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# Bio-membrane integrated systems for nitrogen recovery from wastewater in circular bioeconomy



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#### HIGHLIGHTS

#### Nitrogen recovery from wastewater is evaluated in a circular bio-economy framework.

- Bio-membrane integrated systems are proposed for nitrogen recovery.
- Bio-products generated in the recovery system are critically reviewed.
- Cost and energy analyses of large-scale systems for long-term operations are needed.

#### G R A P H I C A L A B S T R A C T



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#### ABSTRACT

Wastewater contains a significant amount of recoverable nitrogen. Hence, the recovery of nitrogen from wastewater can provide an option for generating some revenue by applying the captured nitrogen to producing bio-products, in order to minimize dangerous or environmental pollution consequences. The circular bio-economy can achieve greater environmental and economic sustainability through game-changing technological developments that will improve municipal wastewater management, where simultaneous nitrogen and energy recovery are required. Over the last decade, substantial efforts were undertaken concerning the recovery of nitrogen from wastewater. For example, bio-membrane integrated system (BMIS) which integrates biological process and membrane technology, has attracted considerable attention for recovering nitrogen from wastewater. In this review, current research on nitrogen recovery using the BMIS are compiled whilst the technologies are compared regarding their energy requirement, efficiencies, advantages and disadvantages. Moreover, the bio-products achieved in the nitrogen recovery system processes are summarized in this paper, and the directions for

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future research are suggested. Future research should consider the quality of recovered nitrogenous products, long-term performance of BMIS and economic feasibility of large-scale reactors. Nitrogen recovery should be addressed under the framework of a circular bio-economy.

#### 1. Introduction

#### 1.1. Principal concepts of the circular bioeconomy

Within the framework of a linear economy, finite natural resources are utilized to generate valuable products, goods and services, energy and chemicals. This may consequently cause a substantial generation of industrial and residential wastes, which will be discarded into landfill or wastewater and simply adds the level of pollution. Thus, treatment risks are increased and waste disposal processes and industrial activities can also result in the production of greenhouse gases (GHGs), which triggers worse global warming. Unlike the linear manner of a fossil fuel-based economy, circular economy aims to recycle and reuse wastes and raw materials in a closed-looped strategy with regenerative and restorative design properties. This in turn contributes to eliminating toxic generation, as shown in Fig. 1.

Within the framework of the circular economy, a bio-economy can achieve the conversion of waste into new clean energy, and valuable material sources are reused through using the least harmful techniques (Guardia et al., 2019). The transition from a finite resource-based economy to a sustainable bio-based one is highly desirable to promote sustainable and viable economic development. Over the last decade, the concept of circular bio-economy has been accepted by many national governments, to achieve: reduced environmental damage, improve sustainable and advanced waste management, and change the way the economy works. The concept of the bio-economy has, indeed, opened up new ways to avoid secondary pollution, enhance environmental protection (from non-degradable and persistent pollutants), produce sustainable resources, create more job opportunities, and mitigate emissions of GHG. Substantial wastewater exists in many forms throughout society; for example, it was reported in Tehran alone,  $186.06 \pm 7.85$  L/capita per day domestic wastewater is produced (Mesdaghinia et al., 2015). Previously, wastewater was considered as a

potential issue and waste in society, but the concept has shifted due to it being considered as an abundant source of energy and chemicals. Thus, the recovery and reuse of resources obtained from wastewater in other industries (e.g., energy and agriculture sectors) will benefit and support economic growth, especially given the final product will be one of higher quality and have negligible effects on soil, water and air.

#### 1.2. Importance of nitrogen recovery in circular bioeconomy

Nitrogen (N) plays an important role in fertilizer production and food security. The explosive increase in global population has resulted in higher demands for nitrogen-based fertilizers (Vineyard et al., 2021; Wei et al., 2018). However, the production of nitrogen-based fertilizers through the Haber-Bosch process occupies ~1.6% of global CO<sub>2</sub> emissions, and 1–2% of global energy consumption (Groenestein et al., 2019; Hou et al., 2018). Therefore, it is essential to look for alternative nitrogen sources to sustainably ensure the supply of green fertilizer, especially given the dual challenges of supply security and climate change. Apart from this, most agricultural nitrogen is lost to the aquatic environment and air, whilst only 17% is consumed by humans via livestock or crops (Beckinghausen et al., 2020). The accumulation of nitrogen in water bodies may result in eutrophication, endangering aquatic environments and ecosystems. Current full-scale techniques to remove nitrogen from wastewater include partial nitrification-anammox processes, nitrification-denitrification, and their combinations. Through these processes complex nitrogenous compounds are finally decomposed by bacteria into atmospheric nitrogen gas (N2). Nonetheless, advanced nitrogen removal technologies are still challenged by energy consumption, in which the energy input for the removal of nutrients (i. e., nitrogen and phosphorus) ranges from 0.39 to 3.74 kWh/m<sup>3</sup> (Gu et al., 2017). Simultaneously, GHGs such as nitrous oxide (N2O) and methane (CH<sub>4</sub>) are generated in the removal process (Law et al., 2011).

Overall, the process related to nitrogen production and removal is

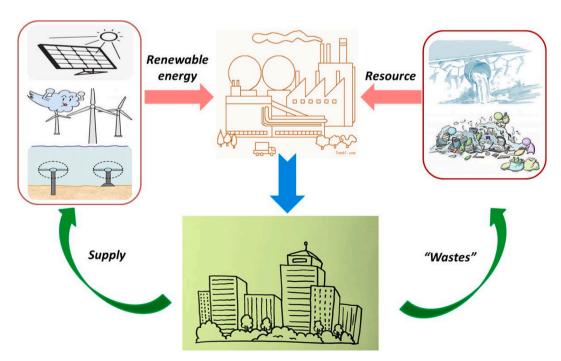


Fig. 1. Concept of circular bio-economy.

energy-intensive, in which the Haber Bosch process transforms the atmospheric nitrogen to ammonia, and then the nitrogen removal process produces atmospheric nitrogen from ammonia. Some researchers have found that nitrogen recovery in wastewater treatments would allow for a circular flow of the nitrogen cycle when compared to nitrogen removal. More specifically, the nitrogen recovery from wastewater can not only resolve to some extent environmental issues such as eutrophication, but also offer supplementary nitrogen sources for fertilizer production. Hence, it is essential to use techniques which are efficient and sustainable, in order to recover nitrogen from wastewater with products available for further use in the industry.

Bio-membrane integrated systems (BMISs) have come into consideration for the nitrogen recovery from wastewater efficiently and sustainably. In this review, BMISs are categorized into side-stream and submerged configurations, in which their energy cost and efficiency were compared. Besides, a generation of bio-products in nitrogen recovery was also reviewed. This research discussed the bio-membrane integrated system for the nitrogen recovery in a circular bioeconomy concept.

#### 2. Nitrogen recovery by BMISs

#### 2.1. Reasons for using BMISs for nitrogen recovery

Nitrogen recovery techniques aim to use the ammonium from waste streams to generate valuable products, including the following: fertilizers in agriculture; a food source for animals or humans; or to cultivate bacteria for their further application in the biogas/biofuels industry (Ye et al., 2021b). It cannot blindly accept to directly recover ammonium from wastewater without any additional treatment because of the possible high content of metals, and other contaminants in wastewater. To make this a viable product with a consistent and safe quality for the future, the ammonium recovery process must involve the biological process to efficiently remove foreign substances so that recovered products are useable. Besides, ammoniation can increase the amount of reactive nitrogen available for its recovery. Although the wastewaters used for nitrogen recovery exist in large volumes, the small concentration of nitrogen in some streams may hinder its recovery. For this reason, membrane technology is employed to enrich the ammonium within the bioreactor, which has low energy consumption compared to other concentrative technologies (Chen et al., 2021; Liang et al., 2019; Paniagua-Michel et al. (2015); San Roman et al., 2010). Therefore, biological process integrating with membrane technology would be beneficial for producing ammonium-rich solutions in wastewater treatment. Here additional steps may be required to achieve final products available for direct application in industry, such as struvite precipitation and adsorption.

