

## REVIEW ARTICLE OPEN

## Membrane-based technologies for post-treatment of anaerobic effluents

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Anaerobic digestion-based processes for converting wastewater into clean water and energy are attracting ever-growing industrial interest. However, apart from the microbial digestion step, current technologies require further progress from an integrated process point of view, including post-treatment steps. Anaerobic effluents normally undergo extensive post-treatment steps to meet stringent discharge standards, while valuable nutrients are rarely recovered. Additionally, a significant portion of the produced methane remains inevitably dissolved in the effluent, which is eventually released into the environment, causing economic loss and global warming concerns. To address these issues, several membrane-based technologies show significant promise. Here, we review current progress in membrane-based recovery of dissolved methane and nutrients, highlighting opportunities where membrane-based technologies can improve the post-treatment of anaerobic effluents. Lastly, we also share our perspectives for promising research directions and how to secure the competitiveness of membrane-based technologies for anaerobic wastewater treatment processes, focusing on current challenges for membrane development, biofouling mitigation strategies, and small-scale to large-scale implementation.

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## INTRODUCTION

The growing environmental concerns and impending global water crisis have changed the landscape of wastewater treatment practices dramatically in the last couple of decades.<sup>1–3</sup> In particular, the anaerobic processes have driven a massive ongoing change owing to their appealing advantages, such as bypassing the need for energy-intensive aeration (of aerobic processes), producing valuable products (e.g., biofuels and biofertilizers), and generating less and easier-to-handle sludges.<sup>2–5</sup> At present, anaerobic processes serve as the core technology behind several wastewater treatment plants worldwide.<sup>2–5</sup> However, the anaerobic wastewater treatment practices are still far from being fully mature, and there is a need for integrating them more efficiently with both conventional and continually emerging separation methods. To our understanding, the membrane-based separation technologies deserve special attention in this regard, which is rationalized below after defining the problems of anaerobic digestion processes.

The anaerobic digestion processes suffer from two major issues. First, due to the metabolic constraints of involved microbial digester communities, nitrogen (N) and phosphorus (P) species are poorly eliminated by anaerobic processes and only break down into ammonium (NH<sub>4</sub><sup>+</sup>) and phosphate (PO<sub>4</sub><sup>3-</sup>) ions (Fig. 1).<sup>6,7</sup> Being nutrients, the oversupply of N and P species may cause eutrophication, if released to the aquatic systems.<sup>8</sup> As a result, the anaerobic effluents rarely meet the discharge standards and therefore require post-treatment processes to improve the water quality further (Fig. 2).<sup>5,9</sup> Second, a significant amount of produced methane (CH<sub>4</sub>) remains dissolved in the effluents.<sup>10–12</sup> Typically, over 60–70% of the organic carbons in wastewater is converted

into CH<sub>4</sub> and carbon dioxide (CO<sub>2</sub>), which leads to the formation of biogas generally consisting of ~55–60% CH<sub>4</sub> and ~40–45% CO<sub>2</sub> (Fig. 1). However, a significant amount of produced CH<sub>4</sub> (biomethane) remains dissolved in the liquid phase (effluent). According to previous reports, dissolved CH<sub>4</sub> level may reach up to ~45% of the total production even under conditions favoring the gaseous form of CH<sub>4</sub> (e.g., at 303 K and atmospheric pressure).<sup>10,11,13</sup> The loss can further increase if the effluents are subjected to low-temperature operations, which increase the solubility of CH<sub>4</sub> in water, or low-mass-transfer conditions that lead to oversaturation.<sup>11,13,14</sup> From the economic standpoint, discharging these effluents means a loss of valuable energy resource, which can otherwise be recovered and combined with the biogas stream for increasing the overall energy efficiency of the wastewater treatment processes.<sup>10–12</sup> Notably, the wastewater industry also faces intense financial pressures and remains partially dependent on government compensations and public funds to run the anaerobic wastewater treatment plants.<sup>4,15</sup> Besides, being a potent greenhouse gas, releasing undue CH<sub>4</sub> to the environment exacerbates the global warming problem.<sup>16,17</sup> Hence, it is essential to rethink the current processes used for post-treating anaerobic effluents so as to move anaerobic wastewater treatment toward a more sustainable future.

Membrane-based technologies are attractive alternatives to many of the conventional post-treatment processes (Fig. 2), owing to their advantages such as cost-effectiveness, energy-efficiency as well as operational robustness and versatility. In the current status, the membrane-based post-treatment technologies implemented in anaerobic processes are largely focused on final polishing and nutrients (N and P) removal. Strategically speaking,

