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The definitive publisher version is available online at [10.1016/j.chemosphere.2021.133175](https://doi.org/10.1016/j.chemosphere.2021.133175)

1 **Bio-membrane integrated systems for nitrogen recovery from wastewater in**
2 **circular bioeconomy**

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37 **Abstract**

38 Wastewater contains a significant amount of recoverable nitrogen. Hence, the
39 recovery of nitrogen from wastewater can provide an option for generating some
40 revenue by applying the captured nitrogen to producing bio-products, in order to
41 minimize dangerous or environmental pollution consequences. The circular bio-
42 economy can achieve greater environmental and economic sustainability through
43 game-changing technological developments that will improve municipal wastewater
44 management, where simultaneous nitrogen and energy recovery are required. Over the
45 last decade, substantial efforts were undertaken concerning the recovery of nitrogen
46 from wastewater. For example, bio-membrane integrated system (BMIS) which
47 integrates biological process and membrane technology, has attracted considerable
48 attention for recovering nitrogen from wastewater. In this review, current research on
49 nitrogen recovery using the BMIS are compiled whilst the technologies are compared
50 regarding their energy requirement, efficiencies, advantages and disadvantages.
51 Moreover, the bio-products achieved in the nitrogen recovery system processes are
52 summarized in this paper, and the directions for future research are suggested. Future
53 research should consider the quality of recovered nitrogenous products, long-term
54 performance of BMIS and economic feasibility of large-scale reactors. Nitrogen
55 recovery should be addressed under the framework of a circular bio-economy.

56 **Keywords:** Bio-membrane integrated system; Ammonium recovery; Nitrogen
57 recovery; Circular bio-economy; Energy requirement.

58

59 **1. Introduction**

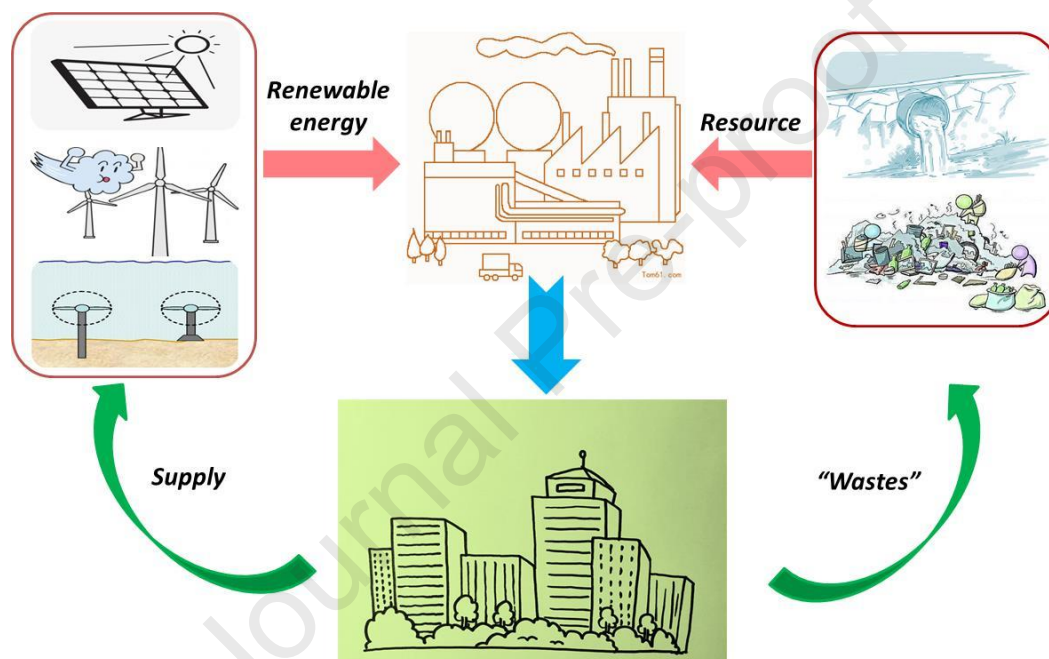
60 **1.1. Principal concepts of the circular bioeconomy**

61 Within the framework of a linear economy, finite natural resources are utilized to
62 generate valuable products, goods and services, energy and chemicals. This may
63 consequently cause a substantial generation of industrial and residential wastes, which
64 will be discarded into landfill or wastewater and simply adds the level of pollution.

65 Thus, treatment risks are increased and waste disposal processes and industrial
66 activities can also result in the production of greenhouse gases (GHGs), which
67 triggers worse global warming. Unlike the linear manner of a fossil fuel-based
68 economy, circular economy aims to recycle and reuse wastes and raw materials in a
69 closed-looped strategy with regenerative and restorative design properties. This in
70 turn contributes to eliminating toxic generation, as shown in Fig. 1.

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

84 produced (Mesdaghinia et al., 2015). Previously, wastewater was considered as a
 85 potential issue and waste in society, but the concept has shifted due to it being
 86 considered as an abundant source of energy and chemicals. Thus, the recovery and
 87 reuse of resources obtained from wastewater in other industries (e.g., energy and
 88 agriculture sectors) will benefit and support economic growth, especially given the
 89 final product will be one of higher quality and have negligible effects on soil, water
 90 and air.



91
 92 **Figure 1.** Concept of circular bio-economy.

93

94 **1.2. Importance of nitrogen recovery in circular bioeconomy**

95 Nitrogen (N) plays an important role in fertilizer production and food security. The
 96 explosive increase in global population has resulted in higher demands for nitrogen-
 97 based fertilizers (Vineyard et al., 2021; Wei et al., 2018). However, the production of
 98 nitrogen-based fertilizers through the Haber-Bosch process occupies ~1.6% of global
 99 CO₂ emissions, and 1–2% of global energy consumption (Groenestein et al., 2019;
 100 Hou et al., 2018). Therefore, it is essential to look for alternative nitrogen sources to

101 sustainably ensure the supply of green fertilizer, especially given the dual challenges
102 of supply security and climate change. Apart from this, most agricultural nitrogen is
103 lost to the aquatic environment and air, whilst only 17% is consumed by humans via
104 livestock or crops (Beckinghausen et al., 2020). The accumulation of nitrogen in
105 water bodies may result in eutrophication, endangering aquatic environments and
106 ecosystems. Current full-scale techniques to remove nitrogen from wastewater include
107 partial nitrification-anammox processes, nitrification-denitrification, and their
108 combinations. Through these processes complex nitrogenous compounds are finally
109 decomposed by bacteria into atmospheric nitrogen gas (N_2). Nonetheless, advanced
110 nitrogen removal technologies are still challenged by energy consumption, in which
111 the energy input for the removal of nutrients (i.e., nitrogen and phosphorus) ranges
112 from 0.39 to 3.74 kWh/m³ (Gu et al., 2017). Simultaneously, GHGs such as nitrous
113 oxide (N_2O) and methane (CH_4) are generated in the removal process (Law et al.,
114 2011).

115
116 Overall, the process related to nitrogen production and removal is energy-intensive, in
117 which the Haber Bosch process transforms the atmospheric nitrogen to ammonia, and
118 then the nitrogen removal process produces atmospheric nitrogen from ammonia.
119 Some researchers have found that nitrogen recovery in wastewater treatments would
120 allow for a circular flow of the nitrogen cycle when compared to nitrogen removal.
121 More specifically, the nitrogen recovery from wastewater can not only resolve to
122 some extent environmental issues such as eutrophication, but also offer
123 supplementary nitrogen sources for fertilizer production. Hence, it is essential to use
124 techniques which are efficient and sustainable, in order to recover nitrogen from
125 wastewater with products available for further use in the industry.