#### 2.2. Current status of nitrogen recovery by BMISs

According to the different implementations of membrane in or outside a bioreactor, there are two typical types of BMISs for the recovery of nitrogen, including side-stream and submerged configurations. In the side-stream BMIS, a separate unit containing membrane module is placed outside of the bioreactor, in which the influent would flow successively through the membrane module and bioreactor (Cerrillo et al., 2021a). For the side-stream BMISs, membranes can be classified into electrodialysis (ED), membrane distillation (MD), forward osmosis (FO), reverse osmosis (RO) and nano filtration (NF) according to the driving force across the membrane. Moreover, the membrane module is directly installed into the liquid phase of the bioreactor (Zhang and Angelidaki, 2015). This system namely submerged BMIS mainly includes anaerobic membrane bioreactor (AnMBR), osmotic membrane bioreactor (OMBR), bio-electrochemical system (BES) and membrane photobioreactor (MPBR). Compared to the side-stream BMISs, the submerged BMISs are favored since they involve less

infrastructure, lower energy consumption and fewer costs. Tables 1 and 2 summarize the side-stream and submerged BMISs for the nitrogen recovery from wastewater.

#### 2.2.1. Side-stream BMIS

2.2.1.1. ED. ED is an electrical-driven separation system with ionexchange membranes (IEMs) equipped. ED stack typically consists of alternating anion exchange membranes (AEMs) and cation exchange membranes (CEMs), in which such IEMs are placed between concentrated and dilute chambers in a multi-chamber cell (Al-Amshawee et al., 2020). Each compartment contains an inert electrode. As a mature separation technology, ED employs an electrical field to drive ions including ammonium ions to pass by IEMs. Consequently, ammonium ions can be enriched at the cathode chamber across the CEM for their further recovery through air stripping, adsorption or chemical precipitation (Desloover et al., 2012; Gurreri et al., 2020; Ippersiel et al., 2012; Vineyard et al., 2020). It is obvious that the ammonium migration highly relies on the current as well as its further recovery in the ED process. By life cycle assessment, Vineyard et al. (2021) reported that ED consumes 26% less electricity than an equivalent nitrification-denitrification reactor but 64% more than an anammox reactor in terms of nitrogen removal/recovery from wastewater. In addition, wastewater with high electrical resistance may seriously influence the performance of the ions exchange membranes. Therefore, it is necessary to effectively control in-situ pH. The application of bipolar membranes in the ED process provides a convenient method of controlling the pH in each compartment.

2.2.1.2. MD. MD is a thermal driven process, in which the temperature difference between the feed side and permeate side drives the mitigation of vapour molecules to pass through the hydrophobic membrane or gas permeable membrane (GPM) (González-García et al., 2021; Guo et al., 2019). In the MD process, the volatile ammonia in the feed side can transfer through the membrane to the permeate stream under acidic conditions (Darestani et al., 2017; Huo et al., 2020), after which the condensed ammonium solution can be further used. It was reported that the temperature and pH of feed solution containing ammonium are the major parameters influencing the ammonia transfer and enrichment (Cerrillo et al., 2021c; He et al., 2018; Laureni et al., 2013; Wu et al., 2016a). Noriega-Hevia et al. (2020) developed a mathematical model to represent the time evolution of pH and nitrogen concentration during the recovery process and argued that the solution pH is the most important factor for enriching the nitrogen in the MD process. Munasinghe-Arachchige et al. (2021) reported the pH of the feed side in the GPM reactor above 9.26 is beneficial to the formation of  $NH_{3(g)}$  due to the pKa for NH3 at 9.26. Garcia-González and Vanotti (2015) found that increasing the solution pH by additional sodium hydroxide could increase the N recovery from 55% to 81% in the GPM reactor. Compared to typical acid absorption and ammonia stripping (23.6-49.6 kWh/kg·N), it was reported that lower energy requirements are needed in the GPM process for the nitrogen recovery (0.22-1.2 kWh/kg·N) while the addition of any alkali reagent is not required (Beckinghausen et al., 2020).

2.2.1.3. FO. The FO process depends on the difference in the chemical potential between the feed solution (i.e., high water chemical potential) and draw solution (low water chemical potential), which drives ions transport by the membrane from a low concentration solution (feed) to a higher one (draw) (Phuntsho et al., 2012). Generally, water in the feed solution can move to the draw solution through the FO membrane while the ammonium can be concentrated in the feed side with appropriate draw solute. It was reported that the aquaporin FO membrane can enrich almost 100% of NH<sub>4</sub>+-N (Engelhardt et al., 2020). Compared to other membrane techniques, the FO process is cost-effective and

Table 1
Summary of side-stream BMISs towards nitrogen recovery from wastewater.

Technology	Feed solution	Efficiency	Energy consumption	Final product	Reference
ED process	domestic anaerobic digester supernatant	$7100 \pm 300$ mg/L of NH <sub>4</sub> –N concentrated with concentration factor of 8	$3.8\pm1.2~\mathrm{kWh/kg\cdot N}$	Liquid ammonium	Ward et al. (2018)
BMED	residual waters	Concentration of TN increased from 1.5 to 7.3 g/L	5.3 kWh/kg·N	Liquid ammonium	Van Linden et al. (2020)
FCDI	Synthesized wastewater	80% of N recovered	20.4 and 7.8 kWh/kg·N for low-strength and high-strength wastewaters respectively	Liquid ammonium	Zhang et al. (2018b)
ED process	municipal wastewater	concentration ratio of 4.6	0.32 kWh/kg·NO <sub>3</sub>	Concentrated nitrate	Mohammadi et al. (2021)
MCDI	simulated wastewater	72% of NH <sub>4</sub> +-N recovered	3.22 kWh/kg·N	struvite	Gao et al. (2020)
ED- membrane stripping	real urine	93% of N recovered	8.5 kWh/kg·N	Volatile ammonia	Tarpeh et al. (2018)
ED-TMCS	Effluent of lab-scale AD reactor	83% of TAN recovered	9.7 kWh/kg·N	Concentrated ammonium	Rodrigues et al. (2021)
EDI	domestic wastewater	over 90% of N recovered	52.34 kWh/kg·N	Concentrated ammonium	Zheng et al. (2017)
ED	Urine	72% of N recovered	13.06 kWh/kg·N	ammonium bicarbonate	Jermakka et al. (2018)
IMD-AC	Urine	approximately 60% of ammonia recovered	2.2 kWh/kg·N	Concentrated ammonium	McCartney et al. (2020)
FCID-GPHFMC	dilute synthetic wastewater	60% of ammonia recovered	9.9–21.1 kWh/kg·N	$(NH_4)_2SO_4$	Zhang et al. (2018a)
BMED-HMFC	wastewater	65.2% of the ammonia capture ratio	0.76 kWh/kg·N	Volatile ammonia	Yan et al. (2018)
FO	Effluent from the treated wastewater	98% of N concentrated	0.015 kWh/kg·N	Liquid ammonium	Zou and He (2016)
GPMR	Swine manure	96.2% of TAN recovered	Methane yield of 105 mL $CH_4/g$ -TCOD	Concentrated ammonium	Molinuevo-Salces et al. (2018)

FCDI: Flow-electrode capacitive deionization; TN: total nitrogen; MCDI: membrane capacitive deionization; ED-TMCS: electrodialysis cell and a transmembrane chemisorption module; TAN: total ammonia nitrogen; BMED-HMFC: bipolar membrane electrodialysis-hollow fiber membrane contactor; IMD-AC: isothermal membrane distillation with acidic collector; FCID-GPHFMC: flow-electrode capacitive deionization unit combined with a hydrophobic gas-permeable hollow fiber membrane contactor.

