, it has been well known that photoautotrophic microalgae are able to assimilate nitrogen and phosphorus without the need for organic carbon. However, in cultivation of suspended microalgae for municipal wastewater treatment, microalgae may exhibit a poor settleability making biosolid-liquid separation extremely difficult, large footprint and high HRT for raceway ponds, flat-plate photo bioreactors, etc. As such, novel technologies including membrane photobioreactors, immobilized processes (e.g. biofilms, algal–bacterial consortia or granular sludge) have been explored for easy biosolid-liquid separation with a small footprint (Abouhend et al., 2018; Ji et al., 2020; Luo et al., 2017; Yang et al., 2020). Algal–bacterial sludge process can provide self-sustaining oxygen supplies as microalgae produce oxygen by utilizing carbon dioxide generated by bacteria, and external oxygen supply may be unnecessary. Recently, increasing attention has been given to microalgal-bacterial granular sludge and microalgal–bacterial biofilms. Different from suspended microalgal-bacterial sludge process, the separation of biomass and treated effluent in the microalgal-bacterial granular sludge or biofilm process could be much easily managed by adopting a smaller clarifier due to the excellent settleability (Abouhend et al., 2019; Quijano et al., 2017). A novel algal-bacterial biofilm reactor equipped with a submerged LED as light source at the reactor center could ensure 90% removal of organic matter and ammonium with hydraulic retention time of 24 h (Gou et al., 2020), and microalgal-bacterial granular sludge process was capable of removing 92.69%, 96.84% and 87.16% of influent organics, ammonium and phosphorus under non-aeration conditions over a short retention time of 6 h (Ji et al., 2020). More interestingly, most of ammonium and phosphorus in municipal wastewater could be assimilated into microalgal-bacterial granular sludge with minimal nitrogen loss through microbial dissimilation, e.g. about 75% of ammonium in municipal wastewater was fixed into microalgal-bacterial biomass through assimilation (Ji et al., 2020). Microalgae are capable of directly capturing

ammonium from municipal wastewater with a COD/N ratio of 10:1. In addition, the GHG emission potential in microalgal-bacterial granular sludge process could be reduced substantially. For example, the GHG production in the daytime was almost negligible in microalgal-bacterial granular sludge process due to the fact that almost all carbon dioxide produced from biological oxidation of wastewater COD by bacteria could be fully utilized by microalgae (Ji et al., 2020; Qiao et al., 2020; Wang et al., 2020), while the CO_2 emission could be significantly reduced in the night-time because only a small amount of wastewater COD was converted to carbon dioxide due to higher assimilation ability of microalgae (Fan et al., 2020; Qiao et al., 2020). In addition, in both day and night times, the emission of N_2O generated from nitrification-denitrification could be negligible due to the fact that most of wastewater ammonium was removed through microbial assimilation. As such, the carbon footprint of microalgal-bacterial granular sludge process could be reduced by about 77.5% compared to that of the conventional activated sludge process (Zhang et al., 2021). Consequently, microalgal-bacterial granular sludge process could offer a practically feasible and manageable engineering approach for concurrent municipal wastewater treatment, nutrients recovery and carbon dioxide capture.

3.3. Waste activated sludge: A source of nutrients

Currently, a huge quantity of waste activated sludge (WAS) is inevitably produced from municipal wastewater treatment plants globally. In China, the WAS production has been projected to increase from 40 million tons as of 2017 to 60 to 90 million tons by 2020 (Zhang et al., 2019b). Nitrogen and phosphorus in municipal wastewater can be assimilated into biomass through microbial synthesis according to the thumb rule of COD:N:P of 100:5:1. In the current BNR processes, only about 10–15% of ammonium-N (averaged at 12.5%) in municipal wastewater could be sequestered in biosolids (Fux & Siegrist, 2004; Rittmann & McCarty, 2001). As discussed above, the quantity of ammonium-N in global municipal wastewater has already stand at 13.2 million tons/year, of which approximately 13.2 million tons/year × 12.5%=1.65 million tons/year ends up in biosolids. In general, nitrogen captured in stabilized biosolids can be applied for agricultural purposes to substitute the use of chemical fertilizers. For example, about 50% of biosolids are recycled to land in the United States (USEPA https://www.epa.gov/biosolids), and 20–40% of WAS is absorbed by agricultural utilization in the most EU countries, whereas sludge incineration continues to plays a vital role in the management of WAS (Wei et al., 2020). The situation in China shows a decreasing trend of land application of WAS from 60.9% in 2009 to 21.9% in 2017 (Wei et al., 2020). However, it should be noted that biosolids in some cases may not be qualified for land application mainly due to the presence of pathogens, heavy metals and other emerging contaminants (e.g. antibiotics). These in turn indicate that biosolids produced from municipal WWTPs may need to be treated or stabilized prior to land applications, e.g. stabilization and inactivation of WAS by digestion or composting.

