# **Challenges in Biogas Production from Anaerobic Membrane Bioreactors**

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#### Abstract

Spectacular applications of anaerobic membrane bioreactors (AnMBRs) are emerging due to the membrane enhanced biogas production in the form of renewable bioresources. They produce similar energy derived from the world's depleting natural fossil energy sources while minimizing greenhouse gas (GHG) emissions. During the last decade, many types of AnMBRs have been developed and applied so as to make biogas technology practical and economically viable. Referring to both conventional and advanced configurations, this review presents a comprehensive summary of AnMBRs for biogas production in recent years. The potential of biogas production from AnMBRs cannot be fully exploited, since certain constraints still remain and these cause low methane yield. This paper addresses a detailed assessment on the potential challenges that AnMBRs are encountering, with a major focus on many inhibitory substances and operational dilemmas. The aim is to provide a solid platform for advances in novel AnMBRs applications for optimized biogas production.

**Keywords:** anaerobic membrane bioreactor, biogas production, methane, inhibitors

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## 1. Background

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## 1.1. Biogas and its sustainability

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categories of biomass to fulfill partially the world's energy needs. Biogas commonly refers to a mixture of gases produced by the biological breakdown of organic matter in the absence of oxygen, of which methane, hydrogen and carbon monoxide can be combusted or oxidized with oxygen. The energy output/ input can reach up to 28.8 MJ/MJ under favorable conditions, contributing to a very efficient use of the valuable biomass [1]. The resultant energy release allows biogas to be used as a biofuel to replace conventional fossil energy sources (coal, oil, natural gas) in power and heat production, and also as a versatile renewable energy source to fuel vehicles with lower sale price compared to diesel and petrol [2, 3]. In general, widespread installation and proper functioning of biogas production systems can provide many benefits to users and the wider community. Advantages include energy sustainability, resource preservation and environmental conservation. On the one hand, the long-term utilization of declining fossil fuels is considered unsustainable because of their limited reservoirs and non-renewable nature. Biogas derived from various biological sources can reduce the heavy dependence on these depleting natural resources, and address the energy insecurity concerns due to its renewable, widely applicable, and abundant characteristics [2, 4]. On the other hand, the valorization of the generated biogas is that it is energy efficient (a typical value for electrical efficiency is 33% while for thermal efficiency it is 45%) and environmentally friendly due to the low emission of hazardous pollutants, for example volatile organic compounds (VOCs) [5]. In terms of the current CO<sub>2</sub>-mitigation policy, biogas,

Biogas represents one of the most highly appreciated opportunities to utilize certain

as a nearly GHGs-neutral replacement for fossil fuels, can be produced from widely available renewable feedstocks, and their production barely contributes to the net carbon emission [6]. Optimistically, the rapid development of biogas production not only can reduce the world's heavy reliance on fossil fuel and thereby global energy needs, but also reduce the carbon footprint from fossil fuel utilization. This means decelerating the drift to global warming and climate change.

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## 1.2. Mechanisms of biogas production

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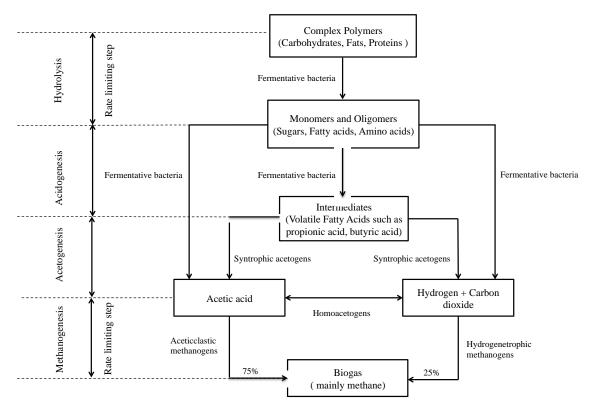
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Biogas can be produced from anaerobic digestion using the locally available residual biomass from various sources (animal waste, domestic sewage, industrial wastewater, agricultural waste, etc.). The anaerobic digestion of complex organic matter to biogas (mainly methane and carbon dioxide) involves four key steps, these being hydrolysis, acidogenesis, acetogenesis and methanogenesis (see Fig. 1.). A balanced methane fermentation process requires individual degradation phases to be carried out by distinct consortia of bacteria, namely fermentative bacteria, syntrophic acetogens, homoacetogens, hydrogenetrophic methanogens and aceticlastic methanogens. The symbiotic relationship among these microorganisms contributes to efficient anaerobic digestion and biogas production. [3,7,8]. The final phase, conducted by methane-forming bacteria, is the most crucial stage in biogas production where the methanogens convert their primary substrates including acetate, hydrogen and carbon dioxide into methane. There are two pathways for methane formation, in which approximately 75% of methane production derives from decarboxylation of acetate and the remaining 25% originates from CO<sub>2</sub> and H<sub>2</sub> [9]. The methane-forming stage is also the most sensitive and rate limiting step in the whole process since methane-forming bacteria have a much slower growth rate compared to acid-forming bacteria, and are sensitive to inhibitors such as ammonia, temperature, pH and other operational conditions. It is therefore

imperative to retain sufficient slow-growing methanogenic bacteria and prevent active biomass from being washed out from the fermenter, and to reduce inhibitory levels.



**Fig. 1.** A schematic diagram showing the comprehensive processes of biogas production from anaerobic processes [2,7].

### 1.3. Biogas production by anaerobic membrane bioreactors (AnMBRs)

The slow growing nature of methanogenic organisms and microbial complexity in the systems have made the operation of biogas fermenters difficult. The success of efficient biogas production depends on the effective retention of methanogenic bacteria in the reactor through decoupling of solids retention time (SRT) and hydraulic retention time (HRT) [10]. Research has mostly focused on retaining a high density of functioning anaerobic microorganisms, in order to achieve efficient biogas production. The most recent development in biogas production is the incorporation of anaerobic bioprocesses with

membrane separation techniques in a membrane bioreactor (MBR), the purpose being to increase biomass concentration extensively in the bioreactor. In an anaerobic membrane bioreactor (AnMBR), high cell concentrations can be sustained under reasonably high hydraulic load and sufficient mixing due to completely decoupling HRT from SRT [11].

Moreover, due to the sufficient retention of active microorganisms, AnMBRs generally have high product concentration and productivity and relatively good toxic resistance, and simplify the separation of product and/or biomass by using micro-filtration or ultra-filtration, thus leading to an improved biogas production economy [6]. The reported biogas production was well documented with the methane yield up to 0.36 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.30 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>, the volume of methane produced at 0 °C Standard Temperature and 1 atm Standard Pressure) and the high methane content up to 90% [12]. However, the optimization of biogas production from AnMBRs has not gained much attention due to the as yet under-developed nature of AnMBRs [7]. For extreme conditions, such as high salinity, thermophilic temperature, high organic loading rate (OLR) and presence of toxicity, membrane assisted anaerobic processes can be hampered and biogas productivity can be compromised.

Several review papers on biogas production (most are recent) are available in the literature. Ylitervo et al. [6] provided a general review of the MBR technology in ethanol and biogas processes and summarized the development of MBRs and the membrane technologies for these biofuels. Wang et al. [11] reviewed the progress in biogas technology in China and briefly introduced AnMBRs as one of the emerging technologies. Mao et al. [2] discussed advances in biogas production from anaerobic digestion in recent years and provided brief information on AnMBRs for biogas production. He et al. [4] summarized the recent performance of AnMBRs for methane and hydrogen production. Minardi et al. [13] reviewed membrane applications for biogas production and purification processes. While these reviews increased researchers' knowledge of AnMBRs for biogas production, they did not

simultaneously either: extensively address detailed application concerns and potential challenges encountered; or update the most recent studies. However, with the recent rapid advances in AnMBR technology for bioenergy recovery, a detailed analysis of research progress will be greatly appreciated.

The objective of this paper is to provide a comprehensive overview of such advances in various AnMBRs, in view of both traditional and advanced reactor configurations for biogas production. Moreover, with the focus on inhibitors and operational dilemmas, a detailed assessment on the potential challenges that AnMBRs are facing is included in this paper. This review provides perspectives on the outlook for an evolution in advanced AnMBRs applications for more economically feasible and productive biogas yield.

## 2. Types of AnMBRs for biogas production

#### 2.1. Conventional AnMBRs

2.1.1. Completely stirred tank reactor (CSTR) AnMBRs

CSTR is by far the most frequently researched anaerobic process in AnMBR systems for biogas production due to the ease of construction and operation. In most cases, CSTRs are in cylindrical or rectangular shapes and employ mechanical turbines for mixing. Side-stream membranes are often used, resulting in high bioreactor liquid turnover rates and a well-mixed hydraulic flow regime. The potential for biological conversion from substrate to methane can be greatly increased due to the prevailing high shear stress and intensive mixing [14, 15]. Lab- and pilot-scale studies have been carried out with all three primary AnMBR configurations: external side-stream membrane [16], submerged membrane [17], and submerged membrane with external membrane tank [18]. In general, a CSTR coupled membrane system is able to achieve a promising methane yield up to theoretical value [19]. Lin et al. [17] found a methane yield rate of 0.26 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.23 L CH<sub>4</sub> (STP)/g

COD<sub>removed</sub>) and high content of methane up to 85% when operating a pilot-scale submerged anaerobic bioreactor (SAnMBR). The compact configuration of such module also allowed more convenient biogas collection. The research on fermentative H<sub>2</sub> production is also typically conducted in CSTR-AnMBRs. Having the largest amount of energy per mass unit than any other known substance (142 kJ/g), H<sub>2</sub> is an ideal energy carrier free from harmful emissions during utilization as it is combusted to form only water. As the hydrogen production stage occurs briefly prior to the methanogenic process, biohydrogen production can be realized by inhibiting the methanogenesis phase using various intervention means. These include the manipulation of hydrogen partial pressure, pH control, chemical inhibition, and promotion of ferric-reducing conditions (the addition of FeSO<sub>4</sub> solution) [6].