**Table 2**Summary of submerged BMISs towards nitrogen recovery from wastewater.

Technology	Feed solution	Efficiency	Energy consumption	Final product	Reference
Scaled-up MEC	Urine	$59\pm31\%$ of TAN recovered	$1.36\pm0.28~\text{kWh/}$ kg·N	Struvite	Zamora et al. (2017)
MEC-FO	High-strength sidestream centrate	$99.7 \pm 13.0\%$ of AN	0.91 kWh/kg·NH <sub>4</sub> +- N;	Struvite	Zou et al. (2017)
Tubular Micro-Pilot MEC	liquid effluent of an anaerobic digestion process	$2.5\pm0.1$ g N/d of ammonium recovered the ammonium recovery	2.3 kWh/kg·N	Volatile ammonia	Cristiani et al. (2020)
MEC	Synthetic digestion effluent of livestock waste	$90.1 \pm 1.3\%$ of ammonia recovered	1.3 kW h/kg·N	$(NH_4)_2SO_4$	Qin et al. (2018)
MEC-chemical precipitation	digested sludge centrate	$53 \pm 5\%$ of TAN recovered	4.5 kWh/kg·N	Struvite	Barua et al. (2019)
BEM	Synthetic wastewater	$68.1 \pm 3.4\%$ of ammonia recovered	2.91 kWh/kg·N	$(NH_4)_2SO_4$	Zhang et al. (2021)
3-chamber BEC	reject water	$75.5 \pm 4.6\%$ of N recovered	6.1 to 8.2 kWh/kg·N	Concentrated ammonium	Koskue et al. (2021)
HRES	synthetic urine	58% of TAN recovered	6.5 kWh/kg·N	Concentrated ammonium	Kuntke et al. (2018)
BES	synthetic wastewater	7.1 g N/m <sup>2</sup> ·d of N recovered	5.7 kWh/kg·N	Volatile ammonia	Qin et al. (2017)
MPC-IE	Raw sewage	37.5% of NH <sub>4</sub> –N recovered	Recovery of 7.4 kWh/kg·N	Concentrated ammonium	Gong et al. (2017)
Gl-AnMBR	Synthetic sewage	95.5% of N recovered	4.5 L/d of CH <sub>4</sub> produced	Concentrated nitrogen	Prieto et al. (2013)
AnMBR-MPBR	Sewage	28% of N recovered	1 kWh/kg·N	Microalgal biomass	Seco et al. (2018)
MPBR	Effluent of AnMBR	51.7 $\pm$ 14.3 mg N/mol recovered	Recovery of 0.058 kWh/kg·N	Microalgal biomass	González-Camejo et al. (2019)

MEDC: microbial electrolysis desalination cell; AN: ammonia nitrogen; BEM: bioelectrochemical membrane; DCP-MFC: dual-membrane cylinder photo-microbial fuel cell; BEC: bioelectroconcentration cell; HRES: Hydrogen Gas Recycling Electrochemical System; MPC-IE: membrane-based pre-concentration combined with ion exchange process; Gl-AnMBR: gas-lift anaerobic membrane bioreactor.

environment-friendly (Van der Bruggen and Luis, 2015). This is despite the fact that the regeneration of draw solute from diluted stream needs additional power, which may be a barrier for commercializing the FO process. Chekli et al. (2016) indicated that it is significant to find an economically feasible hybrid process for regenerating diluted draw solution; for example, integrating FO with MD may be feasible to fulfill the

purpose because the MD process as a post-treatment can recover fresh water from the diluted draw solute of the FO process, which regenerates the draw solution and thus creates a circular economy (Liu et al., 2016; Ray et al., 2019; Volpin et al., 2019). In addition, the reverse salt flux (RSF) in FO processes ranges from 80 to 3000 mg/L (Hancock and Cath, 2009), but the structure of the membrane support layer and

concentration of draw solution have insignificant impacts on the RSF (Phillip et al., 2010). Overall, RSF can not only reduce the concentration of draw solution, but also contaminate the feed solution. However, magnesium-based draw solutes have been investigated to provide magnesium ions to the feed side through RDF, which boosts up the struvite precipitation and thus facilitates the nitrogen recovery in the feed solution (Volpin et al., 2018). Singh et al. (2019) utilized biomimetic aquaporin membranes (thin film composite FO membranes incorporating aquaporin proteins) to recover nitrogen from sewage while using divalent magnesium chloride as draw solution. They found that 66% of ammonia could be recovered within 24 h and regular cleaning enables restoration of membrane performance after every 24 h-cycle.

2.2.1.4. RO and NF. As high-pressure membrane processes, NF and RO have high rejection rates of salts and ions, which are often used to recover ammonium from urine (Adam et al., 2018; Patel et al., 2020; Ray et al., 2020). In the RO process, feed water is driven by osmotic pressure difference to flow from dilute to a concentrated solution through a semi-permeable membrane (Ahuchaogu et al., 2018). In this scenario, the process is driven by additional pressure greater than the osmotic pressure. Ray et al. (2020) reported that 64% and 90% of unionized ammonia can be recovered from hydrolyzed urine by RO and NF processes respectively. It was suggested that almost all the ammonium-nitrogen can be enriched by the RO membrane through solution pH optimization within the reactor because ammonium-nitrogen mainly exists in NH<sub>4</sub><sup>+</sup> form at pH < 7 (Vaneeckhaute et al., 2012). In the NF separation process, 1–10-nm molecule with pore size ranging from 1 to 5 nm can be rejected within the reactor (Shon et al., 2013). Compared to the RO process, lower pressure is applied in the NF process as well as lower energy input (Wafi et al., 2019). Pronk et al. (2006) revealed that the NF membrane could reject nearly all the micro pollutants including phosphate, propanol, ethinylestradiol, carbamazepine, diclofenac and ibuprofen, while nitrogen could permeate by the membrane for its enrichment. The NF process is always used to concentrate nitrogen from urine with 6 kWh/L (Maurer et al., 2006).

#### 2.2.2. Anaerobic membrane bioreactor

Anaerobic membrane bioreactor (AnMBR) has garnered increasing interest in addressing energy challenges in conventional wastewater treatment plants. This is achieved by integrating membrane separation with anaerobic digestion. Compared to aerobic MBR, AnMBR has the advantage of solids-free permeate rich in nutrients including nitrogen, biogas generation, low energy demand because there is no need for aeration, less sludge generation and high COD removal (Liu et al., 2019). This is despite the fact that other nutrients may be also enriched in the permeate solution. Grossman et al. (2021) applied the AnMBR for treating food processing wastewater, where 77% of nitrogen was recovered whilst 57% of total organic carbon was recovered in the form of methane. Simultaneously, the recovery of phosphorus (91%) was also observed. Moreover, the transportation of AnMBR permeate to agricultural areas is still a challenge in practical applications that needs to be solved. Furthermore, methane production in wastewater treatment can drive the operation of the AnMBR to offset input energy while applying the AnMBR in treating municipal wastewater. For instance, Liu et al. (2020b) found that biogas obtained in the AnMBR can produce electrical energy around 0.327 kWh/m<sup>3</sup> of municipal wastewater while treating 400 mg/L of COD.

#### 2.2.3. Osmotic membrane bioreactor

Osmotic membrane bioreactor (OMBR) is developed by placing the FO membrane inside the bioreactor, showing superior permeate quality, a higher pollutants rejection rate, lower fouling propensity, and low energy consumption due to the FO process. The nitrogen in the influent can be enriched in the feed side of the bioreactor and then the stream

containing rich ammonium and other nutrients achieves the nitrogen recovery in the form of struvite through pH elevation and additional necessary chemicals (Hou et al., 2017; Xie et al., 2014). 80% of ammonium can be recovered via struvite precipitation as reported by Qiu and Ting (2014), where NaOH was added to increase pH levels in the 8.0–9.5 range. Some studies utilized the microfiltration (MF) membrane and/or ultrafiltration (UF) membrane in the OMBR to extract the enriched ammonium and reduce the impacts of foreign substances, which works in parallel with the FO membrane (Holloway et al., 2015; Qiu et al., 2015), as illustrated in Fig. 2. To address the issue associated with the draw solution in the OMBR, RO and MD process can be employed to regenerate the draw solute and thus increase the recovery system's feasibility (Chang et al., 2017; Luo et al., 2016).