Currently, biodrying as a combined composting and drying process has been practiced for WAS treatment for the purpose of stabilization and sterilization (e.g. deactivation of pathogens). Generally, the ammonia in air produced from this process is then recovered through scrubbing in the form of mineral salt. In a full-scale biodrying process with a treatment capacity of 150,000 tons WAS per year, 7300 tons of ammonium sulfate could be recovered annually, meanwhile biosolids residue could meet the Dutch quality standards for land application (Winkler et al., 2013). In fact, biodrying as a cost-effective mean has been gaining attraction in many other countries, e.g. Italy, Germany, United Kingdom, Spain, Poland, Greece, China, India, Malaysia etc (Tun et al., 2018). Although WAS generally contains a wide variety of heavy metals, a recent survey showed that seven heavy metals (i.e. Cd, Cr, Cu, Hg, Ni, Pb and Zn) in the sewage sludge collected from a WWTP located in a most industrialized region of Poland did not exceed the quality standards for agricultural uses (Tytła, 2019), while (Agoro et al., 2020) also reported that five heavy metals (i.e. Cd, Pb, Cu, Zn, and Fe) in sludges collected from three sewage treatment facilities in South Africa were all below the hazardous levels. Even such, the national regulations should be put in place when WAS is considered as a fertilizer or a soil stabilizer in order to minimize the potential contamination of soils, water and crops.

3.4. Biological ammonium recovery in the form of N_2O

Nitrogen oxide (N_2O) is viewed as a byproduct of biological nitrogen removal should not be released due to its strong power of greenhouse effect, while it should also be realized that N_2O indeed is a reactive form of nitrogen with a certain amount of chemical energy. As such, attempts have been made to explore the recovery of N_2O from municipal wastewater as a useful fuel oxidizer.

One of such examples to recover N_2O from wastewater was the coupled aerobic–anoxic nitrous decomposition operation, known as CANDO process, which was designed for enhanced production of N_2O from anaerobic digestion liquor (Scherson et al., 2013). The working principle of the CANDO process is primarily based on several integrated biological reactions among different nitrogen species, i.e. ammonium to nitrite (NO_2^-) followed by NO_2^- to N_2O. NO_2^- to N_2O could be reduced with high efficiencies using accumulated poly-hydroxybutyrate as the primary electron donor produced by decoupled substrate addition strategy of CANDO. Finally, produced nitrous oxide is subject to co-combustion with CH_4 for energy generation. Roughly 30% more energy could be produced with the stoichiometric combustion of 1 mol of methane with N_2O compared with oxygen (Scherson et al., 2014). It had been reported that around 60–80% of nitrite generated in the CANDO process could be transformed to N_2O for further harvesting as a fuel oxidizer (Myung et al., 2015; Scherson et al., 2014). As discussed above, only a small amount of wastewater ammonium is assimilated into biomass (i.e. WAS), i.e. the CANDO process could only recover this amount of ammonium originally present in municipal wastewater. In addition to the CANDO process, chemical production of N_2O had also been achieved through inhibition of cytochrome proteins or iron-mediated reaction in biological wastewater treatment process (Schreiber et al., 2012; Wang et al., 2021; Yu et al., 2019). In this process, the N_2O generation efficiency indeed is largely controlled by the intermediates produced through biological reaction (e.g. NO_2^-, NO, HNO_2 and NH_2OH) as recently reviewed by (Zhang et al., 2019a).

At the current technology stage, the environmental sustainability and economic viability to recover N_2O appears to be debatable due to the challenges associated with harvesting, post-purification of N_2O as well as the emission of residual dissolved N_2O (Zhang et al., 2019a), while leading to a complicated process configuration, high operation cost and insignificant recoverable energy against the total in-plant energy consumption. These seem to suggest that a more comprehensive assessment on the N_2O recovery from municipal wastewater and anaerobic digestion liquor is still needed.

3.5. Bioelectrochemical systems

Currently, increasing evidence shows that bioelectrochemical systems, e.g. microbial fuel cell (MFC), microbial electrochemical cells (MECs), and microbial desalination cells (MDCs), could offer an alternative approach for treating wastewater together with ammonium and energy recovery (Arredondo et al., 2015; Ye et al., 2018). In bioelectrochemical systems, bacteria at the anode can convert chemical energy stored in organic matter into electrons, while facilitating ammonium transfer across the cation exchange membrane. Compared to MFC, applied external voltage in MECs could favor ammonium transfer and better ammonium concentration in the cathode chamber with electric current as the driving force (Kuntke et al., 2014). In MECs, elevated pH at the cathode initiates the quick conversion of ammonium to volatile ammonia, without the need to increase water pH. The

concentrated ammonia stream obtained at the cathode could be further harvested through various methods, such as ammonia stripping, precipitation, etc. So far, MECs have been reported for treating wastewater with high-concentration ammonium (e.g. urine, rejected water from sludge dewatering, etc). For example, MEC had been employed for simultaneous hydrogen production and ammonium recovery. In this MEC, the anode chamber received the synthetic wastewater, while synthetic or real ammonium-rich water produced from sludge dewatering served as the catholyte with a ammonium concentration of 1000 mg N/L, where proton reduction into hydrogen gas resulted in a pH increase, leading to ammonium being converted to volatile ammonia which could be stripped off (Wu & Modin, 2013). As such, an overall ammonium recovery efficiency of 79% could be achieved. Similarly, it was demonstrated in study by (Zamora et al., 2017) that 31–59% of ammonium recovery was achievable in a scaled-up MEC treating urine after struvite precipitation at an applied voltage of 0.5 V, and the associated energy consumption was estimated to be 4.9 to 1.0 MJ/kg N, which was much lower than the energy required for chemical production of one kg of ammonia by the Haber-Bosch method (Eq. (1)). More recently, microbial desalination cells (MDCs) have also been exploited for concurrent treatment, desalination and resource recovery from wastewater with ahigh ammonium concentration, such as anaerobic digester liquor, landfill leachate etc (Liu et al., 2020a). MDCs have been designed for ammonium recovery in the two different configurations, i.e. (i) anion exchange membrane (AEM) facing the anode and cation exchange membrane (CEM) facing the cathode and (ii) a revert arrangement with the opposite membrane installation. In the first MDCs configuration, high-strength ammonium wastewater is desalted in the desalination chamber where ammonium ions penetrate through CEM and are further accumulated on the cathode. (Zhao et al., 2019) reported a tubular MDC for recovery of ammonium from a diluted draw solution of forward osmosis. It was found that 72% of ammonium could be removed in the desalination chamber, while 69% being recovered in the catholyte. In the other configuration where CEM and AEM face to the anode and cathode respectively, ammonium ions in the anolyte and anions in the catholyte are driven into the middle chamber by current generation, allowing the recovery of both cation and anion nutrients (e.g. ammonium, phosphate, potassium etc) from a synthetic urine solution (Ledezma et al., 2017).