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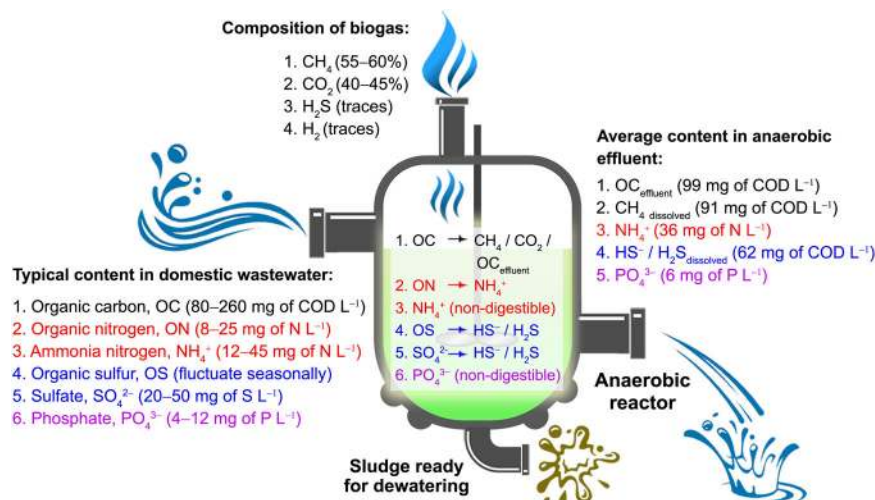
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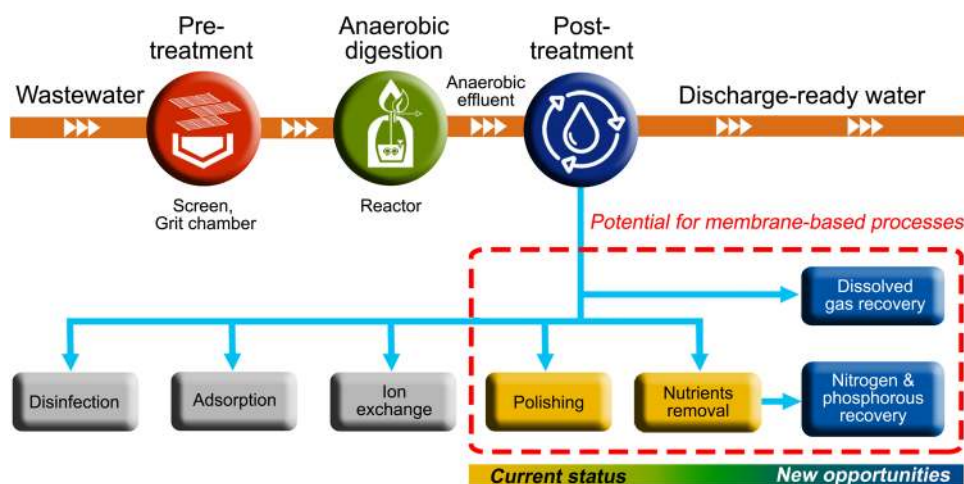
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**Fig. 1** Conceptual layout of wastewater treatment technologies based on anaerobic digestion processes. Prepared based on the information obtained from refs. <sup>5,18,19</sup>



**Fig. 2** Wastewater treatment based on anaerobic digestion and various post-treatment methods deployed for the processing of effluents. The highlighted post-treatment tasks (dashed red box) have compelling opportunities for membrane-based processes and thus elaborated in this review. Prepared based on the information obtained from refs. <sup>6,28</sup> (Note that this general outline also applies to the process designs where certain aerobic treatments are applied prior to the anaerobic digestion step.)

we believe that membrane-based technologies can offer more compelling opportunities for dissolved  $\text{CH}_4$  and nutrients recoveries to help address the challenges of the current anaerobic wastewater treatment processes mentioned above. As such, we focused our discussions on two topics: (1) New opportunities for enhancing resources (i.e., dissolved  $\text{CH}_4$  and nutrients) recovery using membrane-based processes, and (2) current state-of-the-art membrane-based processes for nutrients removal and polishing.

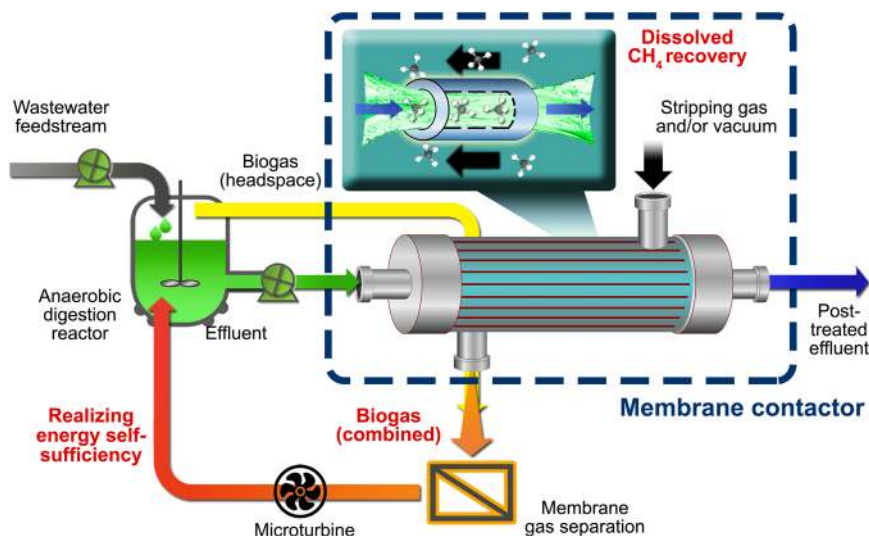
While a previous review dealing with a similar topic is coming from the perspective of the membrane-based processes for wastewater nutrient recovery,<sup>20</sup> we are more concerned with the post-treatment needs point-of-view to evaluate the utility of membrane-based processes for enhancing the process efficiency of the anaerobic digestion. Also, our scope not only covers the effluents arising directly from anaerobic treatment of raw wastewaters but is also relevant to the effluents of anaerobically digested sludge concentrates which are rich in recoverable nutrients such as phosphorus.<sup>21</sup> Lastly, we provided our perspectives to help guide future research directions for improving the feasibility of membrane-based post-treatment technologies in more sustainable anaerobic wastewater treatment processes.

## RESOURCES RECOVERY

Methane ( $\text{CH}_4$ ) and nutrients (N and P) are valuable resources that can be recovered to improve the overall cost-competitiveness of the anaerobic wastewater treatment processes. In particular, achieving net positive energy production from anaerobic wastewater treatment processes is a longstanding vision,<sup>22</sup> which can be potentially realized by recovering  $\text{CH}_4$  via a membrane contacting process, for example. As for N and P recovery, a number of membrane-based processes have also been utilized. However, due to their low concentrations, it is economically less feasible to recover nutrients using membrane-based processes. Our discussion in this section is therefore galvanized toward the strategic use of membrane-based processes for concentrating nutrients.