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

132

133 **2. Nitrogen recovery by BMISs**

134 **2.1. Reasons for using BMISs for nitrogen recovery**

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

151 producing ammonium-rich solutions in wastewater treatment. Here additional steps
152 may be required to achieve final products available for direct application in industry,
153 such as struvite precipitation and adsorption.

154

155 **2.2. Current status of nitrogen recovery by BMISs**

156 According to the different implementations of membrane in or outside a bioreactor,
157 there are two typical types of BMISs for the recovery of nitrogen, including side-
158 stream and submerged configurations. In the side-stream BMIS, a separate unit
159 containing membrane module is placed outside of the bioreactor, in which the influent
160 would flow successively through the membrane module and bioreactor (Cerrillo et al.,
161 2021a). For the side-stream BMISs, membranes can be classified into electrodialysis
162 (ED), membrane distillation (MD), forward osmosis (FO), reverse osmosis (RO) and
163 nano filtration (NF) according to the driving force across the membrane. Moreover,
164 the membrane module is directly installed into the liquid phase of the bioreactor
165 (Zhang & Angelidaki, 2015). This system namely submerged BMIS mainly includes
166 anaerobic membrane bioreactor (AnMBR), osmotic membrane bioreactor (OMBR),
167 bio-electrochemical system (BES) and membrane photobioreactor (MPBR).
168 Compared to the side-stream BMISs, the submerged BMISs are favored since they
169 involve less infrastructure, lower energy consumption and fewer costs. Tables 1 and 2
170 summarize the side-stream and submerged BMISs for the nitrogen recovery from
171 wastewater.

172 **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 ± 300 mg/L of NH ₄ -N concentrated with concentration factor of 8	3.8 ± 1.2 kWh/kg·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 ₃ ⁻	Concentrated nitrate	Mohammadi et al. (2021)
MCDI	simulated wastewater	72% of NH ₄ ⁺ -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) ₂ SO 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)

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

177 **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 ± 31% of TAN recovered	1.36 ± 0.28 kWh/kg·N	struvite	Zamora et al. (2017)
MEC-FO	High-strength sidestream centrate	99.7 ± 13.0% of AN	0.91 kWh/kg·NH ₄ ⁺ -N;	Struvite	Zou et al. (2017)
Tubular Micro-Pilot MEC	liquid effluent of an anaerobic digestion process	2.5 ± 0.1 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 ± 1.3% of ammonia recovered	1.3 kW h/kg·N	(NH ₄) ₂ SO ₄	Qin et al. (2018)
MEC-chemical precipitation	digested sludge centrate	53 ± 5% of TAN recovered	4.5 kWh/kg·N	struvite	Barua et al. (2019)
BEM	Synthetic wastewater	68.1 ± 3.4% of ammonia recovered	2.91 kWh/kg·N	(NH ₄) ₂ SO ₄	Zhang et al. (2021)
3-chamber BEC	reject water	75.5 ± 4.6% of N	6.1 to 8.2 kWh/kg·N	Concentrated	Koskue et al.

		recovered		ammonium	(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 ² ·d of N recovered	5.7 kWh/kg·N	Volatile ammonia	Qin et al. (2017)
MPC-IE	Raw sewage	37.5% of NH ₄ -N recovered	Recovery of 7.4 kWh/kg·N	Concentrated ammonium	Gong et al. (2017)
GI-AnMBR	Synthetic sewage	95.5% of N recovered	4.5 L/d of CH ₄ 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 ± 14.3 mg N/mol recovered	Recovery of 0.058 kWh/kg·N	Microalgal biomass	González-Camejo et al. (2019)

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

192 **2.2.1. Side-stream BMIS**

193 **A. ED**

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

211

212 **B. MD**

213 MD is a thermal driven process, in which the temperature difference between the feed side
214 and permeate side drives the mitigation of vapour molecules to pass through the hydrophobic
215 membrane or gas permeable membrane (GPM) (González-García et al., 2021; Guo et al.,
216 2019). In the MD process, the volatile ammonia in the feed side can transfer through the
217 membrane to the permeate stream under acidic conditions (Darestani et al., 2017; Huo et al.,

218 2020), after which the condensed ammonium solution can be further used. It was reported that
219 the temperature and pH of feed solution containing ammonium are the major parameters
220 influencing the ammonia transfer and enrichment (Cerrillo et al., 2021c; He et al., 2018;
221 Laureni et al., 2013; Wu et al., 2016a). Noriega-Hevia et al. (2020) developed a mathematical
222 model to represent the time evolution of pH and nitrogen concentration during the recovery
223 process and argued that the solution pH is the most important factor for enriching the nitrogen
224 in the MD process. Munasinghe-Arachchige et al. (2021) reported the pH of the feed side in
225 the GPM reactor above 9.26 is beneficial to the formation of $\text{NH}_3(\text{g})$ due to the pK_a for NH_3 at
226 9.26. Garcia-González and Vanotti (2015) found that increasing the solution pH by additional
227 sodium hydroxide could increase the N recovery from 55% to 81% in the GPM reactor.
228 Compared to typical acid absorption and ammonia stripping (23.6 to 49.6 kWh/kg·N), it was
229 reported that lower energy requirements are needed in the GPM process for the nitrogen
230 recovery (0.22 to 1.2 kWh/kg·N) while the addition of any alkali reagent is not required
231 (Beckinghausen et al., 2020).

232

233 C. FO

234 The FO process depends on the difference in the chemical potential between the feed solution
235 (i.e., high water chemical potential) and draw solution (low water chemical potential), which
236 drives ions transport by the membrane from a low concentration solution (feed) to a higher
237 one (draw) (Phuntsho et al., 2012). Generally, water in the feed solution can move to the draw
238 solution through the FO membrane while the ammonium can be concentrated in the feed side
239 with appropriate draw solute. It was reported that the aquaporin FO membrane can enrich
240 almost 100% of $\text{NH}_4^+\text{-N}$ (Engelhardt et al., 2020). Compared to other membrane techniques,
241 the FO process is cost-effective and environment-friendly (Van der Bruggen & Luis, 2015).
242 This is despite the fact that the regeneration of draw solute from diluted stream needs
243 additional power, which may be a barrier for commercializing the FO process. Chekli et al.

244 (2016) indicated that it is significant to find an economically feasible hybrid process for
245 regenerating diluted draw solution; for example, integrating FO with MD may be feasible to
246 fulfill the purpose because the MD process as a post-treatment can recover fresh water from
247 the diluted draw solute of the FO process, which regenerates the draw solution and thus
248 creates a circular economy (Liu et al., 2016; Ray et al., 2019; Volpin et al., 2019). In addition,
249 the reverse salt flux (RSF) in FO processes ranges from 80 to 3000 mg/L (Hancock & Cath,
250 2009), but the structure of the membrane support layer and concentration of draw solution
251 have insignificant impacts on the RSF (Phillip et al., 2010). Overall, RSF can not only reduce
252 the concentration of draw solution, but also contaminate the feed solution. However,
253 magnesium-based draw solutes have been investigated to provide magnesium ions to the feed
254 side through RDF, which boosts up the struvite precipitation and thus facilitates the nitrogen
255 recovery in the feed solution (Volpin et al., 2018). Singh et al. (2019) utilized biomimetic
256 aquaporin membranes (thin film composite FO membranes incorporating aquaporin proteins)
257 to recover nitrogen from sewage while using divalent magnesium chloride as draw solution.
258 They found that 66% of ammonia could be recovered within 24 h and regular cleaning
259 enables restoration of membrane performance after every 24 h-cycle.