In fact, the ammonium removal from wastewater at the anode is primarily determined by (i) the ammonium concentration in wastewater, (ii) the ammonium removal efficiency from the cathode head space by stripping, (iii) applied current density, (iv) the catholyte pH, (v) the type and property of ion exchange membrane and (vi) the equilibria among other ions at both anode and cathode (Arredondo et al., 2015). In addition, bioelectrochemical systems should be integrated with conventional recovery approach, such as air stripping and precipitation. Although bioelectrochemical systems may offer a promising option of ammonium recovery from a concentrated stream, they are still at the early developmental stage and its engineering feasibility has not yet been demonstrated. At the present technology status, they are unlikely feasible for treating municipal wastewater with a low ammonium concentration of around 40 mg/Lin an energy and cost-effective manner.

3.6. Adsorption for ammonium recovery: Feasibility and challenge

Adsorption with a simple process configuration showed the potential to be applied for ammonium removal or recovery (Zhang et al., 2020). To implement the technology for municipal wastewater treatment, it is obvious that adsorbent play a vital role for ammonium removal from wastewater. Many different types of materials have been exploited for ammonium removal from wastewater (Table 1). In addition, suitable and novel process for maximum ammonium recovery by adsorption are highly in need, especially for municipal wastewater.

3.6.1. Adsorbents for ammonium

3.6.1.1. Natural and modified zeolite/inorganic adsorbents. Natural zeolite, clay, clinoptilolite, mordenite, bentonite (mainly montmorillonite), hydrotalcite etc. have been widely studied and applied for removing ammonium from wastewater. For example, natural zeolite is a kind of aluminosilicate mineral with porous structure providing a large inner surface area. Moreover, the framework of zeolite is usually negatively charged. Both electrostatic interaction and aluminosilicate structure may create a strong cation exchange capability of zeolite. It had been reported that natural Chinese zeolite, Australian zeolite, New Zealand's clinoptilolite, and Iranian zeolite with the size of 0.07–0.2 mm had an adsorption capacity of 5.05–12.6 mg/g at an ammonium concentration below 100 mg/L (Huang et al., 2018; Mazloomi & Jalali, 2016). These seem to suggest that the application of natural zeolite for removing ammonium from municipal wastewater is limited due to its relatively low adsorption capacity generally around or below 10 mg/g. As such, artificial synthesis or modification of natural zeolite by surface

Table 1
Summary of common adsorbents for ammonium recovery from wastewater.

Adsorbent	Ammonium concentration (mg N/L)	Adsorption capacity (mg/g)	Contact time (min)	pH	Ref.
Natural Iranian zeolite	40	8.51–10.3	30	7	Huang et al., 2018
Chinese zeolite	20–49	12.6	5–600	6.8	Wang et al., 2016
$NaNO_3$ modified zeolite	1–20	16.9	400	5–8	Fu et al., 2020
Bentonite/Chitosan	10–40	15.9	180	6.0	Han et al., 2020
Chitosan-coated bentonite	26.4	11.6	–	6.0	Gaouar Yadi et al., 2016
Modified zeolite	0–70	21.3	–	–	Han et al., 2020
Na-rich birnessite	2–50	22.6	30	–	Cheng et al., 2017
Polyvinyl alcohol-alginate-zeolite particles	20	28.1	–	–	Putra & Lee, 2020
Synthetic zeolite from fly ash	50	23.9	75	8.0	Zhang et al., 2011
Biochar derived from digested sludge	45	1.4	600	–	Tang et al., 2019
Biochar derived from fruit peel	10–100	4.71	720	8.0	Hu et al., 2020
Biochar derived from rice straw	0–320	4.09	420	–	Xu et al., 2019
Biochar derived from pine sawdust	0–100	5.38	720	7.0	Yang et al., 2018
Modified corncob-biochar	40	12.1	120	–	Vu et al., 2017
Na-type cation exchange resin Amberjet 1200Na	40	27	240	–	Paul Chen et al., 2002
Resin C150H	25–150	28.2	90	6.5	Sica et al., 2014
Sulfonated polystyrene type resin	40	10.4–20	–	7.35–7.77	Malovanyy et al., 2013
Poly(acrylic acid) (PAA)-based hydrogel	50–180	8.8–32.2	240	7.1	Cruz et al., 2018
Modification of PAA-hydrogel with chitosan	100	21.2–36.9	3–5	–	Zheng & Wang, 2009
Nanohydrogel	100	57.6	12	–	Wang et al., 2014

coating, acid, alkali, salt, microwave, and heat treatment methods have been reported for improving ammonium adsorption through increasing active sites, functional groups and cation ions on zeolite (Alshameri et al., 2014; Lei et al., 2008; Leyva-Ramos et al., 2010; Liang & Ni, 2009; Soetardji et al., 2015). For example, zeolite NaY prepared from rice husk ash showed an ammonium adsorption capacity 3 times higher than natural granular mordenite (Yusof et al., 2010), while the zeolite artificially synthesized from fly ash via a solvent-free method exhibited an adsorption capacity of 18.4 mg ammonium/g at an ammonium concentration of 75 mg/L (Liu et al., 2018c).