### Recovery of dissolved $\text{CH}_4$

Produced  $\text{CH}_4$  from anaerobic processes can be used to generate electrical and thermal energies, which can be channeled back to the anaerobic digestion processes to realize energy self-sufficient wastewater treatment plants (Fig. 3). At present, conventional physical processes such as spray aeration, jet tower, packed



**Fig. 3** Illustration showing retrofitting of the membrane contactor unit within the anaerobic digestion and membrane separation processes

column, or diffused aeration have been applied for the recovery/removal of dissolved  $\text{CH}_4$  from the anaerobic effluents.<sup>12</sup> A critical drawback of these techniques is that the stripping gas (usually air), which directly contacts the anaerobic effluents, can cause foaming and flooding. To prevent such operational problems, the use of gas/liquid “membrane contactor” process was proposed.<sup>12</sup> The gas/liquid membrane contacting process uses a stripping gas (or vacuum) to establish a mass transfer driving force between the gas and liquid phases, which are isolated using polymeric membranes as phase barriers.<sup>12,23</sup> To reduce the mass transfer resistance, membranes are commonly designed to have high porosity and low wettability. Typically, small-diameter hollow fiber membranes are used in membrane contactors to provide a much higher mass transfer area per unit volume (Fig. 3). As a result, it is possible to lower the energy consumption of gas/liquid membrane contactor process as compared to that of the conventional separation processes.<sup>12,23</sup>

Several groups have studied the influence of membranes and operation modes on the  $\text{CH}_4$  recovery from anaerobic effluents using membrane contactors. In 2011 and 2012, Bandara et al.<sup>24,25</sup> reported two consecutive studies on the performance of degassing membranes (porous polyurethane layer sandwiched between dense polyethylene layers) for the treatment of a bench-scale upflow anaerobic sludge blanket (UASB) reactor effluent. In their first study, they operated the UASB reactor for 170 days and recovered dissolved  $\text{CH}_4$  from the effluents intermittently.<sup>24</sup> As they have studied different temperatures, hydraulic retention times (HRTs), and transmembrane pressure (TMP) conditions, their  $\text{CH}_4$  recovery efficiencies fluctuated widely, but typically reached around 70 to 90%. In their follow-up work, Bandara et al. operated the UASB reactor for 18 months under ambient conditions and found that the  $\text{CH}_4$  recovery reached “57%  $\pm$  7%” and “66%  $\pm$  8%” at warmer (20–31 °C) and colder (6–10 °C) seasons, respectively.<sup>25</sup> In another study published in 2012, Cookney and coworkers have tested the potential of dense polydimethylsiloxane (PDMS) membrane contactors for  $\text{CH}_4$  recovery from expanded granular sludge bed (EGSB) anaerobic reactors.<sup>26</sup> They found that ~45% of the produced  $\text{CH}_4$  remained in the effluents, and 72% of this can be recovered by using a low liquid velocity (0.0025 m s<sup>-1</sup>) during the membrane contacting process.

Membrane pore wetting is a severe limitation of the membrane contactors. Pore wetting phenomenon occurs when the liquid phase enters the pore of the membranes, leading to a tremendous increase in mass transfer resistance. Henares et al.<sup>27</sup> recently

investigated the performances of porous polypropylene (PP) and dense PDMS membranes for dissolved  $\text{CH}_4$  recovery and found that the fluxes of porous PP membranes were much higher than the dense PDMS membranes. Despite this, porous PP membranes suffered from wetting problems with increased liquid flow rate. As such, Cookney et al.<sup>29</sup> suggested that dense membranes are more suitable as they have a higher capacity to obviate the pore wetting phenomenon. Pore wetting can also be prevented by enhancing the surface hydrophobicity of the membranes. For instance, Wongchitphimon et al.<sup>30</sup> incorporated fluorinated silica nanoparticles on the surface of porous Matrimid<sup>®</sup> membranes. The resulting composite membranes demonstrated a high contact angle of ~126°, which resulted in at least 1.75 times higher performance than a commercial PP membrane for the  $\text{CH}_4$  recovery application. Later, Dilhara et al.<sup>31</sup> evaluated the performance of poly(vinylidene difluoride) (PVDF) membranes that were surface-functionalized with a solution comprising 3 wt% commercial perfluoropolyether, Fluorolink S10, and 2 wt% tetraethoxysilane (TEOS). The surface-modified PVDF membrane exhibited almost two times higher  $\text{CH}_4$  flux and had a stronger resistance toward pore wetting (more than 10 days) as compared to the commercial PP membrane.

Apart from the membrane aspects, the operation mode of membrane contactors is also critical for achieving a high recovery of dissolved  $\text{CH}_4$ . Comparing the two operation modes, i.e., feeding the liquid phase in lumen or shell side of the membrane module, researchers have found that feeding the liquid in lumen side gives better results, owing to more effective cross-flow hydrodynamics which suppress the formation of dead zones.<sup>27,29</sup> However, using the lumen side of the membrane modules increases the risk of clogging when the effluents contain a high solid content. In view of this potential disadvantage, processing the effluents in the shell side of the membrane modules is still the preferred operation mode unless the total soluble solids (TSS) is sufficiently low.

To improve the quality of outlet gas from the membrane contactors, careful optimization of the operating parameters is needed. For example, a trade-off between % $\text{CH}_4$  removal efficiency and concentration of  $\text{CH}_4$  in the gas outlet of the membrane contactor can occur due to the additional stripping gas supplied to increase the mass transfer driving force. While this approach can increase the % $\text{CH}_4$  removal efficiency, it dilutes the  $\text{CH}_4$  concentration in the outlet gas inevitably.<sup>32</sup> To address this problem, McLeod et al.<sup>32</sup> deliberately lowered the stripping gas

flow rate to obtain a gas that possessed a  $\text{CH}_4$  concentration higher than the upper flammable limit and in a readily usable form. Besides, as a highly versatile unit, the membrane contactors allow easy retrofitting to other sources of biogas to mix with the outlet gas and afford a product gas that has on-demand  $\text{CH}_4$  concentration.<sup>12</sup>

Membrane fouling is another critical issue that results in compromised recovery of membrane contactors but not actively explored hitherto. Among the limited number of studies reported on fouling of membrane contactors so far, only Bandara et al.<sup>24,25</sup> noticed an insignificant level of membrane fouling after a ~5.5- and 18-month long investigation using one type of anaerobic effluent. However, when another type of anaerobic effluent was used, Henares et al.<sup>33</sup> observed membrane fouling on PDMS membrane contactors. The removal efficiency remained constant for 120 h before suffering from a fouling-driven flux decline, which lowered the initial flux to ~40% after 175 h. Although the fouling was reversible (the performance was restored after physical cleaning by water), we believe that a deeper understanding of membrane fouling and its mechanisms are essential for the future application of membrane contactors in dissolved  $\text{CH}_4$  recovery.