#### 2.2.4. Bio-electrochemical systems

Bio-electrochemical systems (BES) can recover ammonium with simultaneous wastewater purification and energy recovery, which mainly includes microbial electrochemical cell (MEC), microbial fuel cell (MFC) and microbial desalination cells (MDC) (Cerrillo et al., 2021b; Ye et al., 2018, 2021a). In BES, electrochemically active bacteria are used to convert chemical energy stored in organic matter to electrical energy (Arredondo et al., 2015). In a typical double-chamber BES, the anode chamber and cathode chamber are separated by a proton/cation-exchange membrane (P/CEM). At the anode chamber, the substrates such as organics are anaerobically degraded to produce hydrogen ions (H<sup>+</sup>) and electrons (e<sup>-</sup>), where electrons migrate to the anode surface and then transfer to the cathode electrode through an external circuit. Meanwhile, hydrogen ions pass through the P/CEM to the cathode chamber to react with electron acceptors (e.g., air) to form water (Logan et al., 2006). In MFCs, a thermodynamically favorable reaction takes place in the anode chamber for converting the chemical energy into electrical energy with the reduction of oxygen. In contrast, external electrical energy is required in MECs due to the presence of thermodynamically unfavorable reactions in the cathode chamber (Wu and Modin, 2013). Thus, the main difference between MFCs and MECs is favorability of the thermodynamic reaction taking place in spite of their architecture being virtually the same.

In MFCs, ammonium can be enriched in the cathode chamber due to its transfer from the anode through the CEM. The cathode reaction could generate hydroxyl ions (OH-) localized the cathode electrode, which provides a high pH zone for initiating the quick conversion of ammonium to volatile ammonia without the extra pH adjustment (Kuntke et al., 2011). The concentrated ammonia stream obtained from cathode could be further harvested through various methods, including struvite precipitation, adsorption and ammonia stripping. A decade ago, it was reported that the MFC can simultaneously recover ammonium and energy of up to 3.29 g/d m<sup>2</sup> and 3.46 kJ/g (Kuntke et al., 2011), indicating the positive energy-balance achieved in the ammonium-nitrogen recovery system. Compared to the MFC, MEC revealed better nitrogen recovery from wastewater because additional power applied in MEC favors the ammonium transfer from the anode chamber to cathode chamber. This process contributes to the ammonium enrichment in the cathode compartment (Chen et al., 2017; Haddadi et al., 2013; Hou et al., 2017; Ledezma et al., 2017).

In addition, MDC was used in wastewater treatment for concurrent resource recovery, desalination and wastewater purification, including landfill leachate and anaerobic digester liquor (Iskander et al., 2018; Liu et al., 2020a; Lu et al., 2020; Zhang and Angelidaki, 2015), where its two main configurations are designed for recovering ammonium. From Fig. 3a, it can be seen that CEM faces the cathode while AEM faces the anode in the first configuration. In this scenario, high-strength ammonium wastewater is desalinated in this chamber while ammonium ions mitigate across CEM, and are further accumulated in the cathode chamber. For example, Zhao et al. (2019) found that 69% of ammonium could be recovered in the catholyte, and 72% was removed in the desalination chamber while applying a tubular MDC for recovery of

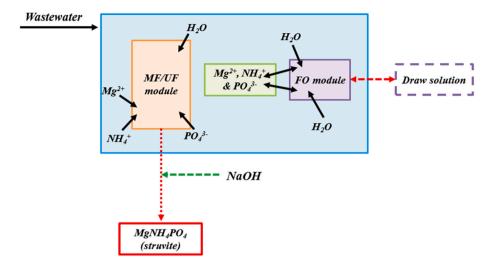


Fig. 2. Schematic of the osmotic membrane bioreactor (OMBR) integrated system for ammonium recovery. Adopted from Ye et al. (2021b).

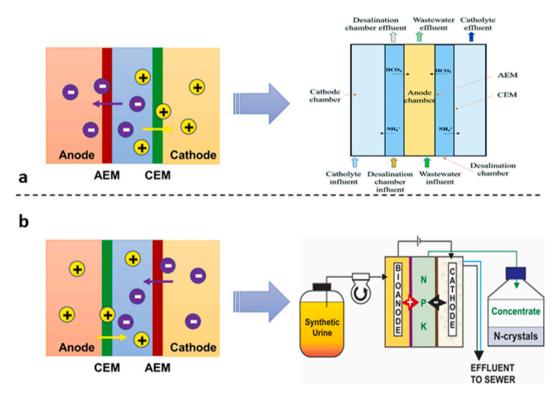


Fig. 3. Two designs of MDC for resource recovery. Adopted from Liu et al. (2020a).

ammonium from a diluted draw solution of the FO process. Apart from this, another configuration drives the anions in the catholyte and cations such as ammonium in the anolyte into the middle chamber through an electrical field. Consequently, both cation and anion nutrients, such as phosphate and ammonium can be recovered from wastewater (Ledezma et al., 2017).

Overall, the ammonium recovery using BES is affected by: (1) the equilibria among other ions at both anode and cathode; (2) the property of IEM; (3) the catholyte pH; (4) applied power; and (5) the ammonium concentration in wastewater (Arredondo et al., 2015, 2017; Mousavi et al., 2021; Qiu et al., 2020; Wan et al., 2010). BES offers a promising option for ammonium recovery from a concentrated stream, but Zhang and Liu (2021) argued that it is not unrealistic to recover ammonium in an energy and cost-effective manner applying BES from wastewater containing low levels of ammonium (40 mg/L). Thus, further

investigations on the engineering feasibility of BES should be conducted.

#### 2.2.5. Membrane photobioreactor

Currently, wastewater treatment, specifically ammonium is generally removed via catabolism (i.e., disassimilation) and microbial anabolism (i.e., assimilation), which is finally assimilated into biomass or converted into nitrogen gas (Rittmann and Mccarty, 2014). The circular economy framework triggers the application of phototrophs (e.g., phototrophic bacteria and microalgae) for concurrent wastewater treatment and nutrient recovery (Liu et al., 2018). Huelsen et al. (2016) reported that the  $\mathrm{NH_4}^+$ -N and COD are assimilated by microbial activity of purple phototrophic bacteria (PPB) in a certain ratio of 1 g·NH<sub>4</sub>+-N/16 g·SCOD, but the concentration of  $\mathrm{NH_4}^+$ -N and COD (40 and 400 mg/L, respectively) in typical sewage cannot satisfy the ratio, so additional carbon source is needed if the acceptable effluent concentrations for discharge

and reasonable nutrients assimilation are prioritized. Besides, since PPB can only use sugars, alcohols, organic acids, etc., as the substrates, the pretreatment of wastewater is also required. However, this inevitably increases the economic burden and limits the commercial application of PPB for the nitrogen recovery from sewage.

In contrast to the phototrophic bacteria, photoautotrophic microalgae can assimilate nitrogen without the extra carbon source, and carbon dioxide can be a carbon source for microalgae growth, which alleviates global warming. Besides, the microalgae biomass can be used to produce various products such as added-value biological derivatives, biopolyesters, antioxidants, dyes, pigments, proteins, sugars and lipids (Assunção et al., 2017; Assunção and Malcata, 2020). More importantly, microalgae biomass can also be applied in developing renewable energy and biofuels, including bioelectricity, biohydrogen, biobutanol, bioethanol and biodiesel (Show et al., 2017; Yap et al., 2021), while effective removal of toxic metals can be achieved through microalgae (Ahmed et al., 2022; Yan et al., 2022). In the photobioreactor (PBR), ammonium-nitrogen can be restored by microalgae as agricultural fertilizer (Tan et al., 2021). The performance of PBR is highly affected applying the ratio of working volume to surface area receiving sunlight when compared to other types of bioreactors (Zhang and Liu, 2021). The poor settleability of microalgae results in the difficult biosolid-liquid separation while cultivating the suspended microalgae at wastewater treatment. Moreover, the high HRT and large footprint also challenge the application of microalgae.