Generally, most of natural adsorbents have a weak affinity to ammonium, implying that further modification should be needed in order to increase adsorption capacity of low-concentration ammonium in municipal wastewater. For example, bentonite possessed a low ammonium adsorption capacity of less than 5 mg/g at an initial ammonium concentration of 50 mg/L (Angar et al., 2017). In contrast, after surface coating with chitosan, the adsorption capacity of chitosan-coated bentonite for ammonium increased to 11.6 mg/g at an even lower initial ammonium concentration of 26.4 mg/L (Gaouar Yadi et al., 2016). In addition, Na-rich birnessite possesses an ammonium adsorption capacity of 22.6 mg/g at an ammonium-N concentration of 2–50 mg/L and adsorbent dosage of 0.5 g/L, with the electrostatic interaction being the main mechanism (Cheng et al., 2017). Chemically modified zeolite has relatively higher adsorption capacity than natural zeolite, but its modification and synthesis are highly complicated and more costly with uses of many chemicals.

Substitution of Si^{4+} in zeolite by other cations have been shown to be beneficial for ammonium adsorption. For instance, zeolite after modification with NaCl solution exhibited an ammonium adsorption capacity 1.5-fold higher than natural one due to increased Na^{+} content (Lin et al., 2013; Zhang et al., 2016). Lei et al. (Lei et al., 2008) reported a zeolite thermal-treated by 2.5 mol/L of NaCl solution at 65 °C for 24 h as well as a Chinese zeolite modified by 2.5 mol/L NaCl solution under microwave irradiation at 119 W and 2450 MHz for 10 min (i.e. microwave-treated zeolite), and their respective adsorption capacities for ammonium could be increased from 6.96 mg/g to 10.61 and 11.59 mg/g. (Zhang et al., 2011) Zhanused an alkali fusion hydrothermal method to produce synthetic zeolite from fly ash for ammonium adsorption. At an ammonium concentration of 50 mg/L and a zeolite dosage of 1.24 g/L, the adsorption capacity was found to be as high as 23.89 mg ammonium/g. In addition, the physical entrapping of zeolite particles in polyvinyl alcohol-alginate could improve the ammonium adsorption capacity by a factor of 4.3 (Putra & Lee, 2020). Consequently, if zeolite is chosen for ammonium adsorption from low-strength municipal wastewater, natural zeolite is not recommendable, instead modified zeolite should be considered, while a cost-benefit analysis is also needed to justify the economic viability and environmental sustainability.

3.6.1.2. Biochar. Biochar is generally derived from thermal decomposition of various carbon-rich biomasses under hypoxic conditions (e.g. crop straw, woods, agricultural and forestry residues, sewage sludge, livestock manure, algae, animal hair etc), and the raw materials and pyrolysis conditions also affect the physicochemical properties of biochar (Yuan et al., 2019). Compared with activated carbon, biochar derived from these biomasses is relatively cheap and could help reduce air pollution associated with uncontrolled biomass burning (Qiu et al., 2016). The adsorption mechanisms of ammonium by biochar can be attributed to the interaction between the acid groups on biochar and ammonium through the formation of amine salts, meanwhile ammonium ions may also exchange with cations carried by biochar. It should be realized that raw materials and production processes for biochar can significantly affect the physicochemical properties of biochar produced. In general, biochar made from wood has a highly crystalline structure with multiple pores and micropores, while biochar from rice husk and coconut husk have more amorphous, but less porous structures. In addition, less negative charges and lower H/C ratio were found in biochar produced under higher pyrolysis temperatures (i.e., 700 °C) than those produced at 300 °C and 500 °C (Xu et al., 2019; Yuan et al., 2011), resulting in decreased ammonium adsorption capacity. In general, biochar produced from digested sludge, rice straw, sawdust, fruit peels and pine sawdust have a relatively low adsorption capacity of 1.4, 2.20, 4.09, 4.71 and 5.38 mg/g, respectively in treating wastewater with a low ammonium concentration (Hu et al., 2020; Tang et al., 2019; Xu et al., 2019; Yang et al., 2018; Yin et al., 2019). These in turn suggest that modification of biochar for improving its adsorption capacity for ammonium is necessary in practical application. For this purpose, calcination of raw materials at different temperatures has been proven to be effective for producing biochar with a large number of oxygen-containing functional groups (e.g. –COOH, C = O and- COC–, etc.), while acidic or alkaline pretreatment of biochar may generate a more negatively charged surface which is favorable for ammonium adsorption (Han et al., 2020; Vu et al., 2017).