#### Nitrogen (N) and phosphorous (P) recovery

Ammonium nitrogen ( $\text{NH}_4^+\text{-N}$ ) and phosphate phosphorus ( $\text{PO}_4^{3-}\text{-P}$ ) do not break down completely during the anaerobic processes and remain in the anaerobic effluents with average values of 36 and 6  $\text{mg L}^{-1}$ , respectively.<sup>18</sup> In this age of depleting natural resources, it is strategic to recycle valuable resources from the effluents, especially in recovering N and P from anaerobic effluents as high-value products such as fertilizers. To date, direct N and P recovery using membrane-based processes are economically less feasible due to low concentrations of nutrients in the effluents.<sup>34</sup> As such, recent focus is on concentrating nutrients prior to their recovery. Several membrane-based processes have been utilized for this purpose and shown promising results. Their basic principles, reported performances, advantages/disadvantages as well as other membrane processes for direct recovery of N and P are summarized in Table 1.

Forward osmosis (FO),<sup>35</sup> reverse osmosis (RO),<sup>36</sup> nanofiltration (NF), and membrane distillation (MD) are competitive candidates for concentrating N and P compounds in the effluents while producing clean water as a permeate. Among them, FO emerges owing to its energy-efficiency since it operates under ambient pressure, unlike RO.<sup>37,38</sup> It was experimentally determined that a seawater-driven FO process effectively concentrate  $\text{PO}_4^{3-}\text{-P}$  (2.3-fold) and  $\text{NH}_4^+\text{-N}$  (2.1-fold) from treated municipal wastewater. Furthermore, the concentrated nutrients can theoretically reach more than tenfold concentration by using a draw-to-feed solution-volume ratio of 2:1.<sup>8</sup> Although clean water is not the final product in this case, the FO process can be coupled with another membrane-based process to realize hybrid FO-based process that has the capacity to produce clean water. For instance, an FO-MD hybrid process demonstrated the feasibility of recovering clean water from a draw solution that was diluted by water extracted from the digested sludge centrate.<sup>39</sup> In addition, the FO process is strategically appropriate for P recovery from the effluents of digested sludge centrates which have high fouling propensity. A study by Ansari et al.<sup>21</sup> revealed that although a 30% decline in water flux occurred as the digested sludge was concentrated by threefold, the water flux was almost recovered to the initial flux by simply flushing the membrane with deionized water owing to reversible fouling observed in the FO process. As a result of the concentration of the effluent, P recovery by precipitation was able to reach 92%. Such a high enrichment also offers the advantage of reducing the incidental expense in terms of storage and transport of the effluent. Moreover, the enriched solution suggests the possibility of using the FO process to facilitate existing methods to

recover nutrients, such as struvite ( $\text{MgNH}_4\text{PO}_4\cdot 6\text{H}_2\text{O}$ ) precipitation, which are less viable when implemented alone. At present, struvite precipitation is known to be the most effective among current methods for P removal when the P concentration is over 100  $\text{mg L}^{-1}$  (higher than average value in anaerobic effluents).<sup>20</sup> In this regard, membrane-based processes can be a cost-effective solution to reduce the water content of the effluents for an efficient nutrient recovery.

For direct recovery of  $\text{NH}_4^+\text{-N}$  from anaerobic effluents, the membrane contactors and electro dialysis are the most commonly utilized techniques. Membrane contactors can recover ammonia ( $\text{NH}_3$ ) from  $\text{NH}_4^+\text{-N}$  at high pH, which can then be used to produce biofertilizers such as ammonium sulfate ( $(\text{NH}_4)_2\text{SO}_4$ ) by separately reacting the recovered N with a sulfuric acid solution. It is noteworthy to mention that the membrane contacting process for producing such biofertilizers is a more cost-effective method compared to air stripping or chemical precipitation. Generally, the membrane contactors expand an operating cost (power and chemicals) of USD 2.15 per  $\text{m}^3$  feed,<sup>40,41</sup> whereas air stripping and chemical precipitation demand a higher USD 3.80 and 13.0 per  $\text{m}^3$  feed,<sup>42</sup> respectively. On the other hand, electro dialysis using ion exchange (IE) membranes is also a promising alternative owing to the following strengths. First, they show high treating capabilities of nutrient streams of low concentrations.<sup>43</sup> Second, they are simple to operate and do not require the use of chemicals.<sup>44</sup> Lastly, they have shown to be resistant toward a sudden spike in the composition of the anaerobic effluents.<sup>44</sup> Recently, Wang et al.<sup>45</sup> coupled the IE resin with membrane capacitive deionization (MCDI) to reduce the detrimental effect of prior cations ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) on the IE adsorption-desorption process. This hybrid process improves the ion exchange capacity and can cover about 2.5 times more inflow than a single IE process after three MCDI charge-discharge cycles, leading to an improved recovery of more than 65% of  $\text{NH}_4^+\text{-N}$ .<sup>45</sup> Despite these promises, membrane contactors require both temperature and pH adjustments,<sup>46</sup> while electro dialysis is impeded by a high energy consumption (e.g., 3.25–3.60  $\text{kWh N-NH}_3 \text{ kg}^{-1}$ ), resin regeneration, and membrane fouling issues.<sup>43</sup> These challenges, if not resolved, will make direct recovery of N and P via membrane-based processes economically less feasible.

