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Chapter 8: Anaerobic membrane bioreactors for emerging pollutants removal

Book: Advanced Membrane Bioreactors for Sustainable Wastewater Management- Current status and recent developments in anaerobic membrane bioreactors

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Abstract

This chapter evaluates the potential of anaerobic membrane bioreactor (AnMBR) to as a technology platform for recovering of clean water, energy, and nutrient from wastewater. AnMBR has a promising potential for removing an wide array of trace organic contaminants (TrOCs) of concern to water reuse, convert organic carbons in wastewater to biogas, and solubilise nutrients (e.g. ammonia and phosphorus) for subsequent recovery for fertilizer production. Research to date reveals several technical challenges to the practical applications of AnMBR for wastewater treatment for resource recovery and water reuse. These challenges include the dilute nature of municipal wastewater, which entails the need for pre-concentrating wastewater prior to AnMBR, and hence, issues related to salinity build-up, accumulation of substances, membrane fouling, and current limited understanding of the removal of TrOCs. Strategies to overcome these technical challenges are proposed to provide a research road map to guide future AnMBR development.

Keywords: Anaerobic membrane bioreactor; Trace organic contaminants; Wastewater treatment; Water use; Resource recovery.

1. Introduction

Wastewater is a vein of resources for the production of clean water, renewable energy, and fertilisers [1]. Water reuse has been practiced globally as an important measure to tackle water scarcity and environmental pollution [2]. As a notable example, through persistent scientific development over the last few decades, Israel has succeeded in reusing most of its wastewater effluent for irrigation, thereby effectively transforming desert into arable and productive farmland [3]. Safe and reliable water reuse requires adequate removal of organic substances and nutrients (nitrogen and phosphorus), which on the other hand, are important resources for our sustainable development. Organic substances considerably embedded in wastewater can be converted to methane rich biogas via anaerobic digestion. The produced biogas can be used directly or converted to electricity and thermal energy for beneficial use. Nitrogen and phosphorus are important nutrients but, in the environment, can also lead to eutrophication of water bodies and result in struvite blockage to wastewater treatment facilities [4]. Thus, nutrient recovery from wastewater for fertiliser production not only secures food and agriculture production, but also reduces the cost of wastewater treatment and facility maintenance.

Membrane bioreactors (MBR) have been increasingly deployed globally for wastewater treatment and reuse. MBR is typically the integration of membrane separation processes, such as microfiltration and ultrafiltration, with conventional activated sludge (CAS) treatment. Compared to CAS treatment alone, MBR is more robust with a much smaller physical footprint and improved effluent quality [5]. Indeed, previous studies have well documented that MBR can offer enhanced removal efficiency of emerging organic contaminants, particularly those biodegradable and hydrophobic compounds in comparison with solely CAS treatment [6-8]. These emerging organic contaminants, often referred to as trace organic contaminants (TrOCs),

such as pharmaceuticals, personal care products, and estrogens, are chemicals of emerging concern that are present in wastewater at trace levels (a few nanogram per litre to several microgram per litre) are arguably the most vexing challenge to water reuse given their threat to the environmental and human health [9, 10]. It is noteworthy that the term MBR is often generically referred to aerobic MBR with a focus only on water reclamation rather than the recovery of energy and nutrients from wastewater. Furthermore, aerobic MBR is energy intensive with aeration being a large electricity consumer to support the CAS treatment [11].

Recent efforts to transform the existing wastewater treatment plants to be energy neutral or positive has resulted in the development of anaerobic MBR [12, 13]. AnMBR combines membrane separation process with anaerobic digestion. Thus, organic substances in wastewater are biologically converted into methane rich biogas, which can then be converted to electricity to offset the energy consumption of wastewater treatment facilities [14]. Moreover, nutrients (e.g. nitrogen and phosphorus) can be transformed to more chemically available forms by anaerobic digestion to facilitate subsequent recovery or direct agricultural utilisation [15, 16]. With the integration of membrane separation processes, AnMBR can enhance the performance of anaerobic digestion regarding improved biogas production and effluent quality [17]. In particular, the membrane process is effective at preventing biomass wash-out, which is critical for anaerobic reactor configuration with low growth rate. AnMBR is capable of treating high strength wastewater with more than 90% removal of organic matter (as indicated by chemical oxygen demand (COD) measurement) and converting up to 98% of influent COD to biogas under optimised conditions [18].