Therefore, membrane photobioreactors (MPBR) were widely investigated for achieving minimum footprint with easy biosolid-liquid separation (Ji et al., 2020). Integrating membrane technology with microalgae can reduce energy consumption and chemical usage with simple operation, easy to scale up, and excellent fractionation capability (Zhang et al., 2020). Chang et al. (2019) utilized a scalable membrane-based tubular photobioreactor (SMPBR) to realize high-efficiency nitrogen recovery (74.31%) from landfill leachate. Compared to the traditional PBR, the microalgal biomass concentration was improved to 2.13 g/L in SMPBR. Similarly, Nguyen et al. (2021) employed the nitrogen from urine to cultivate the microalgae in the MPBR, in which high recovery rate of nitrogen (TN of 90.5 mg/L·d) can be synergistically achieved with biomass production (biomass productivity of 313 mg/L·d) at biomass retention times (BRT) of 7 d.

## 2.3. Generation of bioproducts in nitrogen recovery towards circular bioeconomy

#### 2.3.1. Biofertilizer

Recent studies have shown that the application of conventional chemical fertilizer may have serious impacts on the environment, plants and soil. The possible reason for this is that the increase in soil erosion is associated with the use of synthetic fertilizer whilst the irrational use of nutrient-based fertilizer may reduce the yields of plants and crops, causing some environmental issues (Dineshkumar et al., 2018). Therefore, looking for resource-efficient and eco-friendlier biofertilizers as alternatives to the synthetic fertilizers contributes to more sustainable agricultural practices.

Struvite (MgNH<sub>4</sub>PO<sub>4</sub>·6H<sub>2</sub>O) is the most commonly achieved product in the nitrogen recovery system, which is also a micronutrient critical to the growth of plants and crops due to simultaneously containing nitrogen and phosphorus. Due to the property of struvite such as slow nutrient release, it has advantages over other fertilizers (Song et al., 2011). Since Mg is an essential element of chlorophyll, the existence of Mg also enhances the application prospect of struvite as an efficient fertilizer for grasses. Compared to the commercially available fertilizers, struvite obtained from nitrogen recovery system has low content of heavy metal ions (Latifian et al., 2012). As a result of this, struvite is relatively less harmful to the plant roots. Kern et al. (2008) argued that the loading rate of Cd in struvite obtained from wastewater is reduced by a factor of 10 when compared with triple superphosphate (derived from

natural phosphate deposits). A comparison of fertilizer efficiency between the chemical fertilizer and struvite was conducted by Rasul et al. (2011). They detected no significant difference between the dry matter yield, leaf area and plant height while applying these two fertilizers to the maize crop.

In contrast, some research indicated that lower crop yield of plants and crops treated by struvite fertilizer was observed when compared to chemical fertilizers (Ganrot et al., 2007). The optimal plant growth may require substantial input of struvite to satisfy the plant nitrogen demand because the ratio of N/P in struvite indicates that the plant-available nitrogen is not enough (Beckinghausen et al., 2020). However, an increase in the struvite application may increase the soil pH which is detrimental to the nutrient uptake and availability, and causes the accumulation of P and Mg in soil (Kataki et al., 2016a, 2016b). In soil, the amount of nutrients input should follow the order N > K > P > Mg, so a combination of struvite and other fertilizers may satisfy the requirement to provide sufficient nitrogen content for the plant growth (Latifian et al., 2012). Apart from this, struvite is more technically feasible for application in acidic soils since the solubility of struvite can be improved, thus enhancing the fertilizer efficiency (Cabeza et al., 2011). Overall, struvite can serve as an intermediary solution between global nutrient transfer and on-farm nutrient recycling (Rahman et al., 2014). The expensive production of industrial ammonium can be overcome through connecting all the industrial and municipal wastewater with wastewater treatment plants for mass nitrogen recovery.

In addition, ammonium sulfate can comparatively yield 10% because it can lower the soil pH and increase leaf nitrogen. This is despite the fact that the application of ammonium sulfate may increase soil electrical conductivity and negatively affect the yield. Similarly, Vargas and Bryla (2015) found that ammonium sulfate-based fertilizers are more efficient for the growth and yield of highbush blueberry plant in the first five years compared to the granular application of either nitrogen source. Chien et al. (2011) indicated that the addition of ammonium sulfate provides sulfur to the soil which is lacking in many other proposed fertilizer alternatives. Nitrogen can also be recovered by adsorption via biochar, where the application of nitrogen-loaded biochar can enhance crop yield in agriculture as a slow-releasing fertilizer due to the presence of an aromatic group (Pathy et al., 2021). For instance, the pumpkin yield can be increased four-fold while applying low-dose urine impregnated biochar to the pumpkin field (Schmidt et al., 2015). Wu et al. (2016b) also found the microbial dynamics of soil can be improved by adding biochar. The application of nitrogen-loaded biochar can also sequester carbon in the applied soil, reduce nitrogen loss and improve soil productivity (Yao et al., 2013). Apart from this, the soil's pH can be improved as well as its cation exchange capacity while utilizing the nitrogen-based biochar obtained from the urine, which also strongly suggests the soil conditioner potential of biochar (Bai et al., 2018). In one study, polymer matrix biochar composite was exploited to recover ammonium from urine, and the resulting nitrogen-loaded biochar was applied to the cotton plant (Wen et al., 2017). In this scenario, adding nitrogen-loaded biochar has various positive impacts on cotton plants, including enhanced nitrogen use efficiency and less N leaching. When Mg modified biochar was utilized to recover ammonium from urine, the nitrogen-laden biochar can enhance the plant height of ryegrass and maize (Xu et al., 2018). In contrast, Mg biochar does not exert any positive effect on ryegrass and maize.

Microalgae-based fertilizer can promote microorganisms demand for: plants growth; solubilize phosphate and fix nitrogen through providing proteins; vitamins required for enhancing plant growth; carbohydrates; natural enzymes; polyamines; growth regulators; antifungal substances; amino acids; carotenoids; phytohormones and micro- and macro-nutrients (Patil et al., 2008; Ronga et al., 2019). The plants' physiological processes can be improved and regulated while applying the microalgae-based fertilizer at low doses in agriculture (Morais Junior et al., 2020). Such fertilizers can increase the shelf life, productivity and quality of the plants and crops, improving the tolerance to abiotic

stresses and nutrient absorption (Ronga et al., 2019). Due to an increase in carotenoid content and sugar, the application of microalgae-based fertilizer in tomato cultivars can improve the quality of the fruit (Coppens et al., 2016). In this scenario, the tomatoes processed by microalgae-based fertilizer can contain 44% more carotenoids than the fruit treated by organic fertilizer and 70% more carotenoids than those treated by inorganic fertilizer. Furthermore, it was reported that water extract of *Chlorella ellipsoida* and *Spirulina maxima* can: firstly, improve wheat tolerance for salinity; and secondly, enhance the antioxidant capacity and protein content of the whole grains produced by treating plants with microalgal extracts (El-Baky et al., 2010).

#### 2.3.2. Microalgae biomass

In most current studies, microalgae production is achieved by photoautotrophic cultivation, through which large-scale biomass can be obtained with the use of sunlight that is a free, renewable, and is a clean source of energy (Lam et al., 2012). Currently, MPBRs are investigated for assessing nitrogen recovery and biomass production from different wastewater sources, including agricultural effluent, slurry wastewater and sewage (Gao et al., 2016). Microalgae biomass can be used as raw materials in the feed, and food industry because of its high health and nutritional value (Morais Junior et al., 2020). More specifically, it can provide pigments such as chlorophylls and carotenoids, minerals, monoand n-3 polyunsaturated fatty acids, polysaccharides, essential amino acids and vitamins (Priyadarshani and Rath, 2012). Vermaas (2004) reported that microalgae biomass can be employed as supplements for feed through several nutritional and toxicological tests. In poultry feed, microalgae biomass can supplement conventional proteins to improve the yellow color of egg yolk and broiler skin (Becker, 2003). Further research should consider different biochemical properties of various microalgae species while applying them as supplements in novel animal diets (Batista et al., 2013).