3.6.1.3. Ion exchange resin and others. The cation exchange resin has a networking structure with the functional groups that can exchange with ammonium ion. For example, a Na-type cation exchange resin Amberjet 1200Na has an ammonium adsorption capacity of 27 mg/g for treating synthetic secondary effluent (Paul Chen et al., 2002), and 28.2 mg/g for resin C150H (Sica et al., 2014). In general, the sulfonated polystyrene type resin (e.g. Amberlite IR-120, Diaion SK-1A, Dowex 50 etc) can have an equilibrium adsorption capacity of 10.4–26.6 mg ammonium-N/g for ammonium removal from municipal wastewater (Gu et al., 2019; Malovanyy et al., 2013). It appears that the commercially available cation exchange resins possess a relatively higher adsorption capacity for ammonium compared to zeolite and biochar, while they are much more expensive than both zeolite and biochar, e.g. US$ 390/kg for Dowex 50WX8 resins (Argun et al., 2007). Hydrophilic hydrogels with a three-dimensional polymeric network full of hydrophilic groups (e.g. –COOH, –COO and –OH) on the skeleton can bind with ammonium through complexation, covalent bonding, electrostatic interactions or physical adhesion (Cruz et al., 2018), making the hydrogel having a fast adsorption rate and high adsorption capacity for ammonium. A commercial poly(acrylic acid) (PAA)-based hydrogel exhibited an adsorption capacity of 8.8 mg ammonium-N/g at an ammonium-N concentration of 50 mg/L in treating a synthetic municipal wastewater (Cruz et al., 2018). In addition, it was found that the adsorption capacity of ammonium could be increased to about 30 mg/g after modification of PAA-hydrogel with chitosan (Zheng & Wang, 2009). A novel nano-hydrogel produced from carboxymethyl chitosan and maleic anhydride N-acylated chitosan with palygorskite had been reported to have an maximum ammonium adsorption capacity of 57.6 mg/g (Wang et al., 2014). In fact, nanomaterials with great characterizations (Zhang & Liu, 2020a, 2020c), have also been explored as an adsorbent of ammonium, e.g. Fe_3O_4 nanoparticle (Zare et al., 2016). It should be noted that the nanomaterials used for ammonium adsorption are still at the earlier developmental stage, and there is a long journey ahead of their large-scale application for treating municipal wastewater.

3.6.2. Engineering processes for ammonium recovery from municipal wastewater via adsorption

Different process configurations have been developed for ammonium recovery by adsorption. For instance, a conceptualized process incorporated adsorption as pretreatment followed by post biological assimilation was proposed for municipal wastewater treatment, with sidestream recovery and regeneration (Cruz et al., 2020), in which 70–80% of ammonium-N was firstly harvested by adsorption and 15% was removed through microbial assimilation at a high-rate aerobic stage to ensure very low ammonium concentration in the effluent. While this process may encounter the risk of competitive adsorption on the adsorbents, this means adsorption can be preferentially placed after

organic removal by upstream operation units was proposed (Cruz et al., 2019). However, it should be noted that the upstream operation units (e.g. chemically enhanced primary, anaerobic, high-rate activated sludge etc) proposed for organic removal might not be appropriate from a point view of real application because the effluent produced (i.e. influent to adsorption unit) would contain relatively high-concentration microbes which would inevitably contaminate adsorbent via surface attachment. Obviously, such a situation should be avoided, otherwise the performance of adsorbent is largely compromised, while causing the difficulty in the regeneration of used adsorbent. To address this issue, an anaerobic MBR (AnMBR) coupled with zeolite adsorption for ammonium has been developed (Li et al., 2020), in which the effluent from AnMBR which is nearly particle-free and rich in soluble nutrients is further subjected to adsorption by zeolite for ammonium removal. After which, the RO unit serves a barrier for further removing residual organics, other major ions including heavy metals in AnMBR effluent for producing high-grade product water. Since RO unit is able to reject more than 95% of ammonium, very low ammonium effluent levels can be achieved (<1 mg/L NH_4-N) even when a partial removal of ammonium was achieved by zeolite adsorption. Further, the ammonium-loaded adsorbents could be reused and ammonium recovery by regeneration. However, it should be noted that the underlying mechanism for ammonium adsorption by zeolite is through cation exchange. As such, polyvalent cations on zeolite (e.g. Ca^{2+}, Mg^{2+} etc) may be released into liquid after ammonium adsorption, and this in turn impacted on the performance of subsequent RO unit as RO membrane fouling is more susceptible to multivalent cations than monovalent cations (Li et al., 2020).