260

261 **D. RO and NF**

262 As high-pressure membrane processes, NF and RO have high rejection rates of salts and ions,
263 which are often used to recover ammonium from urine (Adam et al., 2018; Patel et al., 2020;
264 Ray et al., 2020). In the RO process, feed water is driven by osmotic pressure difference to
265 flow from dilute to a concentrated solution through a semi-permeable membrane (Ahuchaogu
266 et al., 2018). In this scenario, the process is driven by additional pressure greater than the
267 osmotic pressure. Ray et al. (2020) reported that 64% and 90% of unionized ammonia can be
268 recovered from hydrolyzed urine by RO and NF processes respectively. It was suggested that
269 almost all the ammonium-nitrogen can be enriched by the RO membrane through solution pH

270 optimization within the reactor because the ammonium-nitrogen mainly exists in NH_4^+ form
271 at $\text{pH} < 7$ (Vaneekhaute et al., 2012). In the NF separation process, 1–10-nm molecule with
272 pore size ranging from 1–5 nm can be rejected within the reactor (Shon et al., 2013).
273 Compared to the RO process, lower pressure is applied in the NF process as well as lower
274 energy input (Wafi et al., 2019). Pronk et al. (2006) revealed that the NF membrane could
275 reject nearly all the micro pollutants including phosphate, propanol, ethinylestradiol,
276 carbamazepine, diclofenac and ibuprofen, while nitrogen could permeate by the membrane for
277 its enrichment. The NF process is always used to concentrate nitrogen from urine with 6
278 kWh/L (Maurer et al., 2006).

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280 **2.2.2. Anaerobic membrane bioreactor**

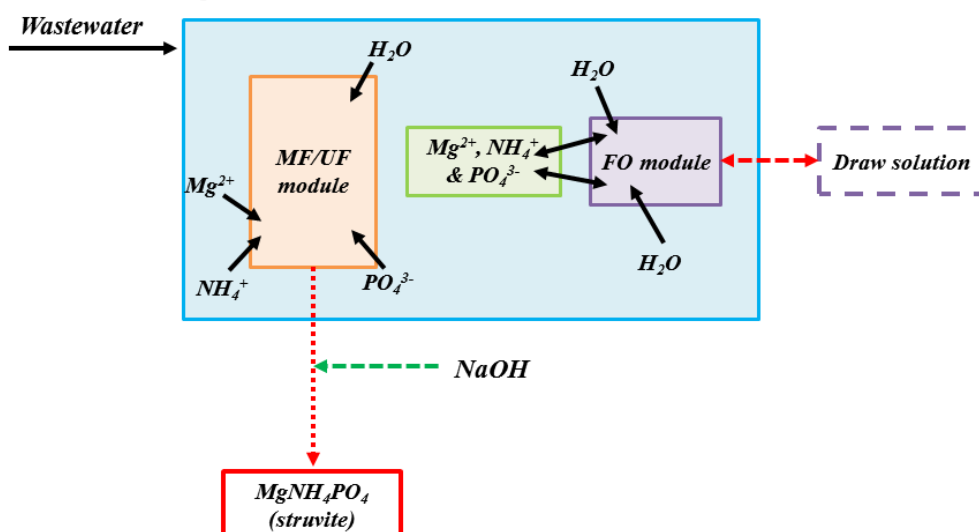
281 Anaerobic membrane bioreactor (AnMBR) has garnered increasing interest in addressing
282 energy challenges in conventional wastewater treatment plants. This is achieved by
283 integrating membrane separation with anaerobic digestion. Compared to aerobic MBR,
284 AnMBR has the advantage of solids-free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

296 COD.

297

298 2.2.3. Osmotic membrane bioreactor

299 Osmotic membrane bioreactor (OMBR) is developed by placing the FO membrane inside the
 300 bioreactor, showing superior permeate quality, a higher pollutants rejection rate, lower fouling
 301 propensity, and low energy consumption due to the FO process. The nitrogen in the influent
 302 can be enriched in the feed side of the bioreactor and then the stream containing rich
 303 ammonium and other nutrients achieves the nitrogen recovery in the form of struvite through
 304 pH elevation and additional necessary chemicals (Hou et al., 2017; Xie et al., 2014). 80% of
 305 ammonium can be recovered via struvite precipitation as reported by Qiu and Ting (2014),
 306 where NaOH was added to increase pH levels in the 8.0–9.5 range. Some studies utilized the
 307 microfiltration (MF) membrane and/or ultrafiltration (UF) membrane in the OMBR to extract
 308 the enriched ammonium and reduce the impacts of foreign substances, which works in
 309 parallel with the FO membrane (Holloway et al., 2015; Qiu et al., 2015), as illustrated in Fig.
 310 2. To address the issue associated with the draw solution in the OMBR, RO and MD process
 311 can be employed to regenerate the draw solute and thus increase the recovery system's
 312 feasibility (Chang et al., 2017; Luo et al., 2016).



313

314 **Figure 2.** Schematic of the osmotic membrane bioreactor (OMBR) integrated system for

315 ammonium recovery. Adopted from Ye et al. (2021b)

316

317 **2.2.4. Bio-electrochemical systems**

318 Bio-electrochemical systems (BES) can recover ammonium with simultaneous wastewater
319 purification and energy recovery, which mainly includes microbial electrochemical cell
320 (MEC), microbial fuel cell (MFC) and microbial desalination cells (MDC) (Cerrillo et al.,
321 2021b; Ye et al., 2021a; Ye et al., 2018). In BES, electrochemically active bacteria are used to
322 convert chemical energy stored in organic matter to electrical energy (Arredondo et al., 2015).
323 In a typical double-chamber BES, the anode chamber and cathode chamber are separated by a
324 proton/cation-exchange membrane (P/CEM). At the anode chamber, the substrates such as
325 organics are anaerobically degraded to produce hydrogen ions (H^+) and electrons (e^-), where
326 electrons migrate to the anode surface and then transfer to the cathode electrode through an
327 external circuit. Meanwhile, hydrogen ions pass through the P/CEM to the cathode chamber
328 to react with electron acceptors (e.g., air) to form water (Logan et al., 2006). In MFCs, a
329 thermodynamically favorable reaction takes place in the anode chamber for converting the
330 chemical energy into electrical energy with the reduction of oxygen. In contrast, external
331 electrical energy is required in MECs due to the presence of thermodynamically unfavorable
332 reactions in the cathode chamber (Wu & Modin, 2013). Thus, the main difference between
333 MFCs and MECs is favorability of the thermodynamic reaction taking place in spite of their
334 architecture being virtually the same.