Due to containing bioactive compounds such as sterols, carotenoids and polysaccharides, microalgae can reduce cholesterol and have various health benefits, for instance anti-cancer properties and other features (Lordan et al., 2011; Raja et al., 2018). Microalgae or its extracts can be also used as medicine against both gram-negative and gram-positive bacteria, and bacterial infections in fish or shrimp feed due to containing long-chain polyunsaturated fatty acids (LC-PUFAs) (Desbois et al., 2009; Shah et al., 2018; Yaakob et al., 2014). Furthermore the microalgae extracts can be widely employed in the cosmetics market for many products including sun protection, hair protection, peelers, emollients, regenerating products, refreshing products and anti-aging creams, and especially for Arthrospira and Chlorella (Morais Junior et al., 2020). Extracts from microalgae species not only can be antimicrobial, but also have potential for anti-aging skincare products, which contributes to their wide-ranging usage in cosmetics-related areas (Mourelle et al., 2017).

Microalgae are also employed to accomplish nitrogen recovery coupled with CO<sub>2</sub> sequestration at wastewater treatment (Molazadeh et al., 2019; Razzak et al., 2017). The capacity of microalgae for CO2 sequestration varies from species; for example, the carbon uptake rate of cyanobacteria A. microcopia Nageli is 28 mg/L min, which is higher than that of diatom P. tricornutum at 1.5 mg/L· min (Francisco et al., 2010). Through investigating the microalgae application in an ethanol synthesis factory, it can be seen that microalgae are feasible for sequestrating CO<sub>2</sub> for its growth in industry (Rosenberg et al., 2011). This finding indicates that microalgae can be utilized capturing CO<sub>2</sub> to save the costs associated with the chemical removal process (Kumar et al., 2010). It has been reported that Chlorella strains from hot springs can be tolerant to CO<sub>2</sub> concentration up to 40% (v/v) (Rizwan et al., 2018). In another study, 50%  $SO_2$ , 70% of NO and 60% of  $CO_2$  can be removed from flue gas by Chlorella sp. cultures, which mitigates greenhouse gases (Chiu et al., 2011). Microalgae biomass was also explored as a material for the production of biofuels to generate clean and sustainable energy (Pienkos and Darzins, 2009). Other potential applications of microalgae

biomass have been explored, including nutraceutics, biofertilizers and bioplastics (Borowitzka, 2013; Lu et al., 2016b; Sathasivam et al., 2019a). Table 3 summarizes the applications of microalgae biomass.

#### 2.3.3. Bioenergy

Conventionally, fossil fuels are combusted to produce energy, but this process will generate carbon dioxide as well, which contributes to global warming. Given the dual challenges of limited fossil fuel sources and increasingly serious global warming, it is of great emergency to look for sustainable and clean energy sources to replace fossil fuels. Biological or natural sources can be converted into renewable, clean and sustainable energy (i.e., bioenergy), including flora, fauna and their byproducts. More recently, bio-energy is considered as an alternative energy source, and a solution for remediating greenhouse gas emissions (Hariz and Takriff, 2017).

Raheem et al. (2018) reported that microalgae biomass can be converted into biofuels such as bioethanol and biodiesel, which has significant effects on global warming and environmental pollution (Khan et al., 2018). Compared to most terrestrial plants, microalgae can effectively convert energy into chemical energy 5-fold due to its simple structure (Yap et al., 2021). More importantly, the microalgae can reach fast growth rates, and some species of microalgae can double their biomass in just a few hours because their growth rate is 5-10 times quicker than traditional food crops (Okoro et al., 2019; Rodionova et al., 2017). Furthermore, there is no need to conduct any modifications of existing fuel engines because microalgae-based biofuels are compatible with these (Ras et al., 2013). Compared to the common oil crop, the lipid productivity of microalgae biomass is 15-300 times larger (Morais Junior et al., 2020; Zullaikah et al., 2019), so it is feasible to supplement the vegetable oils obtained from terrestrial crops. For example, the oil yield of microalgae per hectare is higher than that of rapeseed, soybean and palm oil (Paniagua-Michel et al., 2015). More importantly, the biofuels productivity of microalgae biomass is 10-20 times higher than any other biofuel crop (Mata et al., 2010; Ndimba et al., 2013). One species, Dunaniella tertiolecta, can produce 25.8% of bio-oil yield by hydrothermal liquefaction of microalgal biomass residues and methylation of different fatty acids (Shuping et al., 2010; Tang et al., 2011).

In anaerobic digestion, Duanliella salina can produce biogas because of its easy processing, low cost for pre-treatments, and high biomass productivity (J. Nayeong et al., 2012). In their study, Markou et al. (2013) utilized a medium with phosphorus limitation to cultivate Arthrospira platensis to achieve carbohydrate-enriched biomass. Then the biomass was employed as a substrate to produce bioethanol through with bioethanol productivity at around fermentation g·ethanol/g·biomass. Furthermore, the microalgae biomass can be cultivated by wastewater in non-arable land, which contributes to sustainable wastewater reuse and management, and results in no competition with food crops for farming land (Brennan and Owende, 2010; Christenson and Sims, 2011). Nevertheless, microalgae applications as a bioenergy source are still subjected to the dual challenges of harvesting and management of microalgae from large volumes, and high biomass production (Brennan and Owende, 2010).

Bio-membrane integrated systems can also recover energy in the form of biogas and electricity generation, where biogas production depends on the activity of methanogens in anaerobic conditions whilst the electricity generation occurs in the BES-based integrated systems. Biogas is considered as an alternative to fossil fuels, and it can be recovered from organics through anaerobic biological processes from wastewater treatment (Li et al., 2019). Maaz et al. (2019) reported that the recovered biogas comprises nitrogen (0–15%), hydrogen gas (0–5%), carbon dioxide (3–15%) and methane (50–90%). For the AnMBR, the methane yield is negatively affected due to it being exposed to high salinity levels at the beginning of the operation. This scenario is caused by the reversal of solute leakage (Gu et al., 2015). However, the methanogens may gradually adapt to built-up high salinity environments, and the produced methane can be recovered (Gu et al., 2015).

**Table 3**Summary of microalgae biomass applications.

Applications	Microalgae	Component	Application	Reference
Cosmetics	Dunaliella Salina	Phenols such asferulic acid and caffeic     Glycerol	Smoothen and Moisturize skin	Goiris et al. (2012); Safafar et al. (2015); Ariede et al. (2017); and Sathasiyam et al. (2019b)
Pigments	Chlorella zofingiensis	<ul><li>α-carotene</li><li>β-carotene</li></ul>	Pigmenter for salmon	Sathasivam et al. (2019b); Gouveia and Empis (2003)
Biofuel	Botryococcus braunii	<ul> <li>high lipid content containing 10.54% of polyunsaturated fatty acids 9.95% of saturated fatty acids and 79.61% of monounsaturated fatty acids</li> </ul>	biodiesel production	Tasić et al. (2016) and Caetano et al. (2020)
Bioplastics	Chlorella sp.	<ul> <li>high carbohydrates content (more than 60% dw)</li> </ul>	poly-lactic acid	Wang et al. (2014) and Aikawa et al. (2012)
Biofertilizer	Scenedesmus sp. Artrhospira platensis	higher concentration of abscisic acid, salicylic acid, auxins, gibberellins and cytokinins	<ul> <li>improve the plant nutrient status</li> <li>enhance the number of flowers per plant and root dry matter</li> <li>accelerate plant development</li> </ul>	Plaza et al. (2018)
food industry	Dunaliella salina	<ul> <li>Carotenoid β-content</li> <li>High levels of antioxidant</li> <li>Carbohydrate (32% dw)</li> <li>Protein (57% dw)</li> </ul>	Prevent the intracellular oxidative stress     Coloring beverages     Coloring margarine water-soluble     Animal feed     Human health dietary supplements such as tablets	Mokady et al. (1989); Ye et al. (2008); Mobin and Alam (2017); and Raja et al. (2018); García-González et al. (2005)
food industry	Arthrospira platensis	<ul><li>Carbohydrate (13–16% dw)</li><li>Protein (60–71% dw)</li></ul>	Increase the number of lactic acid bacteria     Produce low cholesterol eggs     Vegan replacement	Mobin and Alam (2017); Niccolai et al. (2019); and Raja et al. (2008); Becker (2007)
Medicine	Arthrospira platensis	<ul> <li>Essential polyunsaturated fatty acids (PUFA)</li> <li>β-carotene</li> <li>Vitamin K and B<sub>12</sub> and B1</li> <li>Provitamin A</li> <li>Isoleucine valine</li> <li>γ -Linolenic acid (GLA)</li> <li>Phycocyanin (colorant)</li> </ul>	Prevent rheumatoid arthritis, diabetes, multiple sclerosis, dermatitis, viral infections and schizophrenia Immune system enhancement Anti-inflammatory Antioxidant Anticancer	Sathasivam et al. (2019b); Vermaas (2004); Spolaore et al. (2006); and
	Chlorella zofingiensis	<ul> <li>Glucan</li> <li>β-carotene</li> <li>Astaxanthin</li> </ul>	Lowering lipids in the blood     Stimulates immune response     Lower blood sugar levels     Increase hemoglobin concentrations	Lu et al. (2016a); Spolaore et al. (2006); Ambati et al. (2014); Régnier et al. (2015); Paniagua-Michel et al. (2015); Varfolomeev and Wasserman (2011)