3.6.3. Adsorption of ammonium: Environmentally sustainable?

Although extensive attention has been dedicated to exploring adsorption as a possible mean for ammonium recovery from municipal wastewater. However, as discussed above, it should be noted that the currently available adsorbents for ammonium have a very low adsorption capacity and selectivity, while frequent regeneration of used adsorbents also adds in an additional barrier. In fact, these make the large-scale application of adsorption for ammonium recovery from municipal wastewater with an extremely large volume almost impossible technically and economically. For the purpose of illustration, a municipal wastewater treatment plant with a flow of 500,000 m^3/day and an influent ammonium-N of 40 mg/L is analyzed for ammonium adsorption, with a fixed-bed zeolite adsorber. If the empty bed contact time (EBCT) is assumed to be 20 min, the working volume of zeolite bed is calculated to be 500,000 m^3/day × 20 min = 6945 m^3. On the other hand, considering the commercially available zeolite has an adsorption capacity of 10 mg ammonium-N/g, the influent ammonium-N loading to the zeolite adsorber can be determined as 500,000 m^3/day × 40 mg/L = 20,000 kg/day. Thus, the theoretical service time of the zeolite adsorber with a working volume of 6945 m^3 is calculated to be (600 kg/m^3 × 6945 m^3 × 10 mg/g)/(20,000 kg/day)≈ 2 days if zeolite has a packing density of 600 kg/m^3, while the actual service time indeed is much shorter than two days. These suggest that regeneration of used adsorbent should be initiated every two days, which appear to be too short for any practical operation. To be more operationally reasonable, if the service cycle is prolonged to 10 days, the volume of the zeolite adsorber would be increased to 34,725 m^3 containing 600 kg/m^3 × 34,725 m^3 = 20,835 tons of zeolite, which would result in a substantial increase in the overall footprint of WWTP. It has been reported that used zeolite could be chemically regenerated in-situ by 1 M of sodium chloride solution for 5 min at a flow rate of 10 BV/h (Rahmani et al., 2009), i.e. about (10 BV/h × 34,725 m^3 × 1 mol/L) × 5 min = 28937500 mol of NaCl = 1693 tons of NaCl for one cycle of regeneration. Meanwhile, to further recover the eluted ammonium, sodium hydroxide solution (i.e. 1.0 M) is also needed for rising the solution pH up to above 11, allowing stripping out of ammonium in the form of ammonia gas. It seems that direct adsorption of ammonium from municipal wastewater may not technically preferable, environmentally sustainable and economically viable due to increased footprint, use of the chemicals and generation of wasted chemical solutions which are difficult to be further managed, while explaining why there is no full-scale application of adsorption process for ammonium removal and recovery from municipal wastewater thus far. In fact, these operational hurdles all result from an extremely large volume of municipal wastewater with a low ammonium concentration and low adsorption capacity of adsorbent.

Recently, an integrated anaerobic fixed-film membrane bioreactor (AnfMBR) and RO process was reported for reclaiming municipal wastewater to high-grade product water (Liu et al., 2020b). It was found that TN and TP were slightly reduced from 34.39 ± 1.18 mg/L and 4.96 ± 0.26 mg/L to 32.47 ± 1.04 mg NH_4^+-N/L and 4.38 ± 0.26 mg PO_4^{3-}-P/L respectively, i.e. almost no ammonium and phosphate were removed in AnfMBR. It was further shown that more than 97% of ammonium and 99% of phosphate could be rejected by RO in this integrated process. As the RO unit was run at a productivity of 75%, the ammonium concentration in RO concentrate could be around 130 mg/L. It is reasonable to consider that the recovery of ammonium from RO concentrate (ROC) with a smaller volume and a higher ammonium concentration is more practically feasible and economically cost-effective than the direct recovery from municipal wastewater. In fact, the adsorption capacity of an adsorbent could be improved at a relatively higher ammonium concentration (e.g. >100 mg/L). For the above example with an operation cycle of 10 days, the volume of zeolite bed could be reduced to the half of that for the direct recovery of ammonium from wastewater as described above, while the chemicals needed for regeneration should also be halved.

Consequently, it appears that the currently available adsorbents with a low adsorption capacity may not be suitable for direct or indirect ammonium recovery from municipal wastewater at a large scale primarily due to the frequent chemical regeneration with the use of a large amount of chemicals, while making the whole wastewater treatment process complicated and environmentally unfriendly.

3.7. Innovative municipal wastewater reclamation processes coupled with ammonium recovery

Although intensive effort has been dedicated to the recovery of ammonium from municipal wastewater, it appears from the analysis above that the regeneration of used adsorbent with a low adsorption capacity makes the recovery of ammonium through adsorption technically unfeasible, environmentally unfriendly and economically inviable. Thus, there is an urgent need to explore innovative engineering processes for concurrent wastewater reclamation and ammonium recovery without involving chemical regeneration.

3.7.1. Integrated AnfMBR-biochar adsorption-RO process

Biochar produced by pyrolysis of various kinds of biomasses has been commonly used as a soil ameliorant for enhancing carbon sequestration as well as improving soil fertility (Beesley et al., 2011; Yuan et al., 2019). As a result, increased agricultural productivity is expectable. Moreover, the porous structure of biochar is helpful for effectively retaining water and nutrients in soil with reduced leaching of fertilizer into surrounding surface water and groundwater (Yao et al., 2012). Biochar can also serve as a harbor for diverse beneficial soil microorganisms (Beesley et al., 2011). As such, biochar has been regarded as an extremely effective soil amendment promoting good soil environment as well as plant health. On the other hand, as discussed above, biochars produced respectively from wood, rice husk and coconut husk have a maximum adsorption capacity of 5 to 10 mg/L. Therefore, an integrated AnfMBR-biochar adsorption-RO process is put forward (Fig. 1a). Different from the processes described in Section 3.6.2, biochar after adsorption of ammonium can be directly deployed as an ammonium-loaded solid fertilizer as well as a soil ameliorant for agricultural applications without the needs for further processing (e.g. regeneration, gas stripping at elevated alkaline pH, dissolution of stripped-out ammonia gas in an acidic solution