Previously, most research has been carried out in an external cross-flow type, but the immersed type has become much more popular recently, due to requiring less energy consumption and less need for fouling mitigation [20]. The carbon source conversion efficiencies in the anaerobic hydrogen producing membrane bioreactor are promising with two cases reaching 100% [21, 22]. FeSO<sub>4</sub> concentration, in particular, was seen as a crucial factor impacting on the dark fermentation pathway for H<sub>2</sub> production from AnMBRs [21]. Hydrogen productivity from the AnMBRs ranged from 2.5 [23] to 66 L/L·d [24], and the hydrogen content of biogas could reach as much as 62.6% [21].

However, to ensure a well-mixed flow regime and sufficient mass transfer, rigorous mechanical mixing is required. Moreover, the disruption of particles as a result of sludge recirculation through the membrane feed pump can have a negatively impact on the orientation between acetogens and sensitive methanogens, thus limiting the essential hydrogen transport for acquiring a superior specific methanogenic activity (SMA) [14, 25]. The impaired syntrophism often leads to a higher concentration of VFA in the system and VFA inhibition is more severe particularly in the thermophilic system. Additionally, CSTR-AnMBR usually operates at a lower biomass concentration (e.g. 5 g/L MLSS) compared to

other high rate anaerobic reactors due to fouling control issues, which results in a lower OLR applied to the system, limiting the biomethane potential from high loading wastewater.

# 2.1.2. Upflow anaerobic sludge blanket (UASB) AnMBRs

The UASB concept was developed by Lettinga et al. in the 1970s for methane production. The secret of such a novel high rate reactor design lies in its ability to: firstly, retain a high concentration of biomass in the form of well settlable methanogenic sludge granules in a thick dense sludge bed at the bottom of longitudinal reactor; and secondly, capture produced biogas through a gas/liquid/solid (GLS) separator at the top. Many researchers have attempted to combine UASB with membrane to optimize the joint benefits such as enhanced methane production and less fouling problems [26, 27]. Xie et al. [28] have investigated the feasibility of a submerged UASB-MBR system for recovering energy from kraft evaporator condensate at 36°C to 38°C for 9 months. The methane production rate of 0.35±0.05 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.31±0.05 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>), which were very close to the theoretical yield of methane with 0.397 L CH<sub>4</sub>/g COD removed at 37°C (0.350 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>), and the methane content in the biogas, reached 90% in this study. Lin et al. [29] also reported a similar methane production rate and excellent fuel quality with 80-90% methane for both mesophilic and thermophilic SAnMBRs from kraft evaporator condensate treatment.

To assist with effective biomass retention, a hybrid UASB-MBR, in which fine fibers were placed at the top of the sludge zone as a biofilter, served to treat real domestic wastewater for biogas production at ambient temperature in Beijing, China [30]. Due to the sufficient retention of biomass by the membrane (MLSS maintained as high as 21.5 g/L), this hybrid Granular-AnMBR (G-AnMBR) system achieved maximum biogas production of 0.42 L/L·d, and methane gas content of 66%. UASB reactors for methane fermentation from low strength wastewater at low and moderate temperatures are often encountered by a poor mixing regime, which undermines biogas productivity due to a decrease in soluble COD treatment efficiency.

The granulation process for UASB using non-granular seeding is also very lengthy, requiring at the least a 3-month start-up period for stable biogas production. Direct Membrane intervention into UASB eliminated the hydraulic selection pressure for sound granules. This can negatively impact the granular sludge properties and hence the methane yield in the long-term [14].

### 2.1.3. Expanded granular sludge bed reactor (EGSB) AnMBRs

EGSB reactors are tall reactors characterized by a higher ratio of height to width and effluent recirculation, and they can provide a very high mixing intensity and sufficient substrate-microbes contact induced by the high upflow force. However, to date, the studies on the feasibility of the EGSB combined membrane process are limited. This is probably due to the fact that manipulating sludge bed expansion in EGSB is relatively difficult due to the absence of solid carriers under high hydraulic upflow force. The only study available was Chu et al. [31] who reported biogas production ranging from 0.28 to 0.58 L/L·d, and 63–72% methane at low HRT of 3.5 h from an EGSB reactor coupled with hollow fiber membrane filtration for energy recovery from domestic wastewater at 15°C. However, granules fragmentation and sludge washout may occur due to the applied high upflow velocity (U<sub>v</sub>>4 m/h), which may affect methane production. In addition, no granulation is expected in EGSB-MBR, which would alter the properties of granular biomass and affect the biogas production in the long-term.

#### 2.1.4. Anaerobic fluidized-bed membrane bioreactor (AFBR) AnMBRs

AFBR is regarded as an effective anaerobic process to be coupled with membrane filtration due to its good mass transfer characteristics and retention of high concentration of active microorganisms at short HRT lasting from a minutes to a few hours. Compared to other gas-sparged AnMBRs, membrane fouling was successfully controlled through the

energy efficient scouring effect of fluidized granular activated carbon (GAC) on the membrane surface, resulting in fouling mitigation having significantly reduced energy costs [32]. A two-stage anaerobic fluidized bed system is often required to fully reclaim methane from wastewaters. Kim et al. [33] proposed a staged anaerobic fluidized membrane bioreactor (SAF-MBR) system, which consisted of an AFBR and an anaerobic fluidized-bed membrane bioreactor (AFMBR) for methane rebate from municipal wastewater primary-clarifier effluent. The methane production from this system was reported as 4.11 mol CH<sub>4</sub>/m<sup>3</sup> (92.1 L CH<sub>4</sub> (STP)/m<sup>3</sup>) with methane composition of 86% at HRT less than 5h. Using only 30% of the gaseous methane energy produced could satisfy total fluidization energy required for the system, meaning such a SAF-MBR is a promising AnMBR for bioenergy production. Similarly, Yoo et al. [34] and Dutta et al. [35] also worked on the SAF-MBRs and concluded that the SAF-MBR system has excellent potential as a low-energy input, high-efficiency, and cost-effective system using methane energy. This is despite the fact dissolved methane representing 63% of the total methane production continued to represent a big issue that needs to be solved if energy production is to increase [34]. Methane production is significantly affected by the temperature, and dissolved methane, particularly in the winter period was a severe issue identified by researchers. Gao et al. [36] investigated an integrated anaerobic fluidized-bed membrane bioreactor (IAFMBR) system with simplified reactor operation and much smaller footprint compared with two-stage systems. In this study the methane content in biogas had typical value of (80±2%) and nearly 50% of the influent COD was converted into methane, of which 25% of produced methane was lost in the liquid phase. As a result of the restrained organic degradation capacity of the AFBR-AnMBRs, VFA accumulation and inhibition was found at shorter HRT, which reduced the specific methane productivity. Moreover, Shin et al. [32] reported that the specific acetoclastic methanogenic activity (SAMA) on the GAC was much lower than the enriched acetoclastic cultures, indicating low levels of such organisms in the GAC VSS. Therefore,

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future research on biogas production optimization should consider the facilitation of attached growth of syntrophic VFA-degrading acetogens and acetoclastic methanogens on GAC.

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#### 2.1.5. Jet flow anaerobic bioreactor (JFAB) AnMBRs

A jet flow anaerobic reactor has certain advantages when coupling with membrane filtration to form an AnMBR for methane fermentation. The liquid circulation inside such a reactor by using an inner tube and a nozzle system offers an adequate homogenization and mass transfer [14]. Applying an UF membrane coupled to the JFAB, Saddoud et al. [37] reported rich methane in biogas (70%) with the average methane yield being 0.27 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.24 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) from domestic wastewater treatment. However, in another study, Saddoud et al. [38] reported the inefficient methanization of such AnMBR with the average methane yield not exceeding even 0.1 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.088 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>). This was due to the considerable fluctuations in the substrate composition and presence of toxic substances emanating from industrial effluents. Saddoud and Sayadi [39], therefore proposed an innovative two-phase anaerobic digestion (TPAD) system coupling anaerobic fixed bed reactor for optimized acidogenesis and the AnMBR for optimized methanogenesis for biogas production from slaughterhouse wastewater. In this combined process, the volatile fatty acid (VFA) inhibition was successfully overcome, and biogas conversion was significantly improved with an average value of 0.31 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.27 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>). **Table 1** summarizes the key features and advantages and challenges of conventional AnMBRs for biogas production.