Taking anaerobic OMBR (AnOMBR) as an example, Yang et al. (2021) reported that the methane produced in the AnOMBR ranges from 60 to 301 mL/g·COD, which is influenced through the conductivity of the feed solution in the range of 2.5–20 mS/cm. Moreover, the AnOMBR could achieve higher methane yield at lower salinity accumulation. The change in the microbial function and composition may result in varying methane production in AnOMBR, which is attributed to RSF (Li et al., 2017). Therefore, it is important to control the salinity level in the OMBR while using an inorganic ionic substance that serves as a draw solute (Ansari et al., 2015). In this scenario, the environment which is beneficial for the growth of anaerobic bacteria can be created, and the efficiency of pollutant removal, and stable methane production can subsequently be maintained. Integrating pressure-driven membrane processes such as MF and UF or BES with AnOMBR may be a possible solution to improve the biogas production through mitigating salinity accumulation; for example, Wang et al. (2017a) found that the salinity level can be effectively kept in a stable range of 2.5-4.0 mS/cm while integrating AnOMBR with the MF membrane (AnOMBR-MF). Compared to the conventional AnOMBR, the AnOMBR-MF system can achieve higher methane production with 280 mL/gCOD during long-term continuous operation. Besides, the practical methane production is 28-39% lower than the theoretical value in AnOMBR-based system due to methane loss. Currently, research on the biogas production by lab-scale AnOMBR is too limited to provide effective analysis (Wang et al., 2016). For this reason, further study should focus on the scaling up of the AnOMBR for the methane production and collection.

The above discussions demonstrate that BES-based systems can generate electricity during the nitrogen recovery from wastewater, including landfill leachate, domestic wastewater and industrial

wastewater. However, the electricity produced by microbes is subjected to low power densities achieved at high reactor volumes (Penteado et al., 2018). For example, approximately 150 W/m³ of power density were obtained in MFC reactor volumes of 1.2 mL, but increasing the volume to 100 mL resulted in power density falling to 50 W/m³ while cultivating bacteria *Shewanella oneidensis* DP-10 at the anode chamber (Biffinger et al., 2007). The electricity generated in the MFC is affected by various factors including the type of wastewater, biofilm formation, membranes, mediators, substrates, the configuration of electrodes, electrolyte temperature, electrolyte pH and MFC design (Apollon et al., 2021).

The dual-compartment MFCs are always used for the nitrogen recovery at wastewater treatment, where their designs mainly include tubular, miniature, flat-plat type, cube-type and H-type MFCs (Gul et al., 2021; Song et al., 2015). While employing glucose as an organic substrate, the power density of cube type and flat plate MFCs were 910 and 212 mW/m<sup>2</sup>, respectively, which is higher than that of H-type MFC (115.6 mW/m<sup>2</sup>) (Min and Logan, 2004; Yang et al., 2017; You et al., 2006). This may be attributed to the lower membrane surface areas for proton transfer in H-type MFCs (Logan et al., 2006). Furthermore, the organic substrates used for microbial growth also affect the MFC performance; for example, the wastewater generated by processing starch provided the power density of 239.4 mW/m<sup>2</sup> (Lu et al., 2009) whilst municipal wastewater treated by MFC presented power density of 52 mW/m<sup>2</sup> (Zhang et al., 2011). Gonzalez del Campo et al. (2013) argued that raising the temperature from 20 to 40 °C can result in expanding power density from 0.73 to 1.01 mW/m<sup>2</sup>. Various investigations were conducted to enhance the efficiency of bioenergy generation in MFC reactors, but the commercialization of MFC still needs significant

refinements to overcome serious practical issues.

#### 3. Challenges and future perspectives

Wastewater as a gradually accepted alternative and renewable source of fresh water and nutrients is due to its existence in extremely large volumes. However, conventional wastewater treatment aims to remove the so-called pollutants, purify the wastewater, but current wastewater treatment prefers to recover renewable resources. In particular, current wastewater treatments generally convert ammonium to valueless nitrogen gas at rather economic and environmental costs. At the same time they simultaneously generate a substantial amount of GHGs which is typical of the linear economy (i.e., take-consumedispose). The primary goal of conventional wastewater treatment is to satisfy the effluent discharge standards, but may result in wasting large amounts of valuable resources which should be recovered, and then reused. Consequently, the spirit of a circular economy departs from the circular bio-economy, and this should be a major driver in future technological developments that benefit society, especially given the challenges of energy, food security, resources depletion and climate change.

In contrast to the linear economic model, circular bio-economy aims to improve the valorization and reuse of residues and organic wastes, which converts bio-waste and their intermediate products into bioenergy, biomaterials, and decouples growth from the consumption of finite natural resources (Mohan et al., 2016). For this reason, it is essential to shift the paradigm for biological wastewater treatment processes from removing to recovering and thereby reusing. It marks an essential step towards greater environmental and economic sustainability. The recovery of nitrogen from wastewater can reduce our dependence on chemical fertilizers, produce bioproducts with economic value, decrease eutrophication problems, and satisfy the effluent nitrogen concentrations required by government legislation. Due to the relatively low ammonium-N concentrations in some wastewaters, the ammonium recovery by current technologies is environmentally unsustainable, and economically costly. To address this issue, the solution to tackle the challenge of concurrent wastewater treatment, and ammonium should rely on BMISs. The use of BMISs for nitrogen recovery can help produce effective biofertilizers, high-value biomass and generate energy. The ammonia–nitrogen recovery from wastewater and its use are shown in Fig. 4.

Researchers should tailor the nitrogen recovery technologies to fulfill the requirement of local soil through cultivating a relationship with local farmers. This would lead to a localized solution for waste to fertilizer conversion. Besides, the nitrogen-based fertilizers derived from the recovery system need to present high efficiency, or, achieve the same if not better performance than the current mineral options. Simultaneously, they should not pose contamination risks to the crops. The cost and benefit analysis for the nitrogen recovery is essential to compare these different processes, including the cost of electricity, chemicals, and the market prices of the recovered products. Furthermore, the nitrogen recovery technologies should also gain the interest of wastewater treatment plants for their full-scale implementation. In this scenario, such technology should compete with current nitrogen removal techniques to meet effluent standards.