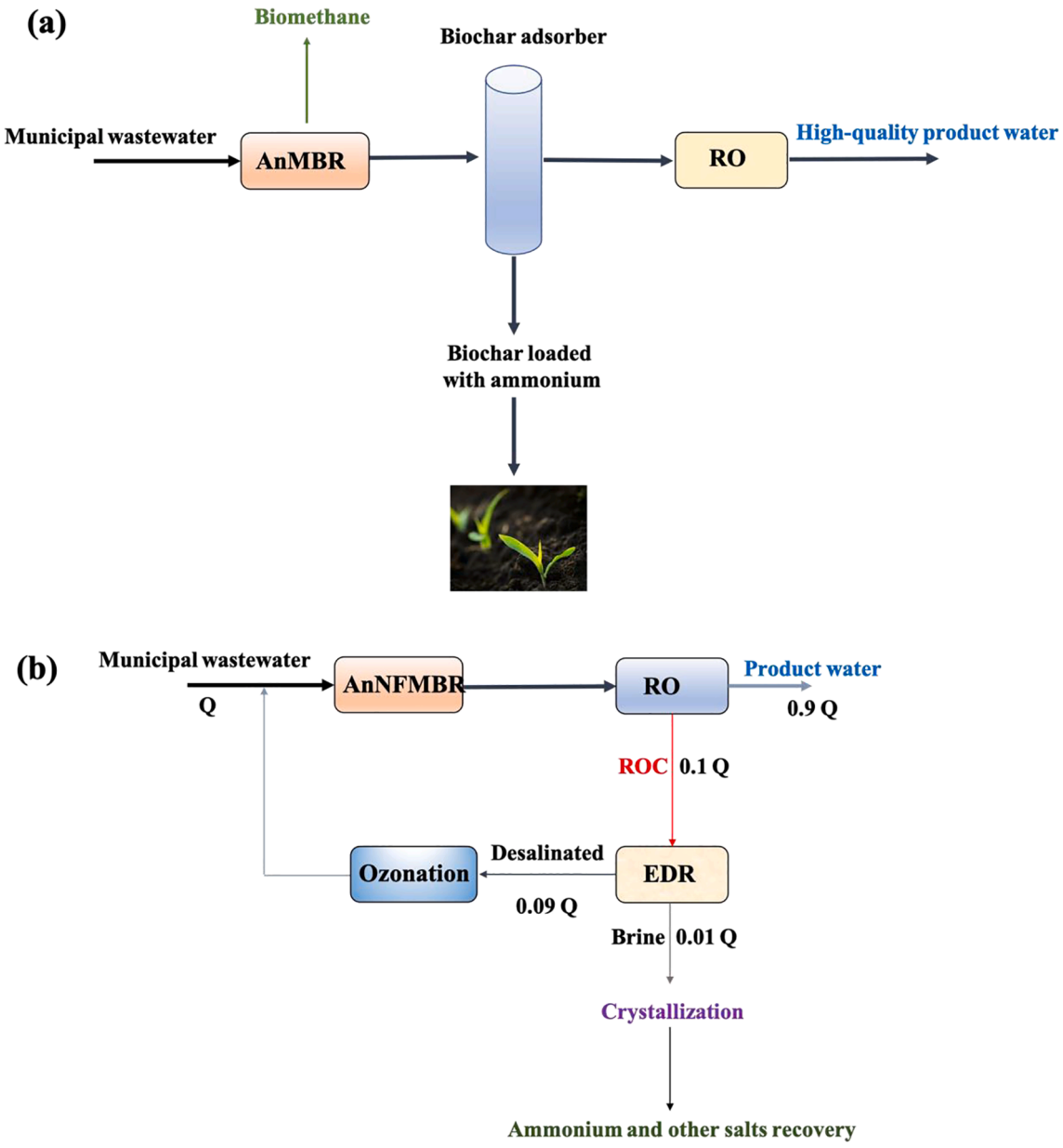

Fig. 1. (a) Integrated AnfMBR-biochar adsorption-RO process for concurrent municipal wastewater reclamation and ammonium recovery. **(b)** An innovative integrated AnNFMBR-RO process coupled with EDR and ozonation towards salts crystallization for resources recovery. Q represents influent wastewater flowrate. Adapted from Zhang and Liu (2020b).

followed by crystallization), making the whole process nearly chemical-free and more environmentally sustainable.

Carbon sequestration in agricultural soils has been demonstrated to be essential for the climate change mitigation since soil organic carbon in most of agricultural lands is being depleted. Returning of biochar to agricultural soils has been suggested as a feasible option for abating global climate change through carbon sequestering (Tan et al., 2017). The maximum sustainable technical potential of biochar to store greenhouse gases (e.g. carbon dioxide, methane and nitrous oxide) has been estimated to be 1.8 Gt CO_2-C(eq)/year (Woolf et al., 2010). It was observed that biochar could help to increase the shoot and root biomass of wheat at a biochar dosage rate below 22.4 ton/ha (Bista et al., 2019). In another study, 30 tons of biochar/ha were supplied to a durum wheat field with up to 30% increase in biomass production and yield (Vaccari et al., 2011). The application of biochar to soils was found to be helpful for increasing aboveground productivity, crop yield, soil microbial biomass, rhizobia nodulation, soil phosphorus, soil potassium, total soil nitrogen and total soil carbon compared with control (Biederman & Harpole, 2013). On the other hand, it has been reported that approximately 0.61 to 0.80 ton of carbon, equivalent to 2.2 to 2.93 tons of carbon dioxide with an average of 2.66 tons of carbon dioxide could be sequestered by one ton of biochar applied to the soils (Blasing et al., 2004). For the purpose of discussion, assuming that 20 tons of ammonium-loaded biochar harvested from the process illustrated in Fig. 1a are applied to every hectare of agricultural land, this will reduce carbon dioxide emission by 53.2 tons/ha, whiling bringing in 257 kg/ha of ammonium if the adsorption capacity of biochar is to be 10 mg ammonium-N/g. These clearly suggest that ammonium-loaded biochar produced from the proposed integrated process (Fig. 1a) holds a great potential promise in being a three-fold win–win–win solution to wastewater reclamation, carbon sequestering, and ammonium fertilizer for sustainable agriculture, deserving further exploitation and exploration.