#### ENHANCING WATER QUALITY OF ANAEROBIC EFFLUENTS

Although nutrients recovery is highly promising as discussed previously, N and P removals from anaerobic effluents, by means of biological processes, remain the mainstream processes as their operating costs are relatively cheap and much less chemical sludge is produced.<sup>47,48</sup> Despite nutrients being recovered or removed, the quality of the anaerobic effluents may not be high enough to meet recycle or discharge standards. Thus, polishing is often required as the final step. In this part, opportunities for membrane processes in these two processes are reviewed.

#### Membrane-coupled bioprocesses for nutrients removal

Recently, a biological process known as the anaerobic ammonium oxidation (or anammox) and its related technologies, such as complete autotrophic nitrogen removal over nitrite (CANON, one stage anammox process), have emerged as powerful techniques for N removal. These biological processes are useful for the treatment of anaerobic effluents with low organic carbon per nitrogen mole ratio (OC/N),<sup>49,50</sup> unlike the conventional nitrification and denitrification processes which demand an additional organic carbon source for the effective removal of N. However, due to the nature of the anammox microorganisms, the conventional bioreactors typically require several months to start-up and have a huge footprint.<sup>51</sup> These limitations can all be addressed by membrane processes which can offer either a

**Table 1.** Membrane-based processes for the recovery of N and P species

Membrane processes	Basic principles	Target species	Membrane required	Reported efficiency	Advantages	Disadvantages	Ref.
FO	Water activity in the effluent is higher than in the draw solution, therefore, water can diffuse from the effluent across the semi-permeable membrane to the draw solution while rejecting other species, including N and P.	Water	Dense membranes with high salt rejection properties.	>98% N rejection >88% P rejection	<ul style="list-style-type: none"> <li>High solute rejection</li> <li>Low fouling tendency and energy saving due to the operation under no hydraulic pressure</li> </ul>	<ul style="list-style-type: none"> <li>Additional treatment to separate a draw solute from the diluted draw solution</li> <li>Possibility of salt leaking to feed solutions</li> </ul>	8,35,39
MD	Vapors from a hot feed effluent transfer across the porous membrane to a cold permeate side while N and P are rejected.	Water	Hydrophobic porous membranes	>89% N rejection P rejection: not reported	<ul style="list-style-type: none"> <li>Suitable for effluent of thermophilic anaerobic processes</li> <li>High solute rejection</li> </ul>	<ul style="list-style-type: none"> <li>Continuing heat loss lowers process performance</li> <li>Low water flux</li> </ul>	52
NF and RO	Feed solutions are pressurized, and small N and P compound molecules were filtered by membranes.	Water	Membranes with selective layers containing subnanometer pores	>94% N rejection >97% P rejection	<ul style="list-style-type: none"> <li>High solute rejection</li> <li>Simple, and compact</li> </ul>	<ul style="list-style-type: none"> <li>Fouling could be severe</li> <li>Membrane cleaning is required</li> <li>High energy required for pumps</li> </ul>	53,54
MCDI-IE	Several cations are adsorbed on a porous electrode by applying an electric field in the pre-treatment using the MCDI. After that, $\text{NH}_4^+$ is recovered by adsorption during an ion exchange process.	N in ionic forms ( <i>i.e.</i> , $\text{NH}_4^+$ )	IE membranes	>65% N recovery <sup>a</sup>	<ul style="list-style-type: none"> <li>Ammonium removal independent of biological treatment</li> <li>Less detrimental effect of prior cations (<math>\text{Ca}^{2+}</math> and <math>\text{Mg}^{2+}</math>) on the IE process</li> </ul>	<ul style="list-style-type: none"> <li>Limitation of charge efficiency in CDI<sup>55,56</sup></li> <li>Scaling possibly caused by precipitation of soluble salts<sup>57</sup></li> </ul>	45
MC <sup>b</sup>	An alkali is put into the anaerobic digestion effluent to convert N compounds to gaseous $\text{NH}_3$ which can be diffused across membranes to dissolve and react with $\text{H}_2\text{SO}_4$ solution feeding from the other side of the membrane.	$\text{NH}_3$	Hydrophobic porous membranes	>70% P removal	<ul style="list-style-type: none"> <li>Highly selective to <math>\text{NH}_3</math></li> <li>Compact process</li> </ul>	<ul style="list-style-type: none"> <li>Cost for alkali addition</li> <li>Absorbent needed to be regenerated</li> </ul>	58

<sup>a</sup> Combined with an IE resin process; <sup>b</sup> Membrane contactor

separation unit to concentrate the anaerobic effluents (such as membrane bioreactors (MBR)) or form a support layer allowing growth of desirable biofilms (such as in membrane biofilm reactors). These membrane processes provide a better control over the cultivation of microorganisms as well as allow a higher quality of effluent and a smaller bioreactor footprint.<sup>67</sup> For P removal, MBR coupled with a biological process known as enhanced biological phosphorous (Bio-P) have demonstrated good P removal efficiency for tertiary wastewater treatment.<sup>68</sup> However, no direct work on the treatment of anaerobic effluents has been reported for these processes so far to our knowledge.

#### Membrane-based processes for polishing step

Anaerobic effluents usually require polishing to further reduce organic matters, suspended solids, and colorations remaining from the biological process before being discharged or subjected to disinfection to remove harmful bacteria and viruses. Being the last step, the process should deliver both high consistency and superior separation performance. Thus, membrane-based filtration techniques are of importance for modern polishing practices.