Despite the promise of AnMBR to achieve energy positive wastewater treatment, there are several fundamental challenges to its further development. These challenges evolve around the

dilute nature of municipal wastewater, inhibitory substances (e.g. inorganic salts and sulfide), the ubiquitous occurrence of TrOCs in municipal wastewater, and membrane fouling. Municipal wastewater contains trace of numerous emerging pollutants but has low concentrations of organic matter, and thus, cannot provide sufficient organic loading rate for anaerobic digestion in AnMBR treatment [19]. Thus, membrane processes, such as reverse osmosis (RO) and forward osmosis (FO), have been used to concentrate municipal wastewater to the level suitable for anaerobic treatment [20]. It is noteworthy that membrane processes not only enrich organic substances, but also inhibitory substances in the concentrated wastewater. In most cases, the accumulation of inorganic salts in the concentrated wastewater is severe when FO is used due to its reverse salt diffusion from the draw solution [21]. In addition, these inhibitory substances are also vexing challenges to AnMBR treating industrial waste streams, for example, from food and paper production processing [22]. Nevertheless, to date, little is known about the comprehensive performance of AnMBR for treating concentrated wastewater with the build-up of these inhibitory substances.

AnMBR can be integrated with additional processes, such as membrane filtration, biological treatment, and advanced oxidation, to enhance contaminant removal and nutrient recovery from the effluent [19]. As a notable example, a novel membrane separation process, membrane distillation (MD), has been recently used to further purify the AnMBR effluent [23, 24]. MD is a thermal-driven process, where water evaporates from the feed solution, across a microporous membrane, into a distillate with vapour pressure difference between these two solutions as the driving force [25]. Since AnMBR is usually operated under mesophilic and thermophilic conditions, its hybridisation with MD (i.e. AnMBR-MD) can potentially reduce the considerable heat requirement for MD operation. On the other hand, MD is highly selective

and allows for further removal of contaminants from AnMBR effluent to advance wastewater treatment and reuse [26]. Such integration also provides new insights to the development of a proof-of-concept anaerobic membrane distillation bioreactor (AnMDBR), which directly combines MD with anaerobic treatment. Nevertheless, investigations on the compatibility of MD with either AnMBR or anaerobic digestion to advance wastewater treatment and reuse are rather scarce.

2. MBR for wastewater treatment and water reclamation

2.1 Fundamentals of AnMBR

Aerobic and anaerobic processes for wastewater treatment can be integrated with membrane filtration to form an aerobic MBR or an AnMBR system, respectively. MBR applications for wastewater treatment, especially water reuse, have increased significantly the last few years. The largest MBR plant of more than 1 GL/d in peak dry weather flow has just been commissioned in Hubei China in early 2019. Given AnMBR's ability to treat concentrated wastewater and simultaneously produce biogas, which is a biofuel, the number of practical applications and research works focusing AnMBRs, have increased significantly [27-29].

The anaerobic treatment process occurs in the absence of oxygen via four interrelated stages including hydrolysis, acidogenesis, acetogenesis, and methanogenesis. The conversion of organic carbon in wastewater to biogas depends on the symbiotic relationship among the different microbes. A few notable groups of these microbes include fermentative bacteria, acetogens, and methanogens [22]. Among these microbes, methanogens appear to be the most significant for biogas production since they convert intermediate products (such as acetic acid) from previous stages to methane gas. Methanogens can be easily washed out from conventional

anaerobic reactors. They are also slow-growing microbes. Thus, the integration of membrane separation such as ultrafiltration (UF) and microfiltration (MF) to the anaerobic process helps to retain methanogens in the reactors. In addition, the hydraulic retention time (HRT) can also be decoupled from the sludge retention time (SRT), allowing for a high treatment intensity (short HRT) but with the necessary SRT. Overall, by integrating anaerobic treatment with the membrane separation process, AnMBR can, in many cases, generate more biogas [12].