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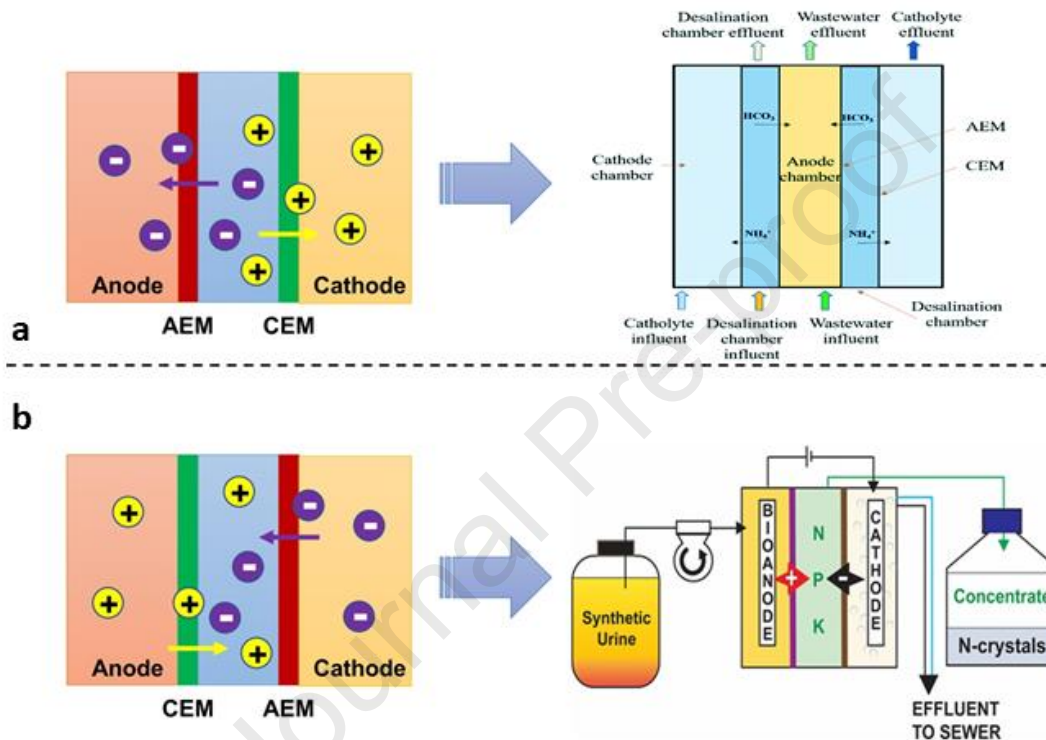
336 In MFCs, ammonium can be enriched in the cathode chamber due to its transfer from the
337 anode through the CEM. The cathode reaction could generate hydroxyl ions (OH^-) localized
338 the cathode electrode, which provides a high pH zone for initiating the quick conversion of
339 ammonium to volatile ammonia without the extra pH adjustment (Kuntke et al., 2011). The
340 concentrated ammonia stream obtained from cathode could be further harvested through

341 various methods, including struvite precipitation, adsorption and ammonia stripping. A decade
342 ago, it was reported that the MFC can simultaneously recover ammonium and energy of up to
343 3.29 g/d m² and 3.46 kJ/g (Kuntke et al., 2011), indicating the positive energy-balance
344 achieved in the ammonium-nitrogen recovery system. Compared to the MFC, MEC revealed
345 better nitrogen recovery from wastewater because additional power applied in MEC favors
346 the ammonium transfer from the anode chamber to cathode chamber. This process contributes
347 to the ammonium enrichment in the cathode compartment (Chen et al., 2017; Haddadi et al.,
348 2013; Hou et al., 2017; Ledezma et al., 2017).

349
350 In addition, MDC was used in wastewater treatment for concurrent resource recovery,
351 desalination and wastewater purification, including landfill leachate and anaerobic digester
352 liquor (Iskander et al., 2018; Liu et al., 2020a; Lu et al., 2020; Zhang & Angelidaki, 2015),
353 where its two main configurations are designed for recovering ammonium. From Fig. 3a, it
354 can be seen that CEM faces the cathode while AEM faces the anode in the first configuration.
355 In this scenario, high-strength ammonium wastewater is desalinated in this chamber while
356 ammonium ions mitigate across CEM, and are further accumulated in the cathode chamber.
357 For example, Zhao et al. (2019) found that 69% of ammonium could be recovered in the
358 catholyte, and 72% was removed in the desalination chamber while applying a tubular MDC
359 for recovery of ammonium from a diluted draw solution of the FO process. Apart from this,
360 another configuration drives the anions in the catholyte and cations such as ammonium in the
361 anolyte into the middle chamber through an electrical field. Consequently, both cation and
362 anion nutrients, such as phosphate and ammonium can be recovered from wastewater
363 (Ledezma et al., 2017).

364
365 Overall, the ammonium recovery using BES is affected by: (1) the equilibria among other
366 ions at both anode and cathode; (2) the property of IEM; (3) the catholyte pH; (4) applied

367 power; and (5) the ammonium concentration in wastewater (Arredondo et al., 2015;
 368 Arredondo et al., 2017; Mousavi et al., 2021; Qiu et al., 2020; Wan et al., 2010). BES offers a
 369 promising option for ammonium recovery from a concentrated stream, but Zhang and Liu
 370 (2021) argued that it is not unrealistic to recover ammonium in an energy and cost-effective
 371 manner applying BES from wastewater containing low levels of ammonium (40 mg/L). Thus,
 372 further investigations on the engineering feasibility of BES should be conducted.



373
 374 **Figure 3.** Two designs of MDC for resource recovery. Adopted from Liu et al. (2020a)

375

376 2.2.5. Membrane photobioreactor

377 Currently, wastewater treatment, specifically ammonium is generally removed via catabolism
 378 (i.e., disassimilation) and microbial anabolism (i.e., assimilation), which is finally assimilated
 379 into biomass or converted into nitrogen gas (Rittmann & Mccarty, 2014). The circular
 380 economy framework triggers the application of phototrophs (e.g., phototrophic bacteria and
 381 microalgae) for concurrent wastewater treatment and nutrient recovery (Liu et al., 2018).

382 Huelsen et al. (2016) reported that the NH_4^+ -N and COD are assimilated by microbial activity

383 of purple phototrophic bacteria (PPB) in a certain ratio of $1 \text{ g} \cdot \text{NH}_4^+ \text{-N} / 16 \text{ g} \cdot \text{SCOD}$, but the
384 concentration of $\text{NH}_4^+ \text{-N}$ and COD (40 and 400 mg/L, respectively) in typical sewage cannot
385 satisfy the ratio, so additional carbon source is needed if the acceptable effluent
386 concentrations for discharge and reasonable nutrients assimilation are prioritized. Besides,
387 since PPB can only use sugars, alcohols, organic acids, etc., as the substrates, the pretreatment
388 of wastewater is also required. However, this inevitably increases the economic burden and
389 limits the commercial application of PPB for the nitrogen recovery from sewage.

390

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

407

408 Therefore, membrane photobioreactors (MPBR) were widely investigated for achieving

409 minimum footprint with easy biosolid-liquid separation (Ji et al., 2020). Integrating
410 membrane technology with microalgae can reduce energy consumption and chemical usage
411 with simple operation, easy to scale up, and excellent fractionation capability (Zhang et al.,
412 2020). Chang et al. (2019) utilized a scalable membrane-based tubular photobioreactor
413 (SMPBR) to realize high-efficiency nitrogen recovery (74.31%) from landfill leachate.
414 Compared to the traditional PBR, the microalgal biomass concentration was improved to
415 2.13 g/L in SMPBR. Similarly, Nguyen et al. (2021) employed the nitrogen from urine to
416 cultivate the microalgae in the MPBR, in which high recovery rate of nitrogen (TN of 90.5
417 mg/L·d) can be synergistically achieved with biomass production (biomass productivity of
418 313 mg/L·d) at biomass retention times (BRT) of 7 d.