Table 1 Key features, advantages and challenges of conventional AnMBRs

Conventional AnMBRs	Key features	Advantages	Challenges
CSTR- AnMBRs	<ul> <li>The first generation of high rate anaerobic reactor and most employed in AnMBR studies</li> <li>A cylindrical or rectangular tank</li> <li>Mechanical mixing</li> </ul>	<ul> <li>Good substrate-sludge contact with slight mass transfer resistance</li> <li>High liquid turnover rates and well-mixed flow regime</li> <li>Enhanced biomethane potential due to prevailing high shear stress and intensive mixing</li> </ul>	<ul> <li>Rapid acidification and VFA inhibition due to continuous mixing and high shear stress</li> <li>Negatively impacted SMA</li> <li>Lower organic loading leads to lower biomethane potential</li> </ul>
UASB- AnMBRs	<ul> <li>A cylindrical or rectangular column</li> <li>Biomass retention in the form of granules</li> <li>Sufficient mixing provided by liquid upflow force and rising biogas bubbles</li> </ul>	<ul> <li>Good wastewater-biomass contact</li> <li>Superior quality of granular sludge for higher biogas production</li> <li>Significantly higher organic/hydraulic loading rates compared to flocculent sludge bed reactor</li> <li>Moderate tolerance to toxic compounds</li> <li>No mechanical mixing device required</li> <li>Reduced gas sparging demand</li> </ul>	<ul> <li>Long start-up period</li> <li>Dead space and poor mixing at psychrophilic conditions</li> <li>Elimination of hydraulic selection pressure causes granules deterioration and disintegration, and unstable biogas production</li> </ul>
EGSB- AnMBRs	<ul> <li>Tall column reactors with smaller footprint and effluent recirculation</li> <li>High upflow velocity (&gt;4m/h), very high mixing intensity, and efficient biomass-substrate contact</li> </ul>	<ul> <li>Improved mass transfer in a compact design</li> <li>Resolve issues with UASB such as hydraulic short cuts, preferential flows, poor mixing regime (dead zones), and temperature constraints</li> <li>Effective in generating biogas from soluble pollutants sources such as domestic</li> </ul>	<ul> <li>No granulation is expected, and this may affect the granule sludge properties and hence biogas production</li> <li>Granules fragmentation and sludge washout due to the high upflow velocity</li> </ul>

wastewater, and wastewater containing
lipids and toxic/inhibitory compounds

# AFBR-AnMBRs

JFAB-

AnMBRs

- Granular activated carbon as the medium for bacterial attachment and growth
- Tall column reactors with a smaller foot print and effluent recirculation
- Two stage submerged membrane configuration is most employed
- A greater surface area per unit of reactor volume
- An inner tube and a nozzle system for mixing
- Jet flow module

- Higher organic and hydraulic loading and greater resistance to inhibitors for biogas production
- Much lower capital cost due to reduced reactor volume.
- Efficient in biogas production from pharmaceuticals and suspended particles of domestic wastewater

- A sound homogenization inside the reactor
- Sufficient mixing and compact design

- Two stage systems are often required for effective biogas production
- Dissolved methane remains as a big issue
- VFA accumulation and inhibition at shorter HRT.
- Low specific acetoclastic methanogenic activity (SAMA) on GAC
- VFA inhibition causing reduction in biogas production
- Ineffective in accommodating toxics and fluctuations in the feed

# 2.2. Modified AnMBRs

# 2.2.1. Anammox AnMBRs

Anammox-AnMBR is a novel process combining energy recovery in the form of methane
with effective nitrogen management. Compared to the conventional
nitrification/denitrification process, anaerobic ammonium oxidization (Anammox) has many
advantages such as high nitrogen removal, cost effectiveness and small footprint [40]. The
Anammox reaction allows microbial oxidation of ammonium to form nitrogen gas (N2) under
anoxic conditions using nitrite as the electron acceptor [41]. Due to the complete separation of
HRT and SRT by membrane, effective domestication and cultivation of the slow growing
anammox bacteria was guaranteed in an Anammox-AnMBR. Dai et al. [42] investigated the
simultaneous methane production and nitrogen removal from concentrated municipal
wastewater by using a membrane-based process combining anaerobic digestion and
nitritation-anammox under ambient temperature. The system achieved a stable methane yield
of 0.223 L $CH_4/g$ $COD_{removed}$ (0.206 L $CH_4$ (STP)/g $COD_{removed}$ ) while a total nitrogen (TN)
removal efficiency of 81% was obtained in the sequential completely (CANON) MBR. This
study concluded that the proposed process was a sustainable approach for biogas recovery and
nitrogen removal. This research also revealed that further treatment is required to reclaim
dissolved methane released into the environment as a powerful GHG, as well as to enhance
the methane recovery efficiency.
Li et al. [40] investigated the sustainable operation of submerged Anammox AnMBR, and
found biogas sparging could greatly reduce small flocs attaching to the membrane to form a
cake layer, thereby alleviating membrane fouling. It should be noted that since nitrite and
ammonia were converted to nitrogen gas by anaerobic ammonia-oxidizing bacteria (AnAOB),
methane composition in the biogas would be lower than that of other types of AnMBRs as a
result of the accumulation of produced N <sub>2</sub> in the biogas. The interaction between AnAOB and

methane-producing bacteria is still unknown, requiring further studies to maximize the methane yield from the Anammox-AnMBR. Last but not least, the feed for the Anammox-AnMBR contains high levels of ammonia, which can inhibit the production of methane.

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## 2.2.2. Anaerobic Dynamic Membrane bioreactors (AnDMBRs)

Dynamic membrane (DM) technology is a new approach for resolving problems such as high cost of membrane modules, low membrane flux, and rapid membrane fouling encountered in conventional AnMBR processes. In an AnDMBR, the solid-liquid separation is mainly accomplished by the cake layer (e.g., dynamic membrane) formed on low cost supporting materials such as meshes and fabrics with macropores [43]. Such a DM transforms one of the most critical disadvantages of AnMBRs, namely membrane fouling, into a competitive advantage. When the dynamic membrane is seriously fouled, the cake layer can be easily removed, cleaned and then replaced by a new deposited layer, thus significantly reducing the membrane cost [44]. The cleaning process and its frequency are determined by the dynamic membrane material and chemical resistance of the filters [45]. Alibardi et al. [46] developed a bench scale anaerobic dynamic MBR using a large pore-sized mesh at 200 µm. They observed varying biogas production with maximum value of 1L/d and methane content fluctuating from 50% to 79%, which was mainly due to the variability of COD removal and HRT during operation. Low CFV (due to the use of larger pore size) contributed to the sustainable aspect of the AnDMBR by reducing high energy input and led to improved methanogenic activity by minimizing the shear stress on the biomass. Methane oversaturation in the effluent was reported in the study. Due to the higher methanogenic activities of the cake layer formed on the external dynamic membrane module, a great amount of biogas was produced by the membrane unit and subsequently released with the effluent stream.

Ersahin et al. [43] applied a monofilament woven as support material for dynamic membrane formation in the AnDMBR for biogas production from synthetic concentrated

wastewater. Average methane yields of 0.31±0.02 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.27±0.02 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) and 0.34±0.04 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.30±0.04 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) were reported at SRTs of 20 days and 40 days, respectively, which were very close to the maximum theoretical value. Methane was solubilized in the permeate. The study also revealed that strong shear stress as a result of biogas sparging might create a physical interruption on the syntrophic anaerobes associated with methane forming, based on the fact that SMAs of the bulk sludge samples were lower than those of the seed sludge at both SRTs. Xie et al. [47] achieved promising average methane yield at 0.34 L/g COD<sub>removed</sub> (0.30L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) and methane content of 70-90% using Dacron mesh (pore size =40 μm) in the AnDMBR for the treatment of raw leachate, high heavy metal concentrations, and high total ammonium concentration above 3000 mg/L. Based on the archaeal taxonomic identification, aceticlastic methanogens were the dominant functional group that produced methane, while hydrogenotrophic methanogens were eliminated at the end of the experiment when ammonium inhibition was observed.

#### 2.2.3. Anaerobic Membrane distillation bioreactors (AnMDBRs)

Membrane distillation (MD) is a thermally driven separation process in which water vapor transfers across a thermal gradient through a hydrophobic, microporous membrane such as polypropylene (PP), polyvinylidene fluoride (PVDF) or polytetrafluoroethylene (PTFE) membranes to form water [48, 49] The competitive advantages of anaerobic processes can be readily utilized when they are combined with the MD process, as the mesophilic or thermophilic operation for the methane fermentation can allow no or less heating requirement for the subsequent MD treatment [49]. AnMDBRs usually require significantly reduced footprints and provide complete retention of incoming organics and microorganisms for maximum bioconversion from waste to energy in the form of biogas [48-50]. The other highlight of the AnMDBR treatment is the complete removal of total phosphorus for the

purpose of controlling eutrophication, which has been recognized as a significant environmental and ecological concern for decades [49].

In a typical AnMDBR system, biogas can be recovered from the system for gas sparging for mixing and fouling control purposes, and additional gas can be utilized for heating and energy use [48]. However, post-treatment is required to recover ammonium nitrogen and methane dissolved in the permeate. Smith et al. [51] reported methane loss in the liquid phase from the anaerobic MBR could be as much as 30% and 50% at 35°C and 15°C, respectively, due to the fact that the solubility of methane gas decreases in response to temperature increase. In an AnMDBR, most of the methane is more likely to exist in the gas phase, thus allowing much easier methane extraction and recovery. Moreover, the dissolved methane is transported with the permeate via the slower gas diffusion process in the thermally-driven AnMDBR, whereas other AnMBRs are mostly pressure-driven, the methane gas would rapidly pass into the permeate across the porous membrane via poiseuille flow. Therefore, dissolved methane in the permeate from the anaerobic MDBR will most likely be much less than those from the other AnMBRs [48]. Xie et al. [49] hybridized anaerobic moving bed biofilm reactor (AMBBR) with the MD process for the treatment of domestic wastewater. A small quantity of biogas with methane content at 58%-72% was produced from the AMMBR while no other biogas data was available from the MD process. Further research regarding biogas production from the AnMDBR and the effects of MD process on the methane-producing species would be valuable so that the benefits of AnMDBR in the sense of bioenergy recovery can be fully explored. It would also be possible to identify possible challenges in biogas production from AnMDBRs.