Membrane filtrations such as microfiltration (MF), ultrafiltration (UF), NF, and RO are pressure-driven processes that use a membrane as a physical barrier to retain large-molecular-weight contaminants while allowing water and low-molecular-weight substances (lower than their cut-off) to pass through. Therefore, the water quality after post-treatment depends critically on the type of membrane-based processes used. As compared to the conventional biological methods for polishing, membrane filtrations have a clear advantage in terms of their simplicity. On the other hand, biological processes such as polishing ponds are usually constrained by their large footprints while other filtration techniques such as sand filtration are much more complex and can be problematic when it comes to cleaning.<sup>72</sup>

Several studies have utilized membrane-based polishing for the post-treatment of anaerobic effluents (Table 2). Particularly, MF demonstrates an almost complete rejection of *E. coli* and *Streptococci* bacteria from anaerobic effluents, making the treated water suitable for non-potable reuse applications.<sup>59,63</sup> NF and UF are also effective in removing emerging pollutants from anaerobic effluents, which are not commonly monitored but can cause adverse environmental effects. For instance, Wei et al.<sup>73</sup> removed fifteen organic micropollutants from an AnMBR effluent using NF and UF, and the results showed that NF obtained an average removal efficiency of 87%. Five organic acids, which were found in high concentrations in a range of 50–500 mg L<sup>-1</sup> in the AnMBR effluent, were treated by NF membranes.<sup>74</sup> Furthermore, through pH adjustment of the feed solution, removal rates can reach 90% and above.<sup>74</sup>

Membrane fouling remains the most pervasive issue to be addressed before any membrane filtration process can be practically implemented as the polishing step. In this context, a pre-treatment step to remove foulants is an effective way to mitigate fouling and prolong membrane lifetime. Addition of coagulants<sup>66</sup> and application of an acid treatment<sup>62</sup> have been successfully demonstrated to remove negatively charged substances (the main foulants) from the anaerobic effluents. Alternatively, membranes with less fouling propensity have been employed for pre-treatment followed by a downstream membrane-based polishing. For example, Debik et al.<sup>65</sup> and Tam et al.<sup>59</sup> authenticated the feasibility of hybrid systems consisting of UF-NF and MF-RO, respectively, for the treatment of anaerobic effluents. Both studies reported excellent qualities of the polished anaerobic effluents as compared to a standalone membrane filtration process.

## CONCLUSIONS AND FUTURE PERSPECTIVES

The prevention of product losses and recovery of reusable resources, coupled with affordable removal of unwanted wastes, may increase the overall profitability and further improve the environmental friendliness of anaerobic processes. In this review, we argue that membrane-based post-treatments have the potential to play a definitive role by leveraging their small carbon footprint, competitive or superior separation performance, and high design flexibility to help reduce both investment and operation costs of post-treatment processes. Particularly, membrane-based processes enable new opportunities in resources recovery, which can drive the realization of energy self-sufficient anaerobic wastewater treatment processes as well as the production of high-value products such as biofertilizers. The state-of-the-art membrane-based post-treatment processes for nutrients removal and polishing have also been reviewed.

We found that, despite some promising results, most of the membrane processes for the treatment of anaerobic effluents are still in lab-scale operations and substantial works remain to be done before these processes can be scaled up into full-fledged operations of which capacities are as large as several thousand m<sup>3</sup> day<sup>-1</sup>. Herein, we focus on the strategies that can significantly improve the membrane processes for the anaerobic effluent treatments, including (1) development of membranes, (2) strategies for biofouling mitigation, and (3) transition from the lab- to real-scale operations.

#### Membrane development

For all membrane-based processes, whether used for recovery or removal, the core of the technology lies in the membrane unit itself. Hence, we contend that continuous efforts ought to be put into developing optimal membranes. For CH<sub>4</sub> recovery using membrane contactors, an ultimate goal is to develop hydrophobic hollow fiber membranes with enhanced antifouling properties and long-term resistance toward pore wetting. This is not easy to achieve since highly hydrophobic membranes capable of preventing pore wetting are at the same time vulnerable to membrane fouling. One potential solution is to develop composite membranes with a thin and dense hydrophilic layer over a hydrophobic membrane substrate. Materials such as poly(2-hydroxyethyl methacrylate) and poly(ether block amide) are possible candidates for the preparation of a hydrophilic layer.<sup>75,76</sup> First, such dense hydrophilic layers can prevent liquid water from entering the membrane pores by acting as a physical barrier. Second, they are able to form thin films even on MF membranes, which are generally difficult to achieve *via* dip- or spin-coating.<sup>75,76</sup> Alternatively, introducing patterns on the surface of hydrophobic membranes can induce turbulent hydrodynamics and high shear stress at the membrane surface to reduce the deposition of foulants. The key advantage here is that the antifouling properties are enhanced without the use of hydrophilic surface modification (Fig. 4a).<sup>69,77,78</sup> Interested readers can refer to discussions on various fabrication methods (e.g., soft<sup>79</sup> or nanoimprint lithography<sup>80</sup>) and several configurations (e.g., flat-sheet<sup>81</sup> or hollow fiber<sup>82</sup> type membranes).

Next-generation membrane development is also focused on membranes with high throughput, good rejection in a single pass, and antifouling properties against both organic and biological foulants. Among those characteristics, antifouling properties are highly demanded in membrane processes for biological N and P removal and polishing because the anaerobic effluents contain both dissolved and suspended solids, which are likely to cause pore blocking or gel layer formation.<sup>83</sup> It is noted in this regard that the surface modification using nanomaterials is one of the most promising approaches to impart antifouling properties. To date, surface-modified composite membranes have been prepared *via* various approaches such as direct coating, surface

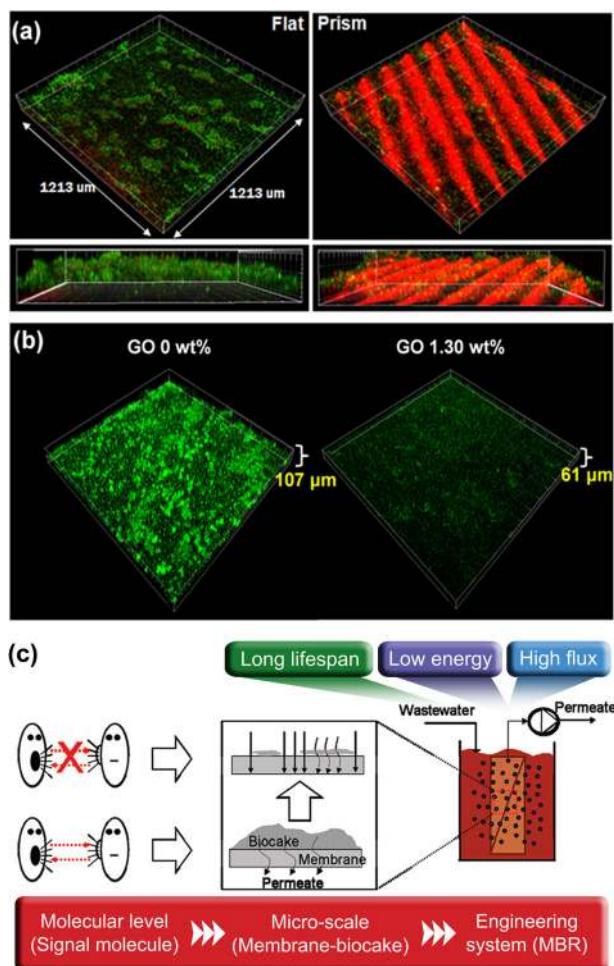
**Table 2.** Membrane-assisted technologies employed for the polishing of anaerobic effluents