419

420 **2.3. Generation of bioproducts in nitrogen recovery towards circular bioeconomy**

421 **2.3.1 Biofertilizer**

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

429

430 Struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) is the most commonly achieved product in the nitrogen recovery
431 system, which is also a micronutrient critical to the growth of plants and crops due to
432 simultaneously containing nitrogen and phosphorus. Due to the property of struvite such as
433 slow nutrient release, it has advantages over other fertilizers (Song et al., 2011). Since Mg is
434 an essential element of chlorophyll, the existence of Mg also enhances the application

435 prospect of struvite as an efficient fertilizer for grasses. Compared to the commercially
436 available fertilizers, struvite obtained from nitrogen recovery system has low content of heavy
437 metal ions (Latifian et al., 2012). As a result of this, struvite is relatively less harmful to the
438 plant roots. Kern et al. (2008) argued that the loading rate of Cd in struvite obtained from
439 wastewater is reduced by a factor of 10 when compared with triple superphosphate (derived
440 from natural phosphate deposits). A comparison of fertilizer efficiency between the chemical
441 fertilizer and struvite was conducted by Rasul et al. (2011). They detected no significant
442 difference between the dry matter yield, leaf area and plant height while applying these two
443 fertilizers to the maize crop.

444

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

461
462 In addition, ammonium sulfate can comparatively yield 10% because it can lower the soil pH
463 and increase leaf nitrogen. This is despite the fact that the application of ammonium sulfate
464 may increase soil electrical conductivity and negatively affect the yield. Similarly, Vargas and
465 Bryla (2015) found that ammonium sulfate-based fertilizers are more efficient for the growth
466 and yield of highbush blueberry plant in the first five years compared to the granular
467 application of either nitrogen source. Chien et al. (2011) indicated that the addition of
468 ammonium sulfate provides sulfur to the soil which is lacking in many other proposed
469 fertilizer alternatives. Nitrogen can also be recovered by adsorption via biochar, where the
470 application of nitrogen-loaded biochar can enhance crop yield in agriculture as a slow-
471 releasing fertilizer due to the presence of an aromatic group (Pathy et al., 2021). For instance,
472 the pumpkin yield can be increased four-fold while applying low-dose urine impregnated
473 biochar to the pumpkin field (Schmidt et al., 2015). Wu et al. (2016b) also found the
474 microbial dynamics of soil can be improved by adding biochar. The application of nitrogen-
475 loaded biochar can also sequester carbon in the applied soil, reduce nitrogen loss and improve
476 soil productivity (Yao et al., 2013). Apart from this, the soil's pH can be improved as well as
477 its cation exchange capacity while utilizing the nitrogen-based biochar obtained from the
478 urine, which also strongly suggests the soil conditioner potential of biochar (Bai et al., 2018).
479 In one study, polymer matrix biochar composite was exploited to recover ammonium from
480 urine, and the resulting nitrogen-loaded biochar was applied to the cotton plant (Wen et al.,
481 2017). In this scenario, adding nitrogen-loaded biochar has various positive impacts on cotton
482 plants, including enhanced nitrogen use efficiency and less N leaching. When Mg modified
483 biochar was utilized to recover ammonium from urine, the nitrogen-laden biochar can
484 enhance the plant height of ryegrass and maize (Xu et al., 2018). In contrast, Mg biochar does
485 not exert any positive effect on ryegrass and maize.

486

487 Microalgae-based fertilizer can promote microorganisms demand for: plants growth;
488 solubilize phosphate and fix nitrogen through providing proteins; vitamins required for
489 enhancing plant growth; carbohydrates; natural enzymes; polyamines; growth regulators;
490 antifungal substances; amino acids; carotenoids; phytohormones and micro- and macro-
491 nutrients (Patil et al., 2008; Ronga et al., 2019). The plants' physiological processes can be
492 improved and regulated while applying the microalgae-based fertilizer at low doses in
493 agriculture (Morais Junior et al., 2020). Such fertilizers can increase the shelf life,
494 productivity and quality of the plants and crops, improving the tolerance to abiotic stresses
495 and nutrient absorption (Ronga et al., 2019). Due to an increase in carotenoid content and
496 sugar, the application of microalgae-based fertilizer in tomato cultivars can improve the
497 quality of the fruit (Coppens et al., 2016). In this scenario, the tomatoes processed by
498 microalgae-based fertilizer can contain 44% more carotenoids than the fruit treated by organic
499 fertilizer and 70% more carotenoids than those treated by inorganic fertilizer. Furthermore, it
500 was reported that water extract of *Chlorella ellipsoidea* and *Spirulina maxima* can: firstly,
501 improve wheat tolerance for salinity; and secondly, enhance the antioxidant capacity and
502 protein content of the whole grains produced by treating plants with microalgal extracts (El-
503 Baky et al., 2010).

504

505 **2.3.2 Microalgae Biomass**

506 In most current studies, microalgae production is achieved by photoautotrophic cultivation,
507 through which large-scale biomass can be obtained with the use of sunlight that is a free,
508 renewable, and is a clean source of energy (Lam et al., 2012). Currently, MPBRs are
509 investigated for assessing nitrogen recovery and biomass production from different
510 wastewater sources, including agricultural effluent, slurry wastewater and sewage (Gao et al.,
511 2016). Microalgae biomass can be used as raw materials in the feed, and food industry
512 because of its high health and nutritional value (Morais Junior et al., 2020). More specifically,

513 it can provide pigments such as chlorophylls and carotenoids, minerals, mono- and n-3
514 polyunsaturated fatty acids, polysaccharides, essential amino acids and vitamins
515 (Priyadarshani & Rath, 2012). Vermaas (2004) reported that microalgae biomass can be
516 employed as supplements for feed through several nutritional and toxicological tests. In
517 poultry feed, microalgae biomass can supplement conventional proteins to improve the yellow
518 color of egg yolk and broiler skin (Becker, 2003). Further research should consider different
519 biochemical properties of various microalgae species while applying them as supplements in
520 novel animal diets (Batista et al., 2013).

521
522 Due to containing bioactive compounds such as sterols, carotenoids and polysaccharides,
523 microalgae can reduce cholesterol and have various health benefits, for instance anti-cancer
524 properties and other features (Lordan et al., 2011; Raja et al., 2018). Microalgae or its extracts
525 can be also used as medicine against both gram-negative and gram-positive bacteria, and
526 bacterial infections in fish or shrimp feed due to containing long-chain polyunsaturated fatty
527 acids (LC-PUFAs) (Desbois et al., 2009; Shah et al., 2018; Yaakob et al., 2014). Furthermore
528 the microalgae extracts can be widely employed in the cosmetics market for many products
529 including sun protection, hair protection, peelers, emollients, regenerating products,
530 refreshing products and anti-aging creams, and especially for *Arthrospira* and *Chlorella*
531 (Morais Junior et al., 2020). Extracts from microalgae species not only can be antimicrobial,
532 but also have potential for anti-aging skincare products, which contributes to their wide-
533 ranging usage in cosmetics-related areas (Mourelle et al., 2017).

534
535 Microalgae are also employed to accomplish nitrogen recovery coupled with CO₂
536 sequestration at wastewater treatment (Molazadeh et al., 2019; Razzak et al., 2017). The
537 capacity of microalgae for CO₂ sequestration varies from species; for example, the carbon
538 uptake rate of cyanobacteria *A. microcopia Nageli* is 28 mg/L min, which is higher than that

539 of diatom *P. tricornutum* at 1.5 mg/L·min (Francisco et al., 2010). Through investigating the
 540 microalgae application in an ethanol synthesis factory, it can be seen that microalgae are
 541 feasible for sequestering CO₂ for its growth in industry (Rosenberg et al., 2011). This finding
 542 indicates that microalgae can be utilized capturing CO₂ to save the costs associated with the
 543 chemical removal process (Kumar et al., 2010). It has been reported that *Chlorella* strains
 544 from hot springs can be tolerant to CO₂ concentration up to 40% (v/v) (Rizwan et al., 2018).
 545 In another study, 50% SO₂, 70% of NO and 60% of CO₂ can be removed from flue gas by
 546 *Chlorella* sp. cultures, which mitigates greenhouse gases (Chiu et al., 2011). Microalgae
 547 biomass was also explored as a material for the production of biofuels to generate clean and
 548 sustainable energy (Pienkos & Darzins, 2009). Other potential applications of microalgae
 549 biomass have been explored, including nutraceuticals, biofertilizers and bioplastics
 550 (Borowitzka, 2013; Lu et al., 2016b; Sathasivam et al., 2019a). Table 3 summarizes the
 551 applications of microalgae biomass.