AnMBR may differ significantly from aerobic MBR with respect to contaminant removal efficiency, treatment intensity, and energy consumption (Table 8-1). Because AnMBR is operated without aeration, it has a lower energy demand compared to aerobic MBR. The produced biogas can be converted to electricity to offset the energy footprint of AnMBR [15]. It is, however, noted that some energy is still required to control membrane fouling. The energy demand in submerged AnMBR can vary over a wide range from 0.03 to 5.7 kWh/m³ due to different energy requirements for gas sparging to control membrane fouling [14]. AnMBR is usually operated at a high biomass content and long SRT, it is most suitable for treating high strength wastewater [30]. In the anaerobic process, carbon dioxide as the terminal electron acceptor and organic carbon is used to produce methane (CH₄) gas rather than for biomass production. As a result, AnMBR generates less sludge than aerobic MBR [12]. On the other hand, in comparison to aerobic MBR, anaerobic respiration is slow and AnMBR usually has a lower treatment performance in terms of contaminant removal efficacy and treatment intensity (or capacity).

Insert Table 8-1

2.2 AnMBR configuration

Several anaerobic treatment processes can be coupled with membrane separation to form different AnMBR configurations (Figure 8-1). Several systematic reviews of anaerobic bioreactors specifically for AnMBR applications can be found in the open literature [19, 22, 30]. Common anaerobic treatment processes for AnMBR include completely stirred tank reactor (CSTR), up-flow anaerobic sludge blanket (UASB), and anaerobic fluidized bed bioreactor (AFBR). CSTR is most commonly used for AnMBR due to its simple construction and operation. In the UASB configuration, biomass can be retained mostly in the bottom zone of the bioreactor, thus, the effluent passed through the membrane unit has a low solid content. Thus, the UASB configuration can help to alleviate membrane fouling. The produced biogas can be captured through a gas and liquid separator. In the AFBR configuration, granular media (e.g. activated carbon or sponge) suspended in the reactor is used to retain biomass in the reactor [37]. Thus, similar to the UASB configuration, membrane fouling can also be alleviated.

Similar to a generic MBR system, the membrane module can be arranged in either side-stream or submerged directly in the anaerobic reactor (Figure 8-1). In the side-stream configuration, the membrane is placed outside of the bioreactor. Digestate in the anaerobic reactor is transferred to the membrane unit and permeate is pushed or extracted through the membrane. In the submerged configuration, the membrane unit is directly immersed in the anaerobic reactor to extract treated water through the membrane. Alternatively, the submerged AnMBR can also be deployed by placing the membrane module in a separate chamber from the working anaerobic reactor. Excess digestate from the membrane tank can be circulated back to the anaerobic reactor for further treatment. This is known as a two-stage configuration.

Insert Figure 8-1

In recent years, researchers have attempted to address the low contaminant removal efficiency of AnMBR by introducing new variations including anaerobic membrane distillation bioreactors (AnMDBR) and anaerobic osmotic membrane bioreactor (AnOMBR). In these systems, membrane distillation (MD) or forward osmosis (FO) process are integrated with anaerobic treatment to enhance the removal of contaminants for water reuse applications.

AnMDBR is defined as the combination of MD and anaerobic treatment. MD is a hybrid thermally driven membrane separation processes. Due to the different in temperature across the membrane, there is a different in partial vapour pressure of water, resulting in water vapour transport through a hydrophobic, microporous membrane. The advantages of anaerobic treatment processes can be utilized when combining with MD, because the thermophilic operation for anaerobic treatment can complement with MD operation [24].

AnOMBR, which integrates forward osmosis (FO) and anaerobic treatment, is another promising technology for advanced wastewater treatment and reuse [39]. In the FO process, water from the feed solution is transported across the semi-permeable membrane to a draw solution under the osmotic pressure difference between these two solutions acting as the driving force. During AnOMBR operation, a draw solution recovery process, such as reverse osmosis (RO), can be used to recover the draw solution and produce clean water [40].

2.3 TrOCs removal

AnMBR is suited promising technology for treating high strength wastewater (i.e. wastewater with a high organic carbon content). Several pilot demonstrations and full-scale AnMBR

systems have been reported for treating wastewater from a variety of sources such as food processing, dairy production, and the beverage industry (Table 8-2).

Insert Table 8-2

TrOCs present arguably the greatest challenge to water reuse by AnMBR [9]. TrOCs have been ubiquitously detected in raw sewage and sewage-impacted water bodies around the world. These TrOCs include steroid hormones, pharmaceuticals and personal care products, industrial chemicals, pesticides, and disinfection by-products [48-53]. Many of these TrOCs are biologically active and may induce adverse impact on human health and the ecosystem [9].