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2.2.4. Anaerobic osmotic membrane bioreactors (AnOMBRs)

AnOMBRs constitute a novel integrated system combining AnMBRs with the forward osmosis (FO) process for effective retention of smaller sized contaminants and prolonging of

their residence time in the reactor, thus leading to improved biodegradation efficiency and biogas yield in one integrated system [52]. In an AnOMBR the FO membrane is usually used, in which water flows from a low-osmotic- pressure feed solution (FS) to a high-osmotic-pressure draw solution (DS) across a semi-permeable membrane. One of the greatest advantages of the FO process is that no energy input is required to drive the filtration process as compared to traditional energy-intensive pressure-driven separation processes such as MF/UF. Gu et al. [52] evaluated the extent of energy recovery in the form of methane gas from an AnOMBR when treating low-strength wastewater at mesophilic temperature. A promising methane production of 0.25–0.3 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.22–0.27 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) was obtained although a loss of methane in the effluent and a high salinity environment (10 mM–200 mM NaCl equivalent) was discovered in the system. Although the salt, alkalinity and ammonia accumulations in the reactor were reported to have no effects on the bioactivity and biogas production, a long-term examination of salt inhibition, pH stability and ammonia inhibition on the biogas production still requires a further assessment.

Chen et al. [53] demonstrated the feasibility of energy recovery from a FO-AnMBR system and reported an average methane yield value of 0.21 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.19 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) with methane content of 65-78%. Nevertheless, the methane yield represented only 58% of the maximal theoretical value due to the loss of methane dissolved in the permeate as well as the inhibition of methanogenic activity under accumulated salinity environment. They also pointed out that under high osmotic conditions, anaerobic biomass tended to consume substrate to produce compatible solutes through osmoregulation and extracellular polysaccharides in order to survive. Therefore, a further investigation of salinity's effect on the microbial kinetics and methanogenic activity is required to optimize the biogas production rate. Other challenges associated with the FO-AnMBR were the disposal of inorganic-rich supernatant, and the membrane's low tolerance to high temperature solution and biological attachment to sustain stable biogas production in the long-term.

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2.2.5. Anaerobic membrane sponge bioreactors (AnMSBRs)

For an AnSMBR, the medium for bacterial attachment and growth is low-cost polyurethane sponges. These sponges represent a viable mobile carrier in many MBR technologies due to their high porosity and endurance, which can immobilize microorganisms and remove organics and nutrients effectively. Their sound mechanical features in relation to membrane scouring are another advantage to counter-attack membrane fouling due to the continuous rubbing behavior of the moving media. Kim et al. [54] investigated both single and two\_stage sponge-submerged AnMBRs using an anaerobic rotary disk MBR (ARMBR) without the membrane cleaning and replacement. They found that disk rotation contributed to enhanced shear force and mass transfer of media, and led to the effective collision between the sponge and membrane surface, thus successfully alleviating fouling and enhancing the membrane permeability in the ARMBR. Apart from the disk rotation, sponges were utilized to maintain microbial growth in the mobile phase as well as effectively control membrane fouling. The reported methane production yield and methane composition in the single system were 12% and 13% higher than those of two-stage systems. Therefore it was suggested that the single ARMBR process was superior to the two-stage process due to higher energy production in the more simplified configuration of the single system. Further research on the effects of sponge size, density and shape, and shear stress of the disk rotation on the bioactivity and biogas production are very much appreciated.

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# 2.2.6. Gas-lifting AnMBRs (Gl-AnMBRs)

Gl-AnMBR is considered to be an advanced hybrid treatment process that combines anaerobic bioprocess with low-pressure membrane filtration. In both external cross-flow and immersed configurations of conventional AnMBRs, considerable energy input is needed for the gas scrubbing requirements of membrane. A Gl-AnMBR, instead, applies the airlift

configuration by using headspace biogas for gas lift to maintain a reasonable membrane flux with minimal energy input, thus optimizing the overall energy footprint of AnMBRs. The utilization of biogas-assisted mixing also facilitates the methane stripping from the bulk liquid, avoiding super-saturation, and allowing a minimum amount of methane dissolved in the permeate [55]. Prieto et al. [56] developed a Gl-AnMBR and evaluated its ability to recover resource from sewage. This suspended-growth bioreactor coupled to a tubular PVDF UF membrane was able to produce 4.5 L/d (0.28 L/gVSS d) biogas, which can be used for membrane scrubbing and energy recovery. Biogas injection was introduced at the bottom of this system where biogas was combined with the sludge to form a two-phase (liquid-gas) flow through the lumen of vertically-placed tubular membranes. The introduction of biogas bubbles into the membrane feed significantly reduced the membrane fouling because of the increased shear force and turbulence over the membrane surface. The ascending biogas bubbles also enhanced the sludge filterability by decreasing the feed density. Therefore, pumping cost for feed flow and permeate extraction was minimized, meaning there were less energy expenses. However, the biogas recirculation for scouring generated high shear force, which was reported to negatively impact on the SMA and subsequently compromise biogas production.

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## 2.2.7. Vibrating AnMBRs (V-AnMBRs)

Membrane fouling is typically controlled by the recirculation of biogas to create shear and turbulence on the membrane surface [57]. However, the high cost of gas pumping, the difficulty of operating gas sparging in certain cases, and its shear stress on the anaerobic microbes remain as major concerns for the gas-sparged AnMBRs. V-AnMBRs, which utilize effective vibratory shear for enhancing the shear at the membrane surface, have attracted the interest of researchers in recent years. Kola et al. [58] introduced a transverse vibration as an innovative membrane fouling mitigation strategy into a membrane coupled UASB reactor.

Based on the observations of significantly increased critical flux and more reversible fouling as compared with conventional fouling control, this study proved transverse hollow fiber membrane vibration provided alternative enhancement of mass transfer. This type of membrane vibration also created vortices in the wake of the vibrating surface, thus facilitating the permeate filtration where gas sparging was often unfavorable. By appropriately incorporating periodical backwash/relaxation with vibrational filtration, such a V-AnMBR would be a promising technology for biogas production. However, the effects of vibratory shear stress on the methane-producing microorganisms require further analysis for the optimal biogas production.

Vrieze et al. [57] investigated a novel V-AnMBR using a magnetically induced vibration membrane filtration system as the solo shear enhancement device in anaerobic digestion for fouling mitigation. The biomethanation performance and membrane fouling of the V-AnMBR was compared with a conventional SAnMBR with biogas scouring (known as NV-AnMBR). Similar CH<sub>4</sub>/CO<sub>2</sub> ratios (around 1.89) in the biogas were observed from both reactors only when treating diluted molasses wastewater but the V-AnMBR resulted in a noticeable increase in transmembrane pressure and failed to prevent the formation of a cake layer due to the absence of a mixing system. The authors also justified that V-AnMBR is still a promising technology and can be applicable if conventional mixing devices or other measures can be implemented to avoid cake layer build-up. VFA accumulation and a decline in methane production were reported when concentrated molasses were applied, which indicated the inhibitory effects of concentrated molasses on biomethanation.

2.2.8. Anaerobic bio-entrapped membrane bioreactors (AnBEMRs)

In view of fouling being the major concern in AnMBRs, the anaerobic bio-entrapped membrane reactor (AnBEMR) has been developed as an alternative to the conventional AnMBRs, particularly those with high biomass concentrations. The competitive advantage of

the entrapped biomass technique was the superior simultaneous removal of carbon and
nitrogen within a simplified single throughput bioprocess. Its robust capacity to tackle
complex organic compounds, and handle high dissolved organics loading at low suspended
biomass concentration was observed in the aerobic bioprocesses [59]. In addition, when
combining the entrapped biomass technique with the membrane, membrane fouling can be
greatly reduced since less soluble organics and suspended biomass were produced in the bio-
entrapped system.

Ng et al. [59], as the sole example in the literature, proposed a novel lab-scale anaerobic bio-entrapped membrane reactor (AnBEMR) packed with bio-ball carriers. In their study, both the traditional AnMBR and AnBEMR were tested for biogas production from pharmaceutical wastewater treatment. The authors found that the AnBEMR was able to produce around 15% more methane than the AnMBR ( $0.142 \pm 0.034 \text{ L CH}_4$ /g COD<sub>removed</sub> ( $0.130 \pm 0.034 \text{ L CH}_4$ (STP)/g COD<sub>removed</sub>) while that of the AnBEMR was  $0.159 \pm 0.035 \text{ mL}$  CH<sub>4</sub>/g COD<sub>removed</sub> ( $0.145 \pm 0.035 \text{ L CH}_4$ (STP)/g COD<sub>removed</sub>)) after a 70-day start-up period. However, both systems encountered the inhibition of methane yield due to organic overloading, high salinity conditions and accumulation of toxic organics when increasing OLRs up to  $34.0 \pm 2.7 \text{ kg COD/m}^3 \cdot \text{d}$ . Furthermore, the AnBEMR showed a longer membrane filtration operating period than the AnMBR due to the release of smaller concentrations of EPS and SMP, and lower suspended biomass concentration. **Table 2** summarizes the key features and advantages and challenges of modified AnMBRs for biogas production.