552
 553 **Table 3.** Summary of microalgae biomass applications

Applications	Microalgae	Component	Application	Reference
Cosmetics	<i>Dunaliella</i> <i>Salina</i>	<ul style="list-style-type: none"> ◆ Phenols such asferulic acid and caffeic ◆ Glycerol 	<ul style="list-style-type: none"> ◆ Smoothen and Moisturize skin 	Goiris et al. (2012); Safafar et al. (2015); Ariede et al. (2017); and Sathasivam et al. (2019b)
Pigments	<i>Chlorella</i> <i>zofingiensis</i>	<ul style="list-style-type: none"> ◆ α-carotene ◆ β-carotene 	<ul style="list-style-type: none"> ◆ Pigmenter for salmon 	Sathasivam et al. (2019b); Gouveia and Empis (2003)
Biofuel	<i>Botryococcus</i> <i>braunii</i>	<ul style="list-style-type: none"> ◆ high lipid content containing 10.54% of polyunsaturated fatty acids 9.95% of saturated fatty 	<ul style="list-style-type: none"> ◆ biodiesel production 	Tasić et al. (2016) and Caetano et al. (2020)

			acids and 79.61% of monounsaturated fatty acids		
Bioplastics	<i>Chlorella</i> sp.	♦	high carbohydrates content (more than 60% dw)	poly-lactic acid	Wang et al. (2014) and Aikawa et al. (2012)
Biofertilizer	<i>Scenedesmus</i> sp. <i>Arthrospira platensis</i>	♦	higher concentration of abscisic acid, salicylic acid, auxins, gibberellins and cytokinins	♦ improve the plant nutrient status ♦ enhance the number of flowers per plant and root dry matter ♦ accelerate plant development	Plaza et al. (2018)
food industry	<i>Dunaliella salina</i>	♦	Carotenoid β -content ♦ High levels of antioxidant ♦ Carbohydrate (32% dw) ♦ Protein (57% dw)	♦ 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	<i>Arthrospira platensis</i>	♦	Carbohydrate (13–16% dw) ♦ Protein (60–71% dw)	♦ Increase the number of lactic acid bacteria ♦ Produce low cholesterol eggs ♦ Vegan	Mobin and Alam (2017); Niccolai et al. (2019); and Raja et al. (2008); Becker (2007)

Medicine	<i>Arthrospira platensis</i>	<ul style="list-style-type: none"> ◆ Essential polyunsaturated fatty acids (PUFA) ◆ β-carotene ◆ Vitamin K and B₁₂ and B1 ◆ Provitamin A ◆ Isoleucine valine ◆ γ-Linolenic acid (GLA) ◆ Phycocyanin (colorant) 	<ul style="list-style-type: none"> ◆ Prevent rheumatoid arthritis, diabetes, multiple sclerosis, dermatitis, viral infections and schizophrenia ◆ Immune system enhancement ◆ Anti-inflammatory ◆ Antioxidant ◆ Anticancer 	replacement Sathasivam et al. (2019b); Vermaas (2004); Spolaore et al. (2006); ; and
	<i>Chlorella zofingiensis</i>	<ul style="list-style-type: none"> ◆ Glucan ◆ β-carotene ◆ Astaxanthin 	<ul style="list-style-type: none"> ◆ 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)

554

555 **2.3.3 Bioenergy**

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

563 Takriff, 2017).

564

565 Raheem et al. (2018) reported that microalgae biomass can be converted into biofuels such as
566 bioethanol and biodiesel, which has significant effects on global warming and environmental
567 pollution (Khan et al., 2018). Compared to most terrestrial plants, microalgae can
568 effectively convert energy into chemical energy 5-fold due to its simple structure (Yap et al.,
569 2021). More importantly, the microalgae can reach fast growth rates, and some species of
570 microalgae can double their biomass in just a few hours because their growth rate is 5–10
571 times quicker than traditional food crops (Okoro et al., 2019; Rodionova et al., 2017).
572 Furthermore, there is no need to conduct any modifications of existing fuel engines because
573 microalgae-based biofuels are compatible with these (Ras et al., 2013). Compared to the
574 common oil crop, the lipid productivity of microalgae biomass is 15–300 times larger (Morais
575 Junior et al., 2020; Zullaikah et al., 2019), so it is feasible to supplement the vegetable oils
576 obtained from terrestrial crops. For example, the oil yield of microalgae per hectare is higher
577 than that of rapeseed, soybean and palm oil (Paniagua-Michel et al., 2015). More importantly,
578 the biofuels productivity of microalgae biomass is 10–20 times higher than any other biofuel
579 crop (Mata et al., 2010; Ndimba et al., 2013). One species, *Dunaliella tertiolecta*, can
580 produce 25.8% of bio-oil yield by hydrothermal liquefaction of microalgal biomass residues
581 and methylation of different fatty acids (Shuping et al., 2010; Tang et al., 2011).

582

583 In anaerobic digestion, *Dunaliella salina* can produce biogas because of its easy processing,
584 low cost for pre-treatments, and high biomass productivity (J. Nayeong et al., 2012). In their
585 study, Markou et al. (2013) utilized a medium with phosphorus limitation to cultivate
586 *Arthrospira platensis* to achieve carbohydrate-enriched biomass. Then the biomass was
587 employed as a substrate to produce bioethanol through fermentation with bioethanol
588 productivity at around 16 g·ethanol/g·biomass. Furthermore, the microalgae biomass can be

589 cultivated by wastewater in non-arable land, which contributes to sustainable wastewater
590 reuse and management, and results in no competition with food crops for farming land
591 (Brennan & Owende, 2010; Christenson & Sims, 2011). Nevertheless, microalgae
592 applications as a bioenergy source are still subjected to the dual challenges of harvesting and
593 management of microalgae from large volumes, and high biomass production (Brennan &
594 Owende, 2010).

595

596 Bio-membrane integrated systems can also recover energy in the form of biogas and
597 electricity generation, where biogas production depends on the activity of methanogens in
598 anaerobic conditions whilst the electricity generation occurs in the BES-based integrated
599 systems. Biogas is considered as an alternative to fossil fuels, and it can be recovered from
600 organics through anaerobic biological processes from wastewater treatment (Li et al., 2019).
601 Maaz et al. (2019) reported that the recovered biogas comprises nitrogen (0 ~ 15%), hydrogen
602 gas (0 ~ 5%), carbon dioxide (3 ~ 15%) and methane (50 ~ 90%). For the AnMBR, the
603 methane yield is negatively affected due to it being exposed to high salinity levels at the
604 beginning of the operation. This scenario is caused by the reversal of solute leakage (Gu et al.,
605 2015). However, the methanogens may gradually adapt to built-up high salinity environments,
606 and the produced methane can be recovered (Gu et al., 2015).