Ref.	Anaerobic effluent source	Membrane process	Treated species and inlet concentration	Reported efficiency (%)	Operating conditions	
Tam et al. <sup>59</sup>	MBR	RO	COD 17.5 mg L <sup>-1</sup>	88.6	Pilot-scale study RO flow rate 19 m <sup>3</sup> day <sup>-1</sup> MBR flow rate 40 m <sup>3</sup> day <sup>-1</sup> Pressure not reported	
			Total nitrogen 3.7 mg L <sup>-1</sup>	91		
			Total dissolved solid 337 mg L <sup>-1</sup>	87.5		
			Apparent color 37 Hazen unit	97		
			Conductivity 0.57 mS cm <sup>-1</sup>	95.3		
			<i>E. coli</i> 3.4 CFU per 100 mL	100		
			Viruses 1.2 PFU per 100 mL	100		
Andrade et al. <sup>60</sup>	MBR	NF	Estrogens 38.2 (µg L <sup>-1</sup> )	88	Feed flow rate 5.6 L min <sup>-1</sup> Pressure not reported	
			TOC 24.9 mg L <sup>-1</sup>	97.4		
			Total solid 1.482 mg L <sup>-1</sup>	64.8		
			Color 36.8 Hazen unit	82.7		
Jacob et al. <sup>61</sup>	MBR	RO	Conductivity 2.28 mS cm <sup>-1</sup>	89.3	Batch operation, size 500 mL Inlet pressure 8 bar	
			TOC 7 mg L <sup>-1</sup>	96		
			Conductivity 0.76 mS cm <sup>-1</sup>	97.5		
Grundestam and Hellstrom <sup>62</sup>	MBR	RO	TOC 7 mg L <sup>-1</sup>	93.5	Batch operation, size 1 m <sup>3</sup> Pressure 40 bar	
			Conductivity 0.7 mS cm <sup>-1</sup>	93		
			TOC 17 mg L <sup>-1</sup>	88		
Al-Malack and Anderson <sup>63</sup>	EGSB	MF	Kjeldahl-nitrogen 67 mg L <sup>-1</sup>	90.6	Permeate flow rate 0.27 L min <sup>-1</sup> Pressure not reported	
			Total P 10	99.6		
			COD 120 mg L <sup>-1</sup>	87.5		
			SS 35 mg L <sup>-1</sup>	91.4		
			PO <sub>4</sub> 2 mg L <sup>-1</sup>	86.5		
Fernández et al. <sup>64</sup>	EGSB	UF	<i>E. coli</i> 1.9 × 10 <sup>3</sup> CFU mL <sup>-1</sup>	99.8	Pressure 1.3 bar Permeate flow rate 4.9 L min <sup>-1</sup>	
			<i>Streptococci</i> 1 × 10 <sup>3</sup> CFU mL <sup>-1</sup>	99.7		
			COD 1978 mg L <sup>-1</sup>	70–75		
Debik et al. <sup>65</sup>	SGBR	UF	COD 630 mg L <sup>-1</sup>	41	Pressure 3 bar	
			Color 400 Hazen unit	37.5		
			Conduct 9.87 mS cm <sup>-1</sup>	16		
		NF	COD 630 mg L <sup>-1</sup>	85		Pressure 10 bar
			Color 400 Hazen unit	87.3		
Duan et al. <sup>66</sup>	ASBR	UF	Conduct 9.87 mS cm <sup>-1</sup>	45.8	Constant flux at 30 L m <sup>-2</sup> h <sup>-1</sup> Pressure between 1.1 to 1.2 bar Constant pressure at 12.4 bar	
			COD 96.5 ppm	38		
			Turbidity 16 NTU	94.4		
			Conductivity 1260 mS cm <sup>-1</sup>	91.2		
			VSS 16.2 ppm	99		
		NF	TSS 86 ppm	95.3		
			COD 96.5 ppm	97		
			Turbidity 16 NTU	99		
			Conductivity 1260 mS cm <sup>-1</sup>	89.8		
			VSS 16.2 ppm	99		
TSS 86 ppm	99					

MBR membrane bioreactor, EGSB expanded granular sludge bed, SGBR static granular bed reactor, ASBR anaerobic sequencing batch reactor, VSS volatile suspended solid, TSS total suspended solid

grafting, plasma treatment, or simply adding nanomaterials into the dense rejection layer of the membranes (e.g., graphene oxide (GO) in Fig. 4b).<sup>70,84</sup> Further discussion on the strategies to develop surface-modified composite membranes with antifouling properties can be found elsewhere.<sup>85</sup> Key challenges to be solved

include feasibility issues regarding large-scale fabrications, higher investment costs, and environmental concerns arising from the use of nanomaterials as well as the long-term performance stability of such composite membranes.