Table 2 Key features, advantages and challenges of modified AnMBRs

Modified AnMBRs	Key features	Advantages	Challenges
Annamox- AnMBRs	<ul> <li>Completely autotrophic nitrogen removal</li> <li>Anammox bacteria</li> <li>Homogeneous distribution of substrates and biomass</li> <li>Anammox bacteria in forms of flocs or granules</li> </ul>	<ul> <li>Production of Anammox bacterial as flocs or granules with high growth rate</li> <li>High nitrogen removal</li> <li>Suitable for biogas production from wastewater containing a high ammonium concentration and low COD content</li> <li>Overcome long start-up issue with the Anammox process</li> </ul>	<ul> <li>Dissolved methane</li> <li>AnAOB compete with methane-producing bacteria</li> <li>Methane composition can be altered due to the production of nitrogen gas</li> <li>Ammonia inhibition</li> </ul>
AnDMBRs	Dynamic membrane was formed on the supporting materials such as meshes and fabrics with macropores	<ul> <li>Much lower capital costs for membrane and its cleaning and replacement</li> <li>Higher membrane flux</li> <li>Reduced energy consumption and shear stress on the biomass by using low CFV</li> <li>Cope well with large OLR, high heavy metal and high ammonium concentrations</li> <li>Promising methane production due to the higher methanogenic activity</li> </ul>	<ul> <li>Methane oversaturation in the permeate</li> <li>Strong shear stress due to biogas sparging for fouling control can affect methanogenic activity</li> <li>Biogas escape from the external membrane unit and effluent collection vessel</li> <li>Ammonium inhibition on hydrogenotrophic methanogens</li> </ul>
AnMDBRs	<ul> <li>Thermally-driven MD process using microporous hydrophobic membranes</li> <li>The organic retention times much greater than</li> </ul>	<ul> <li>Complete retentions of non-volatile organics</li> <li>Lower operating pressures than conventional pressure-driven membrane processes</li> <li>Particularly suitable for treating refractory organics which require a long residence time</li> <li>Complete rejection of total phosphorus</li> </ul>	<ul> <li>High energy requirement for heating, and uneconomical for large-scale applications</li> <li>Post-treatment required to recover methane from the permeate</li> </ul>

	<ul> <li>the hydraulic retention time</li> <li>Thermophilic bacteria at about 50°C</li> <li>Stable fluxes can be sustained</li> </ul>	<ul> <li>Shorter start-up time</li> <li>Less dissolved methane</li> </ul>	
AnOMBRs	<ul> <li>High retention forward osmosis semi-permeable membrane</li> <li>Draw solution (such as seawater) required</li> <li>Separation driven by osmotic pressure difference</li> <li>High rejection capacity</li> </ul>	<ul> <li>Better organic removal and higher biomethane potential</li> <li>Suitable for methane fermentation from dilute wastewater under mesophilic conditions</li> <li>Retention of alkalinity and avoiding reactor acidification</li> <li>Complete total phosphorus removal and partial ammonia and total nitrogen removal</li> <li>Promising methane production</li> </ul>	<ul> <li>Possible effects of alkalinity, salt and ammonia accumulation on the long term stable biogas production</li> <li>The inorganic-rich supernatant disposal</li> <li>Membrane's low endurance to biological attachment and high temperature solution for the long time operation</li> </ul>
AnSMBRs	<ul> <li>Rotary disk-supporting media for membrane fouling control</li> <li>Sponges for sustaining microbial growth and fouling control</li> </ul>	<ul> <li>Enhanced membrane permeability</li> <li>Low cost sponge media for mobile carrier</li> <li>Successful membrane fouling control (scouring) without any membrane cleaning</li> </ul>	<ul> <li>Limited data available to examine the biogas production from such AnSMBRs</li> <li>Further studies required to determine the optimal sponge characteristics for the optimized biogas production</li> </ul>
Gl- AnMBRs	<ul> <li>Two-phase flow in the membrane unit</li> <li>Enhanced sludge retentate recirculation</li> </ul>	<ul> <li>Lower cross-flow velocity</li> <li>Biogas-assisted mixing can help with reducing methane super-saturation</li> </ul>	<ul> <li>Helium gas required for the start-up</li> <li>Continuous scouring can cause varying gas equilibriums, methane oversaturation and changing pH</li> <li>High shear stress on the methanogens due to the gas recirculation</li> </ul>

V- AnMBRs	Membrane vibration system for fouling control instead of traditional gas-sparging means	<ul> <li>No mixing system required</li> <li>Suitable for fouling control when biogas gassparging is not feasible</li> <li>No biogas sparging shear stress on the biomass</li> </ul>	<ul> <li>Inhibited methanogenic activity due to high salinity and toxic sulfide levels</li> <li>Vibratory shear stress may affect the microbial activity for biogas production and requires further investigation</li> </ul>
AnBEMRs	Biomass entrapped in the bio-carriers/bio-balls	<ul> <li>Better organic removal and higher methane yield than conventional AnMBRs</li> <li>High organic loading</li> <li>Significantly reduced fouling</li> <li>Suitable for claiming biomethane from complex wastewater (pharmaceuticals)</li> </ul>	<ul> <li>Acidogenesis and methanogenesis inhibition at high salinity conditions and organics overloading</li> <li>Dissolved methane causes the lower methane yield</li> <li>Long start-up period</li> </ul>

## 3. Inhibitors of biogas production

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#### 3.1. Ammonia

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During methane fermentation, ammonia is generated by the biodegradation of the nitrogenous compounds mostly in the form of proteins [60]. As ammonia concentration grows above 3500mg/L, methane yield starts to suffer from decreasing below the theoretical value [61]. As ammonia concentrations climb up to the values of 4051–5734 mg NH<sub>3</sub>-N L<sup>-1</sup>, the methanogenic bacteria can lose 56.5% of its activity. Ammonia inhibition includes the increase of maintenance energy requirement, a change in the intracellular pH, and inhibition of a specific enzyme reaction. Free ammonia (FA) (NH<sub>3</sub>) is more toxic than ionized ammonia (NH<sub>4</sub><sup>+</sup>) because it is able to penetrate through the cell membrane, resulting in the disruption of cellular homeostasis, potassium deficiency and/or proton imbalance [60]. A higher temperature and pH value can exacerbate the inhibition by releasing more FA [62]. Release of ammonia is a primary concern for a high rate or intensified process such as AnMBR treating high strength waste, where shorter HRT and higher OLR may cause substrate and inhibitory intermediates to accumulate in the reactor [63]. Measures to minimize ammonia inhibition are demanded especially for thermophilic methane fermentation from AnMBRs. Kanai et al. [61] proposed the Kubota Submerged Anaerobic Membrane Bioreactor, in which the membranes could retain methanogenic bacteria while ammonia could be filtered out with the permeate, allowing the efficient production of methane from Japanese garbage with high protein levels (TN concentration at 10,000 mg/L). The recovered energy from this process was well above the overall energy consumption, enabling such a SAnMBR to be a net energy producer. Meabe et al. [62]

suggested that the acclimatization of biomass due to the high SRT in the AnMBR system

could affect the degree of ammonia inhibition, and no critical inhibition by ammonia was expected in the their mesophilic and thermophilic study.

Jensen et al. [63] also reported a successful methanation process from anaerobic digestion of slaughterhouse wastewater containing high proteins using a pilot-scale AnMBR. They found over 95% of COD in the wastewater feed was converted into biogas with 70% methane, and 78–90% of nitrogen was released to the permeate as ammonia, which meant ammonium inhibition was minor in this system. Landfill leachate also features high concentrations of organic contaminants and ammonia, which can be problematic when used for biogas production. Xie et al. [47] have successfully applied an AnDMBR for biomethane production with average methane yield at 0.34 L/g COD<sub>removed</sub> (0.30L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) from high strength landfill leachate digestion. Although free ammonium nitrogen (FAN) concentration was completely inhibitory in this case, detrimental effects were not observed on the performance due to the microorganisms' adaption to high free ammonia concentrations. The authors also suggested an interesting finding that the acetate-consuming methanogens were less inhibited than hydrogen utilizing methanogens, despite high ammonium concentrations in the reactor (over 3000 mg/L NH<sub>3</sub>-N at mesophilic conditions). This has proved to be controversial to other researchers [60, 64] indicating that acetic lastic methanogens were more sensitive to ammonium inhibition compared with hydrogenotrophic methanogens.