607

608 Taking anaerobic OMBR (AnOMBR) as an example, Yang et al. (2021) reported that the
609 methane produced in the AnOMBR ranges from 60 to 301 mL/g·COD, which is influenced
610 through the conductivity of the feed solution in the range of 2.5-20 mS/cm. Moreover, the
611 AnOMBR could achieve higher methane yield at lower salinity accumulation. The change in
612 the microbial function and composition may result in varying methane production in
613 AnOMBR, which is attributed to RSF (Li et al., 2017). Therefore, it is important to control the
614 salinity level in the OMBR while using an inorganic ionic substance that serves as a draw

615 solute (Ansari et al., 2015). In this scenario, the environment which is beneficial for the
616 growth of anaerobic bacteria can be created, and the efficiency of pollutant removal, and
617 stable methane production can subsequently be maintained. Integrating pressure-driven
618 membrane processes such as MF and UF or BES with AnOMBR may be a possible solution
619 to improve the biogas production through mitigating salinity accumulation; for example,
620 Wang et al. (2017a) found that the salinity level can be effectively kept in a stable range of
621 2.5 ~ 4.0 mS/cm while integrating AnOMBR with the MF membrane (AnOMBR-MF).
622 Compared to the conventional AnOMBR, the AnOMBR-MF system can achieve higher
623 methane production with 280 mL/gCOD during long-term continuous operation. Besides, the
624 practical methane production is 28–39% lower than the theoretical value in AnOMBR-based
625 system due to methane loss. Currently, research on the biogas production by lab-scale
626 AnOMBR is too limited to provide effective analysis (Wang et al., 2016). For this reason,
627 further study should focus on the scaling up of the AnOMBR for the methane production and
628 collection.

629

630 The above discussions demonstrate that BES-based systems can generate electricity during
631 the nitrogen recovery from wastewater, including landfill leachate, domestic wastewater and
632 industrial wastewater. However, the electricity produced by microbes is subjected to low
633 power densities achieved at high reactor volumes (Penteado et al., 2018). For example,
634 approximately 150 W/m³ of power density were obtained in MFC reactor volumes of 1.2 mL,
635 but increasing the volume to 100 mL resulted in power density falling to 50 W/m³ while
636 cultivating bacteria *Shewanella oneidensis* DP-10 at the anode chamber (Biffinger et al.,
637 2007). The electricity generated in the MFC is affected by various factors including the type
638 of wastewater, biofilm formation, membranes, mediators, substrates, the configuration of
639 electrodes, electrolyte temperature, electrolyte pH and MFC design (Apollon et al., 2021; Qiu
640 et al., 2021).

641
642 The dual-compartment MFCs are always used for the nitrogen recovery at wastewater
643 treatment, where their designs mainly include tubular, miniature, flat-plate type, cube-type and
644 H-type MFCs (Gul et al., 2021; Song et al., 2015). While employing glucose as an organic
645 substrate, the power density of cube type and flat plate MFCs were 910 and 212 mW/m²,
646 respectively, which is higher than that of H-type MFC (115.6 mW/m²) (Min & Logan, 2004;
647 Yang et al., 2017; You et al., 2006). This may be attributed to the lower membrane surface
648 areas for proton transfer in H-type MFCs (Logan et al., 2006). Furthermore, the organic
649 substrates used for microbial growth also affect the MFC performance; for example, the
650 wastewater generated by processing starch provided the power density of 239.4 mW/m² (Lu
651 et al., 2009) whilst municipal wastewater treated by MFC presented power density of 52
652 mW/m² (Zhang et al., 2011). Gonzalez del Campo et al. (2013) argued that raising the
653 temperature from 20 to 40 °C can result in expanding power density from 0.73 to 1.01
654 mW/m². Various investigations were conducted to enhance the efficiency of bioenergy
655 generation in MFC reactors, but the commercialization of MFC still needs significant
656 refinements to overcome serious practical issues.

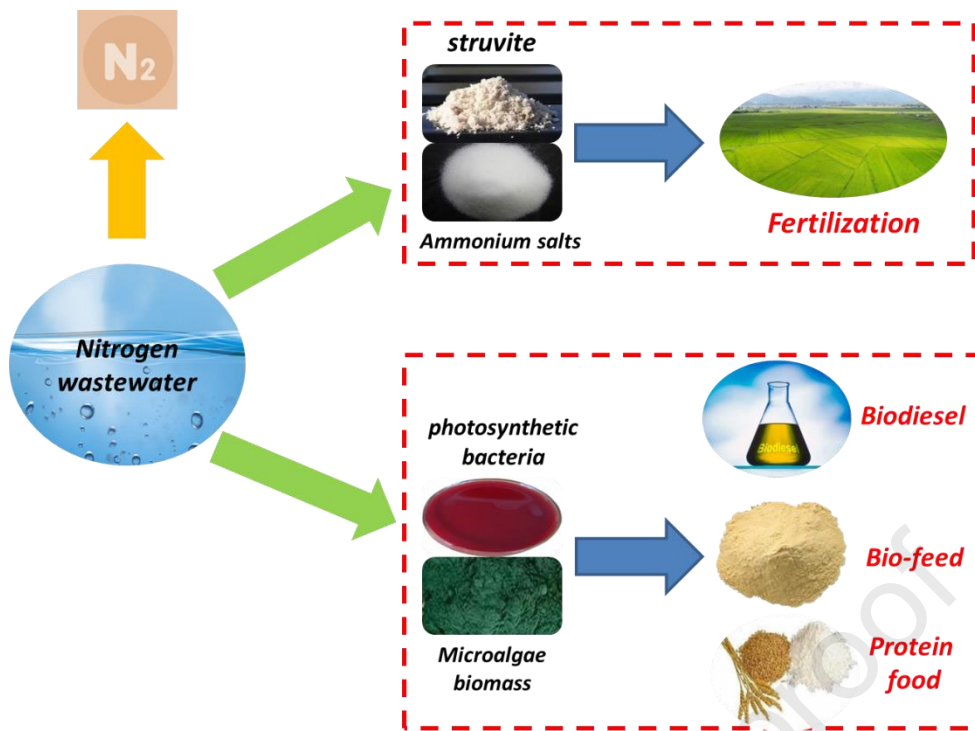
657

658 **3. Challenges and future perspectives**

659 Wastewater as a gradually accepted alternative and renewable source of fresh water and
660 nutrients is due to its existence in extremely large volumes. However, conventional
661 wastewater treatment aims to remove the so-called pollutants, purify the wastewater, but
662 current wastewater treatment prefers to recover renewable resources. In particular, current
663 wastewater treatments generally convert ammonium to valueless nitrogen gas at rather
664 economic and environmental costs. At the same time they simultaneously generate a
665 substantial amount of GHGs which is typical of the linear economy (i.e., take-consume-
666 dispose). The primary goal of conventional wastewater treatment is to satisfy the effluent

667 discharge standards, but may result in wasting large amounts of valuable resources which
668 should be recovered, and then reused. Consequently, the spirit of a circular economy departs
669 from the circular bio-economy, and this should be a major driver in future technological
670 developments that benefit society, especially given the challenges of energy, food security,
671 resources depletion and climate change.

672
673 In contrast to the linear economic model, circular bio-economy aims to improve the
674 valorization and reuse of residues and organic wastes, which converts bio-waste and their
675 intermediate products into bioenergy, biomaterials, and decouples growth from the
676 consumption of finite natural resources (Mohan et al., 2016). For this reason, it is essential to
677 shift the paradigm for biological wastewater treatment processes from removing to recovering
678 and thereby reusing. It marks an essential step towards greater environmental and economic
679 sustainability. The recovery of nitrogen from wastewater can reduce our dependence on
680 chemical fertilizers, produce bioproducts with economic value, decrease eutrophication
681 problems, and satisfy the effluent nitrogen concentrations required by government legislation.
682 Due to the relatively low ammonium-N concentrations in some wastewaters, the ammonium
683 recovery by current technologies is environmentally unsustainable, and economically costly.
684 To address this issue, the solution to tackle the challenge of concurrent wastewater treatment,
685 and ammonium should rely on BMISs. The use of BMISs for nitrogen recovery can help
686 produce effective biofertilizers, high-value biomass and generate energy. The ammonia–
687 nitrogen recovery from wastewater and its use are shown in Fig. 4.