#### 3.1. Economic analysis

The cost-effectiveness of nitrogen recovery is a key incentive driving the development of BMISs on a commercial scale. For this reason, the techno-economic analysis involving energy consumption, reaction efficiencies, and operating parameters are important for evaluating the applicability of one technology for recovering nitrogen from wastewater. Besides, the production of ammonium through BMISs must be more economical than the conventional ammonia synthesis process, so the energy consumption per unit of nitrogen recovery was the most crucial factor. Although some studies have conducted such analyses (Mohammadi et al., 2021; Qin et al., 2017; Ward et al., 2018; Zou et al., 2017), most of these values used for the current calculation are achieved in lab-scale reactors for short-term experiments. Hence, the commercial application of BMISs is still questionable, including lack of solid outcomes in pilot-scale operations of BMISs, uncertain durability, and costly component materials (e.g., membrane and electrode). Long-term

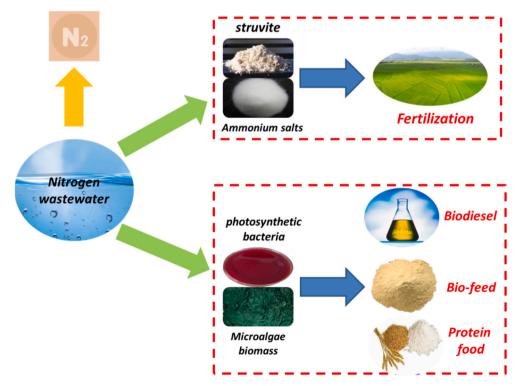


Fig. 4. Ammonia-nitrogen recovery from wastewater and its use.

operation of large-scale BMISs in real wastewater environments is currently rarely investigated and evaluated, so the current techno-economic analysis's reliability is in doubt. For this reason, it is necessary to evaluate the performances and challenges of large-scale systems for long-term operations.

#### 3.2. Technical analysis

The bio-membrane integrated systems involve microbial activity, so there are many additional problems in systems, such as, undesired substrate consumption via competing metabolic processes, unwanted biomass growth, and incomplete biodegradation of substrates, which would curtail amount of nitrogen recovered in the system (Pandey et al., 2016). Ahmed et al. (2019) believed that a combination of technologies may be a solution to maximize the efficiencies of nitrogen recovery in BMISs. Besides, scaling of the membrane is required in order to advance application of membrane-based techniques. Membrane fouling may not only contaminate permeate, but also deteriorate the membrane's efficiency over long-term operation of membrane-based processes. Another concern about the membrane-based treatment process is the cost of membranes. Further studies should focus on the advanced development of membranes to improve their properties and reduce the synthesis costs.

Compared to other membrane separation technologies, OMBR integrated system presents its unique advantages of low energy input and membrane fouling potential (Yang et al., 2021). Nevertheless, the recovery of draw solute in the FO process still hinders the commercial application of OMBR integrated systems (Chekli et al., 2012). Conventional draw solute recovery processes including ED, MD, and RO were investigated, however, these methods require high energy consumption, and thus increase the overall outlay. For instance, high energy consumption of  $1.88-4.01 \text{ kWh/m}^3$  (i.e., an operation cost of  $0.2-0.4 \text{ s/m}^3$ ) is required to recover salt from the draw solution (Ortiz et al., 2008). Similarly, the use of a MD process can successfully recover water from the draw solute, in which additional energy input of 29 kWh/m<sup>3</sup> (i.e., an operation cost of 2.9 \$/m<sup>3</sup>) is required to drive the MD operation (Zhao et al., 2014). This is despite the fact that solar energy and power plants can provide energy sources to reduce energy consumption in the MD process. Therefore, further efforts should be made to devise effective and economic recovery of draw solute with possible use of renewable energy in the OMBR integrated systems.

Due to its ability to be integrated with the currently existing wastewater treatment system, for the purposes of alleviating energy demand, and pollution problems, BES is considered as a platform for the conversion of waste into energy and chemicals. The rapid degradation of organic wastes in the anode chamber would allow for the prospect of their commercial application (Kadier et al., 2016; Sadhukhan et al., 2016; Santoro et al., 2017). These advantages make BESs a key part of the promising circular bio-economy model. The internal resistance is an important parameter affecting the BES performance (Arredondo et al., 2015). At the anode chamber of BESs, the microbial environments need effective controls to improve their performance. The possible reason for this is that the electricity generation in BES may be detrimentally affected by the deactivation undesired side reactions of anode (Tota--Maharaj and Paul, 2015; Zhang et al., 2013). More specifically, accumulation of organic wastes on the anode surface, consumption of organics and electrons for undesired side reactions, and interruption of bio-electrochemical reactions due to the presence of impurities may compromise the power density of BESs (Fornero et al., 2010; Rabaey et al., 2005; Zhang et al., 2013). Therefore, a pretreatment process is essential. This is despite the fact that additional processes will increase the costs, so more economic analysis is required here. In addition, the selection of materials including membrane and electrodes are significant for BES commercialization. For example, bio-electrochemical efficiency of cathodic reactions is influenced by the cathode materials and the production rate of chemicals (Shahgaldi and Hamelin, 2015; Sonawane

et al., 2013). Pt-supported cathode electrodes are traditionally used at the cathode chamber of BESs (Wang et al., 2017b), but their high costs (>\$1500/g, Sigma-Aldrich) are a barrier to commercialization. This is despite the fact that the application of Pt-based electrodes improves the oxidation-reduction potential. Apart from this, the use of expensive and complicated membranes (e.g., Nafion®, >\$1500/m², Sigma-Aldrich) results in costly BESs. Several issues may occur to negatively affect the performance of the membrane, such as fluctuating wastewater treatment conditions, contamination and accumulation of microbes, and gas leakage between two chambers (Xu et al., 2012). To successfully commercialize the BESs, it is of great importance to develop membranes and electrodes that are cheaper and more efficient compared to the currently existing commercial ones.

The nitrogen recovery through MPBR integrated systems is affected by photosynthetic efficiency, dissolved gases, solution pH, mixing, temperature and irradiance (Anto et al., 2020). The biggest challenge of MPBR integrated systems is the availability of sunlight in the case of light irradiance, so it is recommended to use an acrylics type material to realize the cost-effectiveness of the establishment. It is also essential to prevent the microalgae accumulating on the walls of the tubes, as this may undermine sunlight transmittance. Microalgae can continuously produce biomass under simulated illumination, even during night hours. Another important factor affecting the performance of MPBR integrated systems is nutrient solubility, so effective pH control is significant. Consequently, sparger or the bubblier kind of arrangement is always used to control gaseous CO2 which may result in the generation of HCO<sub>3</sub> and subsequently affecting the solution pH. The development of a MPBR integrated system for processing, harvesting and cultivating microalgae biomass is vital to enhance the system's sustainability, and make the system more economically viable.

#### 4. Conclusion

It is imperative to recover nitrogen from wastewater given the fact that nitrogen in wastewater is becoming a serious and worldwide environmental problem, and there is a high global demand for nitrogenous fertilizers. The recovered nitrogen can generate some revenue by providing supplementary nitrogen as a source for fertilizer production and industrial activity. This review paper assessed nitrogen recovery, and circular bio-economy options from wastewater via BMISs. All the technologies discussed in this paper were able to recover a high percentage of ammonium from wastewater, where the recovered products can be used as bio-fertilizer, biomass and bioenergy to help society. Of all the approaches currently available for ammonium recovery from wastewater, the BES integrated, MPBR and OMBR-based processes have a great potential for concurrent recovery of nitrogen and energy as well as their combinations, which would transform wastewater management from a linear into a circular economy model.

#### Author contribution statement

Yuanyao Ye: Investigation, Writing – original draft, Methodology, Formal analysis, Data curation, Huu Hao Ngo: Supervision, Investigation, Project administration, Conceptualization, review & editing, Wenshan Guo: Investigation, review & editing, Soon Woong Chang: Investigation, Project administration, review & editing. Dinh Duc Nguyen: Methodology, Formal analysis, Resources, review, Sunita Varjani: Methodology, Resources, review, Qiang Liu: Investigation, Conceptualization, review & editing. Xuan Thanh Bui: Methodology, Validation and review. Ngoc Bich Hoang: Methodology, Data curation, review

#### **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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