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# 3.2. Sulfide

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Problems associated with the methane fermentation of sulphate-rich wastes are the toxicity of sulphide to anaerobic microorganisms and the competition for the influent COD from the sulphate reducing bacteria (SRB) (approx. 2g COD/g SO<sub>4</sub>-S<sub>removed</sub>), which suppresses methane productivity [60,65,66]. In particular, methane production from municipal wastewater can be challenging because it can be easily characterized by low COD/SO<sub>4</sub>-S

ratios. The fierce competition between methane producing bacteria (MPB) and SRB can negatively impact on the quantity and quality of the biogas produced. Although it is evident that the AnMBRs are more resistant to toxics due to the sufficient SRTs for methanogens, many studies have reported increased operational costs during the treatment of high sulphate containing wastewaters by the AnMBR, especially at psychrophilic conditions and lower SRTs. Both Ferrer et al. [65] and Pretel et al. [66] concluded that AnMBR systems represented more energy surplus potential, thus being a net energy producer when treating low-sulphate municipal wastewater in warm/hot climates. The cost savings of up to 28% (Ferrer et al., [65]) in treating low-sulphate can be achieved as compared to the scenario with sulphate-rich municipal wastewater. Liao et al. [12] reported the complete inhibition of biological activity caused by feed toxic shock (high concentration of H<sub>2</sub>S in feed) in a thermophilic SAnMBR with mesophilic sludge as the inoculum. Thus, the pretreatment of the feed should be in place to remove toxic sulfur substances so that the biological activity of thermophiles can be maintained. Gimenez et al. [67] also observed a low methane yield at 0.069 L/g COD<sub>removed</sub> (0.061 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) from a pilot-scale mesophilic SAnMBR treating wastewater with a low COD/SO<sub>4</sub>-S ratio, and this was mainly attributed to the SRB competition for 90% of influent COD. The methane recovery efficiency from SAnMBRs was greatly influenced by sulphate content in urban wastewater, and higher biogas production would be expected if high COD/SO<sub>4</sub>-S or no sulphate were present in the substrate [55]. The effective countermeasures to sulfide toxicity include the dilution of the wastewater, and the implementation of sulfide removal techniques such as physico-chemical measures (stripping), chemical reaction (coagulation, oxidation, precipitation), and biological conversions (micro-aerobic sparging and partial oxidation to sulfur) [60]. Acclimatization of MPB to free H<sub>2</sub>S to increase the tolerance of aceticlastic and hydrogenotrophic MPB to sulfide can also be a possible solution. Nevertheless, above-mentioned sulfide toxicity control

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techniques require further research to obtain valuable data from AnMBRs, in order to validate their applicability and effectiveness in sulfide control.

On the other hand, methanogenic activities are not inhibited if the ratio of COD/sulphate in substrate is higher than 10, and low concentrations of sulphate and sulphide are also necessary for effective biogas production [68,69]. Li et al. [70] compared the performance of two AnMBRs with and without the addition of sulphate for the anaerobic co-digestion of coffee grounds, milk and waste activated sludge. They concluded that sulphate addition (at a COD/SO<sub>4</sub><sup>2-</sup> ratio of 200:1 to 350:1) wielded positive effects on propionate degradation and methane fermentation in a thermophilic AnMBR at higher OLRs. Without the addition of sulphate, the thermophilic AnMBR system at higher OLRs entered a "sub-health state" as a consequence of propionate acid accumulation.

#### 3.3. Salinity

The presence of high salt concentrations is common in many saline wastes from fish and seafood processing, chemical, petroleum and pharmaceutical industries. High salinity can cause bacterial cells to dehydrate due to the osmotic pressure. With its toxic effects on non-adapted biomass mainly attributed to cations, high salinity is regarded as one of the most important factors influencing methane fermentation processes [60]. Enzyme inhibition, cell activity decline and plasmolysis are the typical manifestations of salt stress on anaerobic microbes [10]. Ng et al. [59] investigated strong salinity conditions' inhibitory effect on methane yield from both conventional AnMBR and advanced AnBEMR when treating the pharmaceutical wastewater. They found microbial flora was negatively impacted (methane yield below 0.16 L/g COD<sub>removed</sub> (0.14 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) was reported) in a hypersaline scenario, which was due to the disrupted ordinary metabolic functions and degradation kinetics with high salt concentrations. Jeison et al. [71] attributed the presence of

very small sized and weak granules in both UASB and AnMBR systems to the high salinity of the wastewater, despite the fact that membrane enhanced retention of active halotolerant bacteria contributed to a superior sludge activity than UASB. They also revealed that the long-term continuous adaption periods resulted in better levels of sodium tolerance, with the observed 50% activity inhibitory concentration (IC<sub>50</sub>) value for acetotrophic methanogenesis at approximately 25 g Na<sup>+</sup>/L. In addition, high salinity as a consequence of salt accumulation in the reactors is regarded as a significant concern for the AnOMBR in terms of fouling and excess flux loss rather than inhibition or toxic effects on the biological processes [52,53].

# 3.4. Long chain fatty acids (LCFAs)

Long chain fatty acids (LCFAs), are potentially suitable substrates for biogas production. However, the toxicity of LCFAs is known to impair granule formation, sludge flotation and washout, suppress methanogenic activity, mass transfer limitations of substrate, nutrients and biogas in anaerobic granular sludge bed reactors when treating high strength-lipid wastewaters [10, 60, 72, 73]. Dereli et al. [72] assumed that the major drawbacks mentioned above could be addressed by membrane assisted biomass retention in AnMBRs. However, they found the AnMBR process still suffered from reversible LCFA inhibition at 50 days SRT and in turn process instability, which was mainly caused by LCFA adsorption, although the membrane guaranteed excellent biomass retention when treating lipid rich wastewaters. The authors suggested AnMBR operation at shorter SRTs was preferred due to the deliberate washout of adsorbed and free LCFA, thus reducing high concentration LCFA inhibition or transport limitation. Nevertheless, a major fraction of LCFA would not remain degraded, therefore lowering the biomethane potential. Dereli et al. concluded that sustainable methane fermentation from all LCFAs required only very low applied Lipid/Mass ratios. Furthermore they [73] observed LCFA inhibition at high SRTs in their lab-scale AnMBR system when

639 treating wastewater with Fat, Oil, and Grease (FOG) concentration at  $11.3 \pm 0.5$ g/L. The 640 inhibitory effect accelerated biomass deflocculation and SMP release. 641 On the other hand the LCFA absorption on sludge flocs modified their hydrophobicity, 642 resulting in less fouling propensity. Jensen et al. [63] reported minor LCFA inhibition from an 643 AnMBR when treating slaughterhouse wastewater with average FOG concentration of 1407 644 mg/L. Ramos et al. [74] reported that the long-term sludge adaption to LCFAs was required 645 for high rate methanogenesis from LCFAs in an UASB coupled membrane system. 646 Acclimated sludge quickly reached maximum methane production from the digestion of 647 substrate with high oil and grease (O&G) content (4.6-36 g O&G/L) at OLR of 17 kg 648 COD/(m<sup>3</sup>d), without any notable inhibitory effects. The advantages and disadvantages of 649 AnMBRs to mitigate problems induced by inhibitors are summarized in **Table 3**.

Inhibitors	Inhibitory effects on biogas production	Advantages/Disadvantages of AnMBRs
Ammonia	<ul> <li>Disrupting intracellular pH</li> <li>Increasing maintenance energy requirement</li> <li>Inhibiting a specific enzyme reaction</li> <li>Methanogenic activity loss</li> </ul>	<ul> <li>Complete retention of biomass</li> <li>Ammonia were filtered out with the permeate</li> <li>Acclimatisation and adaption of biomass to free ammonia due to the applied high SRT in AnMBRs</li> </ul>
Sulfide	<ul> <li>The toxicity of hydrogen sulphide to anaerobes</li> <li>The reduction on quality and quantity of biogas production</li> <li>H<sub>2</sub>S can cause corrosion in boilers, engines, and pipes causing higher maintenance and replacement costs</li> <li>Downstream oxygen demand required for oxidising H<sub>2</sub>S</li> </ul>	<ul> <li>Complete retention of slow-growing methanogens</li> <li>Higher methane production potential at higher temperature and higher SRT</li> <li>Sulfate content in the substrate significantly affected the overall operating cost.</li> <li>Promising for treating low/non sulphate-loaded wastewater</li> <li>Cost associated with membrane fouling</li> </ul>
Salinity	<ul> <li>Reduced methanogenic activity</li> <li>Biomass decay</li> <li>Long adaptation time</li> <li>Negative impact on granule stability and granule size.</li> </ul>	<ul> <li>Enhanced retention of active halotolerant bacteria</li> <li>Flux decline due to salt accumulation</li> <li>Long term adaption leads to high tolerance</li> </ul>
LCFAs	<ul> <li>Impairment of granulation</li> <li>Sludge flotation, washout, and foam/scum accumulation</li> <li>LCFA precipitation on sludge particles</li> <li>Methanogenic inhibition due to mass transfer limitations</li> </ul>	<ul> <li>No biomass washout</li> <li>Lesser fouling due to increased sludge hydrophobicity</li> <li>Inhibition due to floc deterioration and SMP release.</li> </ul>