3.7.2. Integrated AnNFMBR-RO process coupled with EDR and ozonation

Recently, Zhang & Liu (2020b) proposed an integrated anaerobic nanofiltration membrane bioreactor (AnNFMBR)-RO coupled with electrodialysis reversal (EDR) and ozonation (Fig. 1b) which provides a potential opportunity for high-efficiency recovery of various mineral resources including ammonium. In general, more than 95% of nutrients in the AnNFMBR effluent can be retained by RO unit and ends up in ROC which is further subjected to EDR for further water recovery and desalination with a productivity of 90%. However, EDR is incapable of removing soluble organic matters accumulated in ROC, thus ozonation is proposed to improve biodegradability of residual COD in ROC. Afterwards, ozonated water is directed back to the AnNFMBR. Overall, only about 1.0% as the final concentrate with high ammonium concentration of about 4000 mg/L is produced after EDR from the proposed integrated process (Fig. 1b). The cost analysis of the integrated process in Fig. 1b can be found elsewhere (Zhang and Liu, 2020b), while it should be noted that fouling in AnNFMBR may be a potential issue that should be tackled in future development.

Different from other metallic salts, ammonium is readily converted to highly volatile ammonia gas at an elevated pH, which can be further harvested through a simple stripping. As recommended by the (USEPA, 2000), for a higher ammonia concentration above hundred milligrams per liter, steam stripping instead of conventional air stripping may be more economically viable. Meanwhile, the recovery of other salts is achievable via selective crystallization which is determined by the different saturations of salts (Kieselbach et al., 2020). For example, if the saturation of a target salt is reached during evaporation with the formation of precipitate, the process can be continued until the next salt reaches its saturation. In fact, the selective crystallization has been demonstrated to be effective in separating single salts as solid crystals from a concentrated stream with a complex salt composition (Kieselbach et al., 2020). It should be realized that the recovery of resources from municipal wastewater represents a growing interest for future municipal wastewater management under new circular economy, and it indeed is not only meant for water recovery, but also for other valuable components in municipal wastewater (e.g. ammonium, metallic salts etc). Although zero liquid discharge (ZLD) has been extensively explored for managing various kinds of industrial wastewater, this concept had not yet been actively promoted in municipal wastewater reclamation. As described in Fig. 1b, ZLD may offer an ambitious and innovative strategy for municipal wastewater management that maximizes water and resource recovery towards a circular economy. There is a general perception that achieving ZLD is energy intensive and costly. However, for a highly concentrated brine with a very small volume (Fig. 1b), the ZLD approach would not lead to a substantial increase in the overall treatment cost of municipal wastewater (Zhang & Liu, 2020b). Moreover, with the implementation of more and more stringent environmental regulations as well as increasing values of freshwater and resources, ZLD could be expected to be a beneficial or even a necessary option for municipal wastewater management in future.

4. Perspectives

Municipal wastewater with an extremely large volume has been gradually accepted as an alternative and renewable source of fresh water and nutrients. However, the current BNR processes designed with the concept of biological oxidation–reduction transform most of wastewater ammonium to valueless nitrogen gas at the high economic and environmental costs, while generating a substantial amount of greenhouse gases. It appears that the current BNR processes have become a road barrier towards ammonium recovery from municipal wastewater. In fact, the design and operation of current biological processes for municipal wastewater treatment are largely based on the concept of linear economy (i.e. take-consume-dispose), with a primary goal of satisfying the effluent discharge standards, but almost all resources in municipal wastewater are wasted without necessary recovery and reuse, while this obviously departs from the spirit of circular economy. It is apparent that circular economy should be a major driver in the future technological development for municipal wastewater management with the primary focus on the recovery of freshwater, energy and resources. Different from linear economic model, circular economy is regenerative, while aiming to gradually decouple growth from the consumption of finite resources. As discussed in this article, the currently available technical approaches for ammonium recovery from municipal wastewater all appear to be economically costly and environmentally unsustainable due to relatively low ammonium-N concentration (e.g. only about 40 mg/L). It should be realized that the ultimate engineering solution to tackle the challenge of concurrent municipal wastewater reclamation and nutrient recovery should rely on zero liquid discharge with crystallization of salts including ammonium as recently proposed (Zhang & Liu, 2020b). Along with this working philosophy, the integrated anaerobic AnNFMBR-RO process coupled with EDR and ozonation for the post-treatment of ROC would be able to realize the crystallization of final EDR brine at an affordable energy cost, while ammonium and other salts could be easily recovered from the crystalized brine. Therefore, a paradigm shift of biological wastewater treatment processes from removing to recovering is absolutely necessary, which is an essential step towards the greater environmental and economic sustainability.

5. Conclusion

One should be aware that circular economy is unavoidably game-changing technological development for municipal wastewater management towards a greater environmental and economic sustainability, and this requires engineering solutions that can realize the concurrent water, energy and resource recovery. Among all approaches currently available for ammonium recovery from municipal wastewater, the innovative integrated anaerobic fixed-film MBR-biochar adsorption-RO and anaerobic AnNFMBR-RO coupled with EDR and ozonation processes have a great potential for concurrent recovery of high-grade product water, energy and nutrients. Such technological advances could gradually transform wastewater management from a linear to a circular economy model.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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