688

689 **Figure 4.** Ammonia–nitrogen recovery from wastewater and its use

690

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

702

703 **3.1. Economic analysis**

704 The cost-effectiveness of nitrogen recovery is a key incentive driving the development of

705 BMISs on a commercial scale. For this reason, the techno-economic analysis involving
706 energy consumption, reaction efficiencies, and operating parameters are important for
707 evaluating the applicability of one technology for recovering nitrogen from wastewater.
708 Besides, the production of ammonium through BMISs must be more economical than the
709 conventional ammonia synthesis process, so the energy consumption per unit of nitrogen
710 recovery was the most crucial factor. Although some studies have conducted such analyses
711 (Mohammadi et al., 2021; Qin et al., 2017; Ward et al., 2018; Zou et al., 2017), most of these
712 values used for the current calculation are achieved in lab-scale reactors for short-term
713 experiments. Hence, the commercial application of BMISs is still questionable, including lack
714 of solid outcomes in pilot-scale operations of BMISs, uncertain durability, and costly
715 component materials (e.g., membrane and electrode). Long-term operation of large-scale
716 BMISs in real wastewater environments is currently rarely investigated and evaluated, so the
717 current techno-economic analysis's reliability is in doubt. For this reason, it is necessary to
718 evaluate the performances and challenges of large-scale systems for long-term operations.

719

720 **3.2. Technical analysis**

721 The bio-membrane integrated systems involve microbial activity, so there are many additional
722 problems in systems, such as, undesired substrate consumption via competing metabolic
723 processes, unwanted biomass growth, and incomplete biodegradation of substrates, which
724 would curtail amount of nitrogen recovered in the system (Pandey et al., 2016). Ahmed et al.
725 (2019) believed that a combination of technologies may be a solution to maximize the
726 efficiencies of nitrogen recovery in BMISs. Besides, scaling of the membrane is required in
727 order to advance application of membrane-based techniques. Membrane fouling may not only
728 contaminate permeate, but also deteriorate the membrane's efficiency over long-term
729 operation of membrane-based processes. Another concern about the membrane-based
730 treatment process is the cost of membranes. Further studies should focus on the advanced

731 development of membranes to improve their properties and reduce the synthesis costs.
732
733 Compared to other membrane separation technologies, OMBR integrated system presents its
734 unique advantages of low energy input and membrane fouling potential (Yang et al., 2021).
735 Nevertheless, the recovery of draw solute in the FO process still hinders the commercial
736 application of OMBR integrated systems (Chekli et al., 2012). Conventional draw solute
737 recovery processes including ED, MD, and RO were investigated, however, these methods
738 require high energy consumption, and thus increase the overall outlay. For instance, high
739 energy consumption of 1.88-4.01 kWh/m³ (i.e., an operation cost of 0.2-0.4 \$/m³) is required
740 to recover salt from the draw solution (Ortiz et al., 2008). Similarly, the use of a MD process
741 can successfully recover water from the draw solute, in which additional energy input of 29
742 kWh/m³ (i.e., an operation cost of 2.9 \$/m³) is required to drive the MD operation (Zhao et al.,
743 2014). This is despite the fact that solar energy and power plants can provide energy sources
744 to reduce energy consumption in the MD process. Therefore, further efforts should be made to
745 devise effective and economic recovery of draw solute with possible use of renewable energy
746 in the OMBR integrated systems.
747
748 Due to its ability to be integrated with the currently existing wastewater treatment system, for
749 the purposes of alleviating energy demand, and pollution problems, BES is considered as a
750 platform for the conversion of waste into energy and chemicals. The rapid degradation of
751 organic wastes in the anode chamber would allow for the prospect of their commercial
752 application (Kadier et al., 2016; Sadhukhan et al., 2016; Santoro et al., 2017). These
753 advantages make BESs a key part of the promising circular bio-economy model. The internal
754 resistance is an important parameter affecting the BES performance (Arredondo et al., 2015).
755 At the anode chamber of BESs, the microbial environments need effective controls to
756 improve their performance. The possible reason for this is that the electricity generation in

757 BES may be detrimentally affected by the deactivation undesired side reactions of anode
758 (Tota-Maharaj & Paul, 2015; Zhang et al., 2013). More specifically, accumulation of organic
759 wastes on the anode surface, consumption of organics and electrons for undesired side
760 reactions, and interruption of bio-electrochemical reactions due to the presence of impurities
761 may compromise the power density of BESs (Fornero et al., 2010; Rabaey et al., 2005; Zhang
762 et al., 2013). Therefore, a pretreatment process is essential. This is despite the fact that
763 additional processes will increase the costs, so more economic analysis is required here. In
764 addition, the selection of materials including membrane and electrodes are significant for BES
765 commercialization. For example, bio-electrochemical efficiency of cathodic reactions is
766 influenced by the cathode materials and the production rate of chemicals (Shahgaldi &
767 Hamelin, 2015; Sonawane et al., 2013). Pt-supported cathode electrodes are traditionally used
768 at the cathode chamber of BESs (Wang et al., 2017b), but their high costs ($> \$1500/\text{g}$, Sigma-
769 Aldrich) are a barrier to commercialization. This is despite the fact that the application of Pt-
770 based electrodes improves the oxidation-reduction potential. Apart from this, the use of
771 expensive and complicated membranes (e.g., Nafion®, $> \$1500/\text{m}^2$, Sigma-Aldrich) results in
772 costly BESs. Several issues may occur to negatively affect the performance of the membrane,
773 such as fluctuating wastewater treatment conditions, contamination and accumulation of
774 microbes, and gas leakage between two chambers (Xu et al., 2012). To successfully
775 commercialize the BESs, it is of great importance to develop membranes and electrodes that
776 are cheaper and more efficient compared to the currently existing commercial ones.

777

778 The nitrogen recovery through MPBR integrated systems is affected by photosynthetic
779 efficiency, dissolved gases, solution pH, mixing, temperature and irradiance (Anto et al.,
780 2020). The biggest challenge of MPBR integrated systems is the availability of sunlight in the
781 case of light irradiance, so it is recommended to use an acrylics type material to realize the
782 cost-effectiveness of the establishment. It is also essential to prevent the microalgae

783 accumulating on the walls of the tubes, as this may undermine sunlight transmittance.
784 Microalgae can continuously produce biomass under simulated illumination, even during
785 night hours. Another important factor affecting the performance of MPBR integrated systems
786 is nutrient solubility, so effective pH control is significant. Consequently, sparger or the
787 bubbler kind of arrangement is always used to control gaseous CO₂ which may result in the
788 generation of HCO₃⁻ and subsequently affecting the solution pH. The development of a
789 MPBR integrated system for processing, harvesting and cultivating microalgae biomass is
790 vital to enhance the system's sustainability, and make the system more economically viable.

791

792 **4. Conclusion**

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

805

806 **Acknowledgement**

807 This research was supported by University of Technology Sydney, Australia (UTS, RIA NGO
808 and UTS SRS 2021) and the Korea Institute of Energy Technology Evaluation and Planning

809 (KETEP) and the Ministry of Trade, Industry & Energy (MOTIE), Republic of Korea (No.
810 20183020141270 and No. 20194110300040).

Journal Pre-proof

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