# 4. Influential factors on biogas production

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# 4.1.Temperature

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Temperature is a vital parameter that profoundly influences anaerobic processes. Attempts to produce biogas from AnMBRs have been made under all three different temperature ranges: psychrophilic (0-20°C) [18, 31], mesophilic (20-42°C) [19, 75] and thermophilic (42-75°C) [76]. Thermophilic AnMBRs are known to have a rate-advantage over the others due to a faster reaction time and higher volumetric loading rate, thus demonstrating higher biogas productivity [2]. Both Liao et al. [12] and Lin et al. [29] investigated thermophilic SAnMBRs for biogas production from kraft evaporator condensate treatment at 55±1°C, and proved that it was a feasible technology to produce a promising methane yield at average value of 0.35 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.29 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>) with an excellent fuel quality close to 85– 90% methane in the biogas. They attributed higher methane yield to the higher sludge digestion rate making a larger contribution under thermophilic temperatures. Qiao et al. [76] also reported that producing methane from coffee grounds via the thermophilic co-digestion SAnMBR was feasible, and traceable hydrogen content of 100-200 ppm was found in the biogas. However, thermophilic AnMBRs are more sensitive to the presence of toxic compounds such as hydrogen sulfide [12] and inhibitory substances in the feed, and environmental changes. The thermophilic process's extreme temperature is also believed to cause severe ammonia toxicity, digestion inhibition and unstable fermentation processes [3, 77]. Furthermore, higher membrane flux can be retained using lower energy requirements at higher temperatures due to the reduced viscosity of the biomass suspension [10, 78]. Membrane permeability can be further enhanced by decreasing the transmembrane pressure (TMP) due to a lower permeate viscosity at high temperature. However, Jeison and van Lier

[79] observed long-term fluxes in the thermophilic SAnMBR were in fact 2–3 times lower than those attained under mesophilic conditions. Therefore, more studies regarding temperature's effects on the sustainable flux are required so that biogas production from AnMBRs is at its most efficient. Membrane fouling is another critical area of interest, which impedes the development of thermophilic AnMBRs for biogas production. Lin et al. [29] compared thermophilic and mesophilic AnMBRs at different OLRs but under similar hydrodynamic conditions. A more compact, less porous cake layer with higher cake resistance was observed in the thermophilic AnMBR, which was mainly due to the higher concentration of fine particles and EPS release at higher temperatures. Furthermore, permeate from the mesophilic AnMBR had much better quality than the thermophilic one. More attention should therefore be paid to the sustainable fouling mitigation measures in order to ensure the economic feasibility of thermophilic AnMBRs for biogas production.

Psychrophilic AnMBRs has recently attracted significant attention particularly in terms of generating biogas from low strength wastewaters [18]. It was found that both psychrophilic and mesophilic AnMBRs achieved comparable methane production rates [75, 80], although the former corresponded to significant methane loss in the permeate [75, 81] and a slightly higher fouling rate due to VFA accumulation [18, 75] and protein-dominated EPS release [82]. Last but not least, Gao et al. [83] reported temperature shocks led to a temporary increase in biogas generation rate, but shocks with larger magnitude at higher temperatures resulted in performance being significantly disrupted.

# 4.2. pH

Most AnMBR systems operate at near neutral pH since methane fermentation takes place within the pH 6.5–8.5 range with the optimal range from 7.0 to 8.0 [3]. Such a pH range was usually maintained through neutralization, which requires the excessive use of chemicals such

as sodium carbonate/biocarbonate or calcium carbonate since some streams have extreme pH values, and hydrolysis and acidogenesis phases will decrease pH values. Extreme pH conditions during AnMBRs operation can not only upset biological performance and methane yield but also affect membrane permeability and lifespan [84]. Gao et al. [85] investigated the effects of elevated pH shocks (pH 8.0, 9.1 and 10.0) on biogas production from a SAnMBR, and found that the pH 8.0 shock had a minor impact, yet pH 9.1 and 10.0 shocks did exert significantly negative impacts on the methane yield. This was mainly due to the ammonia toxicity and VFA accumulation at increased pH value. Serious membrane fouling resistance was reported, due to pH shock induced sludge flocs breakage and the accumulation of fine particles in the bulk sludge. In light of the difference in growth rates, and optimum pH for the growth of acidogens (5.5-6.5) and methanogens (6.5-8.2), many researchers have worked on phased AnMBRs, which separate acidogenesis and methanogenesis processes into the twostage reactor configuration [86]. Optimizing each stage separately in its own reactor reduces VFA accumulation, facilitates process stability, and enhances the system's tolerance to greater loading rate and toxicity. These features will lead to higher methane production. Such a phased AnMBR has been successfully applied to high loading wastewaters treatment for maximum methane yield [39, 86, 87].

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# 4.3. Hydraulic retention time (HRT)

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HRT is a key parameter from an economic perspective as it has a significant impact on the capital cost, meaning shorter HRTs allow smaller biogas-producing AnMBRs [88]. Many researchers have worked on the influence of HRT on biogas production from AnMBRs. Generally, HRTs can range from as low as 2 h [33] to as high as 30 d [89] depending on feed characteristics, system hydraulics, sludge properties, etc. Ho and Sung [90] reported that methane recovery decreased by 13% from municipal wastewater as a result of the increased

COD accumulation in the AnMBR when reducing HRT from 12 to 6 hours. Therefore,
AnMBR operation with relatively long HRTs may maximize methane recovery. However,
Yuzir et al. [91] observed reduced methane productivity from AnMBRs with longer HRT due
to less COD available as substrate for methane production. Significantly enhanced methane
production was evident when high hydraulic shock load was applied (HRT 1 d) and they
attributed this high yield to enhanced levels of hydrogenotrophic methanogenesis rather than
acetoclastic activity. Huang et al. [92] also reported that a shorter HRT increased biogas
production due to increased organic loading rate in a SAnMBR. However, too short an HRT
was not recommended due to higher biomass concentrations and higher SMP that could
worsen membrane fouling. Gao et al. [36] observed something different when they
investigated the effects of decreasing HRT on biogas production from an integrated anaerobic
fluidized-bed membrane bioreactor. They found that methane productivity increased when the
HRT decreased from 8h to 6h, which was linked to the increased OLR. Meanwhile the
productivity decreased as more VFAs accumulated with a much shorter HRT.
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cause AnMBRs reactor volume to be used inadequately and shortened HRT may lead to VFA accumulation, reduced methane productivity and severe membrane fouling.

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### 4.4. Solid retention time (SRT)

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Unlike other types of anaerobic reactors, AnMBRs enable SRT to become completely independent from HRT, irrespective of the sludge properties. SRT values ranged from 20 d [72,94] to infinite days [92], although most researchers worked using SRT values higher than 160 d. As a thumb of rule, AnMBRs operating at longer SRTs produce greater quantities of biogas because any decrease in the SRT may reduce the extent of reactions needed for stable digestion. For example, Huang et al. [92] in 2011 reported that a longer SRT would enhance the dominance of methanogenesis and lead to more biogas generation. In their study, methane yield rates of 0.670±0.203 L CH<sub>4</sub>/d, 0.906±0.357 L CH<sub>4</sub>/d, 1.290±0.267 L CH<sub>4</sub>/d (0.610±0.203 L CH<sub>4</sub> (STP)/d, 0.825±0.357 L CH<sub>4</sub> (STP)/d, 1.175±0.267 L CH<sub>4</sub> (STP)/d) were reported at SRTs of 30, 60 and infinite days, respectively. Yeo and lee [95] suggested that AnMBR operation under a long SRT could permit low dissolved methane concentration in AnMBR permeate, along with high methane recovery. They attributed 45% more methane production at higher SRT to supplemental methane formation originating from biomass electrons via endogenous decay. It is obvious long SRTs are more favorable in AnMBRs' operation since it results in minimal sludge production and hence significantly reduces disposal cost. However, longer SRTs operation can also impact on methanogenic activity due to a decrease in viable biomass concentration [10]. The effects of long SRTs on membrane fouling, furthermore, require urgent attention. Prolonged SRTs can hinder sludge flocculation and reduce particle size, and increase the release of soluble microbial products (SMP) [92]. On the other hand, high sludge concentration at high SRT can result in a rapid cake formation and compaction, leading to

excess flux decline [10]. Additionally, the accumulation of inorganic solids at high SRTs may also increase inorganic fouling, which in many studies was found to be serious [73, 96].

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# 4.5. Organic loading rate (OLR)

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AnMBR processes have the competitive advantage of accommodating fluctuations in the organic loading, and OLRs ranging from 0.23 [37] to 33.7 kg COD/m<sup>3</sup>·d [76] have been applied in AnMBRs for biogas production. In general, OLR represents the quantity of volatile solids fed into a biogas digester per day under continuous operation [2]. When an increase in OLR occurs, therefore, the biogas yield is supposed to also increase to a certain extent. An et al. [93] reported that biogas yield from an AnMBR rose linearly with an increase in the organic loading. Wijekoon et al. [97] also observed a continuous increase in biogas production rate from 5L/d to 35L/d with increasing loading rate from 5 to 12 kg COD/m<sup>3</sup>·d in a two-stage thermophilic AnMBR. Bornare et al. [16] reported an increase in the average biogas generation from 159 to 289 L/d but a decrease in the biogas yield from 0.48 to 0.42 L biogas/g COD<sub>removed</sub> when increasing OLR from 0.62 to 1.32 kg COD/m<sup>3</sup>·d. They attributed this conflicting outcome to a better food-to-microorganisms (F/M) ratio (0.08kg COD/kg MLVSS/day) at a lower OLR. Dereli et al. [10], however, stated that the effect of OLR should be assessed together with SRT and biomass activity as the system's OLR was not an independent parameter. Applied temperature also exerts a profound influence on the applicable OLRs in AnMBRs. Thermophilic AnMBRs emerge as being more effective in coping with higher volumetric loading than AnMBRs operating in the mesophilic range [98]. It should be also noted that as the organic loading rate increases, the risk of deteriorating biogas production due to VFA accumulation may occur due to the inhibition of microbial activity [97]. For example, Saddoud and Sayadi [39] documented a process failure in their study, i.e. a drastic decrease in

the methane yield at OLR of 16.3 kg COD/m³·d due to VFA accumulation and methanogenic
inhibition in a one-phase AnMBR. They subsequently suggested a two-stage AnMBR with
the anaerobic filter as acidogenic reactor and jet flow AnMBR as methanogenic reactor at
high OLR and achieved a significant improvement in biogas conversion in the staged
AnMBR.
Serious fouling caused by the release of EPS/SMP and the accumulation of large fine
particles, became another issue associated with high loading AnMBRs when using AnMBRs
for biogas production [99]. In the face of the comprehensive effects of OLR on methane
production, Wei et al. [19] proposed the concept of sustainable OLR to optimize energy
recovery potential from typical municipal wastewater through mesophilic AnMBRs. They
reported sustainable OLR of 6kg COD/m <sup>3</sup> ·d could result in maximum methane yield up to a

theoretical value of 0.382 L CH<sub>4</sub>/g COD<sub>removed</sub> (0.318 L CH<sub>4</sub> (STP)/g COD<sub>removed</sub>). Table 4

summarizes the operational factors affecting biogas production from AnMBRs and the

recommendations for optimized biogas production.