**Fig. 4** **a** Confocal laser scanning microscopy (CLSM) images of a biofilm layer formed on the flat (left) and prism-patterned (right) membranes after 2 h filtration of the mixed liquor taken from an MBR at a cross-flow rate of  $0.05 \text{ m s}^{-1}$  (Reynolds number  $\approx 1100\text{--}1200$ ). Green and red colors indicate the cells and membranes, respectively. Image size:  $1213 \times 1213 \mu\text{m}^2$ . Adapted with permission from ref. <sup>69</sup>, copyright American Chemical Society 2012. **b** CLSM images of biofilm (green dots) formed on GO-free (left) or GO-incorporated (right) polysulfone membranes. Adapted with permission from ref. <sup>70</sup>, copyright Elsevier 2013. **c** Schematic illustration showing the concept of quorum sensing-based biofouling control in MBR. Adapted with permission from ref. <sup>71</sup>, copyright American Chemical Society 2009

### Strategies for biofouling mitigation

The anaerobic effluents still contain microorganisms that can reproduce and cause biofouling of membranes. Unlike organic fouling, which could be alleviated through hydrophilic modification (see “Membrane development” section above), biofouling is harder to tackle as the microorganisms can persistently foul even hydrophilic surfaces by secreting extracellular polymeric substances (EPS).<sup>71,86</sup> Thus, it is of importance to take into account the fundamental mechanisms underlying biofilm formation. Indeed, inhibiting the intercellular communication by quenching quorum sensing (a.k.a. quorum quenching) has been shown to be an effective method of suppressing biofilm formation on the membrane surfaces (Fig. 4c).<sup>71</sup> For mitigating membrane biofouling in MBRs, diverse quorum quenching approaches ranging from enzymatic<sup>71</sup> to bacterial<sup>87,88</sup> methods are available in the literature.

Improving membrane module designs is another proven strategy to reduce biofouling. Particularly, feed spacers in flat sheet membrane modules can effectively enhance mass transport properties by increasing local shear rates and velocities at the membrane surfaces, leading to less attachment and lower accumulation of the bacteria on the membrane surfaces.<sup>89</sup> For hollow fiber membranes, baffles are typically installed in the shell side of the modules to obtain turbulent hydrodynamic cross-flow velocities.<sup>90</sup> The 3D printing technology offers a fabrication resolution of  $0.1\text{--}1 \mu\text{m}$ , which can realize innovative spacers and membrane module components, including turbulence promoters, air diffusers, flow distributors, and membrane holders.<sup>91</sup> The components feature intricate and complex geometric designs that have a high potential to further improve the antibiofouling performance.<sup>91</sup> However, at present, such designs are technically challenging for conventional manufacturing approaches.

For biofouling cleaning, the use of chemical solvents is an expensive and aggressive method although it is one of the most effective approaches. It should be minimally employed or used as a last resort only in the case that physical cleaning is no longer effective in recovering the membrane performances. Although not considered an ultimate mitigation measure, the use of scouring agents is also deemed as a promising physical cleaning strategy.<sup>92</sup> It requires less energy than gas sparing techniques and is more effective in cleaning the membrane surfaces. In addition, scouring technique is applicable for several membrane processes and can be coupled with the quorum quenching techniques by using scouring media that contain quorum quenching bacteria to obtain a synergistic effect for reducing membrane biofouling.<sup>93–95</sup>

### Transition from lab-scale to real-scale operations

Several membrane technologies, such as membrane contactors, FO, and MCDI, have demonstrated great potential for post-treatment of anaerobic effluents. However, they remain to date as lab-scale investigations. For successful implementation of these membrane processes in large-scale operations, we recommend three crucial steps, namely, optimization, scale-up, and development of membrane maintenance strategies.

First, both the membranes as well as process operating parameters need to be optimized for maximizing membrane performance and minimizing energy consumption. Membranes can be optimized by tuning the morphologies and choosing the right materials to obtain high-performing membranes with the highest flux and rejection capacity. This will help to reduce the unit sizes required and further enhance the energy-efficiency of the membrane system. For process operating parameters, trade-offs are commonly observed. In a membrane contactor process for dissolved  $\text{CH}_4$  recovery, it is often found that increasing the liquid flow rate in the system can improve the mass transfer rate, which may decrease the membrane area required and lower the investment cost. However, this also leads to a higher energy penalty and an increase in operating costs. Thus, it is important to optimize all involving operating parameters to achieve effective and more economically viable processes.

Second, process scale-up should be coupled with theoretical studies as well as mathematical modelling. It is challenging to directly scale-up the membrane-based post-treatment technologies from lab- to real-scale applications due to the involvement of various parameters such as complex microbial consortia, variable effluent quality, and different environmental conditions. Thus, it is needed to optimize operating conditions in order to find possible solutions to the above variables prior to the actual implementation. Scale-up modelling in this regard has advantages in that it makes us understand key areas of focus in the upscaled process without the need to carry out the actual implementation. Mathematical modelling can also be used as a tool for energy and cost-benefit analyses, and the data obtained can be



compared with other competing technologies. In addition, it can be used to define a suitable process configuration. Specifically, the numbers of pumps per membrane modules<sup>96</sup>, numbers of membrane modules connected in series, as well as sizes of each module<sup>97</sup> can be optimized to find the best configuration with the lowest energy consumption. Through systematic theoretical studies and mathematical modelling, we can identify potential problems with the design for pilot-scale and actual-scale implementation and troubleshoot them before they are implemented.

Lastly, it is essential to develop membrane maintenance strategies to reduce the operating costs regarding membrane cleaning and replacement. It is important to identify major foulants in the anaerobic effluents for each membrane processes so that an effective cleaning protocol or necessary pre-treatment can be developed. Membrane cleaning is an unavoidable part of the membrane processes, and the cost could become overwhelming when the fouling is severe. In this regard, membrane autopsy by instrumental techniques, such as Fourier-transform infrared spectroscopy (FTIR), liquid chromatography organic carbon detection (LC-OCD) or scanning electron microscopy (SEM),<sup>98,99</sup> might be powerful tools for foulant characterization.

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## AUTHOR CONTRIBUTIONS

T.H.B. conceived the concept and provided guidance for this work. W.R. and J.L. organized the materials and drafted the manuscript. K.G. and H.E.K. revised the manuscript and provided critical suggestions during the revision.

## ADDITIONAL INFORMATION

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