Factors	The effects on biogas production process	Possible suggestions for optimized biogas production from AnMBRs
Temperature	<ul> <li>Faster reaction rates→higher-load bearing capacity→higher biogas productivity</li> <li>Possible acidification→ inhibition of biogas production</li> <li>Decreased stability and increased toxicity→poor methanogensis→higher net energy input and larger investments</li> <li>Difficulty in anaerobic biomass immobilization→poor sludge settling characteristics→ reduced methanogenic activities→poor effluent quality</li> <li>Less cooling required→improved process economics</li> <li>Sludge decay with non-adapted mesophilic sludge→serious membrane fouling</li> <li>Reduced sludge viscosity→A higher flux→process efficiency  →Lower shear rates→ lower energy requirement</li> <li>A lower permeate viscosity→increased membrane permeability by decreasing TMP</li> <li>More compact cake layer→higher cake layer resistance→server fouling issues→very low long-term flux→process inefficiency</li> </ul>	<ul> <li>Two phase AnMBRs with thermophilic hydrolysis/acidogenesis and mesophilic methanogenesis</li> <li>Avoidance of drastic temperature changes</li> </ul>
	Mesophilic:	
	Better process stability, higher biomass richness, better permeate quality but possible low methane yields and poor biodegradability and nutrient imbalance	
	Psychrophilic:	
	<ul> <li>Enhanced methane solubility→loss of methane in effluent→lower methane recovery</li> <li>TSS and soluble COD accumulation and a higher viscosity→increased filtration resistance→increased fouling and operational cost</li> <li>Enhanced membrane removal and compensation for the decreased SMA and bulk sludge removal</li> <li>Energy requirement for operating the system is lower</li> <li>Reduced reaction and hydrolysis rates→reduced methanogenic activity</li> </ul>	

Temperature	changes:
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- Temperature decrease → decreases in the VFA production rate, the ammonia concentration, the substrate utilization rate and the metabolic rate of the microorganisms → increased start-up times → decreasing CH<sub>4</sub> and H<sub>2</sub> yields
- Temperature increase in pH, hydrolysis of organic particulates→increase in methane potential
- Temperature increase→ Free ammonia concentration→ methanogenic inhibition
- Temperature fluctuation→stress on biomass →increase membrane fouling and operational cost

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- Extremely low pH value→acidification and VFA accumulation→reduced methane yield
- Extremely high pH value→increased ammonia toxicity and VFA inhibition→reduced methane yield
- pH shocks→dispersion of sludge flocs→the accumulation of colloids, solutes or biopolymers in the bulk sludge suspension→deteriorated membrane performance and biogas production potential

**HRT** 

- Optimum HRT exists, which ensures the maximum methane productivity
- HRT lower than the optimal value→VFA accumulation→ reduced methane yield→ server fouling
- HRT above the optimal value→insufficient utilization of biogas digester component→reduced methane production

SRT

- Long SRT—enhance dominancy of methanogenesis—enhanced methane yield
- Long SRT—reduced dissolved methane—higher methane recovery
- Long SRT→ reduced sludge disposal and cost
- Long SRT→reduced sludge particle size and release of SMP→membrane fouling
- $\bullet \quad \text{Long SRT} {\rightarrow} \text{cake formation and consolidation} {\rightarrow} \text{increased fouling cost}$
- Long SRT 

  -- accumulation of inorganic solids 

  -- inorganic fouling

- Two phase AnMBR with optimized conditions for both acidogenic and methanogenic reactor to bring biogas yield optimization
- Minimize pH shock loading by neutralizing the feed with chemicals such as sodium biocarbonate
- Avoid operation at too high or too low a HRT
- Operate AnMBRs for maximum biogas production at optimal HRT
- Long SRT is generally recommended for AnMBRs operation
- Additional care is required for fouling mitigation at long SRT

# **OLR**

- Increased OLR→higher metabolic activity of methanogens→increase biogas yield and methane content in the biogas to certain extent
- High OLR→VFA accumulation→irreversible acidification→risk of a deteriorated biogas yield
- High OLR or organic shock loading—release of tight EPS/SMP and accumulation of fine particles—serious membrane fouling
- Operating AnMBR at sustainable OLR to maximize the methane yield.
- Thermophilic systems and effluent recirculation can help relieve systems from the overloading issues

#### 5. Future perspectives

The critical analysis of recent literature reveals that much progress has been made in the research and development of AnMBRs for biogas production. The green perspectives of AnMBRs including biogas production, high effluent quality, waste minimization, high capacity, footprint efficiency (reducing capital costs), lower energy requirements, and decentralized operation, mean that such a technology can produce bioenergy through a sustainable bioprocess. For example, a substantial commercial value of US\$341640/year could be obtained from a full-scale AnMBR's recovery of biogas [17]. Most research on AnMBRs has focused on investigating traditional configurations such as CSTR, UASB, AFBR, etc. Recent advances in the modification of conventional AnMBRs and the development of novel AnMBR processes have witnessed greater outcomes. Incorporating a dynamic membrane into AnMBRs significantly reduced capital exploitation costs for the membrane and its cleaning and replacement. Thermally-driven membrane distillation minimized the methane loss through the liquid phase and completely rejected total phosphorus when coupled with AnMBRs. The forward osmosis process in the AnOMBR provides nearly complete removal of total phosphorous and records a relatively good rejection rate for total nitrogen and ammonia [52, 53]. The developments of V-AnMBRs, Gl-AnMBRs, AnBEMRs and AnSMBRs offer new insights into sustainable fouling mitigation strategies that enhance the economic feasibility of AnMBRs for biogas production. The literature review revealed that most of the research reported on biogas production from AnMBRs was confined to lab scale experiment. The results from lab testing most often could not directly transfer to industrial and commercial practical application. Fundamental information regarding the energy consumption of each specific AnMBR and its capital cost for installation and operation is still lacking. Further research at well controlled pilot or full scale AnMBRs studies is greatly appreciated to obtain valuable data, in order to support the wide

implementation of such green technology at industrial scale.

Depending on the origin, the feedstock for AnMBRs may contain inhibitory compounds such as ammonia, sulfide, salt or LCFAs. Accumulation of these inhibitors in AnMBR may cause process upset or even reactor failure, indicated by the reduction in daily biogas production and methane composition. Therefore, co-digestion with other wastes, acclimatization of microorganisms to toxic compounds, and pretreatment to remove or counteract toxicants can enhance the process stability and optimize biogas production [60]. Despite extensive research on the operational parameters' effects on the biogas productivity from AnMBRs, a universal operational protocol has not yet been developed to optimize biogas production due to the complexity for the consortia of microorganisms, variations in the feed characteristics, and differences in system hydraulics. Although thermophilic AnMBRs have a rate-advantage over mesophilic ones, greater investment is required to deploy thermophilic systems and mitigate the severe membrane fouling.

Prolonged SRT is believed to minimize the toxic impacts and enhance methanogensis but the resultant fouling issues and subsequent flux decline are problems that remain unsolved. Decreasing the HRT to acquire the optimal OLR is a possibility. However, published information on the maximum sustainable loading rate is still lacking and its relationship to the applied HRT requires further investigation. Numerous researchers have reported the issues associated with dissolved methane at the effluent, despite variations in AnMBR configurations. Due to economic and environmental concerns, methane leakages must be minimized [75]. Developing effective and applicable dissolved methane recovery process is of great interest to optimize bioenergy recovery and minimize direct greenhouse gas emissions to the atmosphere.

### 6. Conclusions

AnMBR technology, as future green bioprocess has much potential for renewable energy production. Currently AnMBRs are starting to evolve to the next level. Advanced AnMBRs have attracted a lot of interest in producing high fuel quality biogas when they are combined with many novel technologies such as Anammox, dynamic membrane, membrane distillation, etc. The biogas production from AnMBRs is greatly influenced by many factors including temperature, pH, HRT, SRT, and other variables. The opportunity to apply such green technology on a commercial scale first needs prospective research studies to overcome many challenges such as methane recovery, product inhibition, membrane fouling, and membrane cost.

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