TrOCs removal by MBR has been extensively investigated over the last two decades. Studies in the current literature have focussed almost exclusively on the aerobic MBR process. They have resulted in a systematic understanding of the fate of TrOCs during aerobic MBR treatment. By contrast, little is known about the fate and removal of TrOCs by AnMBR.

TrOCs removal from the aqueous phase during aerobic MBR treatment is governed mostly by biodegradation and adsorption. Molecular structure of the TrOCs is an important factor governing both of these mechanisms. Of a particular note, Tadkaew et al., [54] have successfully developed a framework for qualitatively predicting the removal of TrOCs by aerobic MBR treatment based on the presence of molecular electron donating groups (EDGs) or electron withdrawing groups (EWGs) and their hydrophobicity. In their study, Tadkaew et al., [54] shown that TrOCs with EDGs (e.g. hydroxyl and amine) can be effectively removed by aerobic MBR whereas TrOCs with EWGs (e.g. chloro and amide) in their structure are not effectively removed. In a later study, Wijekoon et al., [55] have adapted this framework and successfully elucidated the fate of TrOCs according to their molecular properties during aerobic MBR treatment. Wijekoon et al., [55], showed that recalcitrant and hydrophobic/hydrophilic

contaminants were mainly removed via adsorption to sludge. On the other hand, biodegradation/transformation was a major removal mechanism for readily biodegradable and hydrophobic/hydrophilic TrOCs. Previous work also suggests anoxic conditions could favour the removal of some TrOCs (e.g., carbamazepine and diclofenac [56]) that are otherwise recalcitrant to aerobic treatment. Similarly, there is also some evidence that nitrifying bacteria can enhance the removal of several TrOCs [55, 57].

Current understanding of the removal of TrOCs by AnMBR is still limited [58, 59]. Abargues, Robles [58] and Czajka and Londry [60] reported negligible anaerobic removal of 17 α -ethinylestradiol. By contrast, Monsalvo et al., [59] observed 20% removal of 17 α -ethinylestradiol. This discrepancy in the current literature can be attributed to the fact that several different microbial consortiums can be responsible for anaerobic biodegradation of TrOCs. Anaerobic biodegradation of TrOCs may be carried by sulphate, iron, and nitrate reducing bacteria that act as the final electron acceptors [60-62]. When nitrate is available, 17 α -ethinylestradiol can be effectively removed by biodegradation. In the absence of nitrate, removal of 17 α -ethinylestradiol via adsorption to biosolids was the main removal mechanism [62]. By contrast, Czajka and Londry [60], reported no biodegradation of 17 α -ethinylestradiol over 3 years of incubation period in anaerobic, sulphate, nitrate, or ion reducing condition. The anaerobic condition is particularly favourable for biological reduction of halogenated TrOCs (e.g., polyaromatic hydrocarbons) [61]. Anaerobic reductive dehalogenation appears to be a major mechanism for biodegradation of halogenated compounds since they can act as the terminal electron acceptor for a number of anaerobic microorganisms [61].

Research to date indicates that TrOCs removal by AnMBR is often lower than that by aerobic MBR. Monsalvo et al., [59] investigated the removal of 38 TrOCs by AnMBR. They reported over 90% removal of nine TrOCs; while the remaining 29 TrOCs were removed by less than 50%. In a subsequent study, Wijekoon et al., [63] compared the removal of TrOCs from the aqueous phase by AnMBR to values previously reported by Tadkaew et al., [54] (Table 8-3). Data from Wijekoon et al. [63] also showed that TrOC removal by AnMBR is also governed by their intrinsic physiochemical properties. The predictive framework previously proposed by Tadkaew et al., [54] was successfully used to relate the removal of TrOCs by AnMBR to three groups: namely A – hydrophilic TrOCs with only electron donating functional groups, B- hydrophilic TrOCs with only electron withdrawing functional groups, and C – hydrophobic TrOCs (Figure 8-2). Hydrophobic TrOCs were effectively removed by more than 70% since they readily adsorb to sludge and thus can be further degraded in the sludge phase (Figure 8-2). Hydrophilic compounds with electron donating functional groups (e.g. hydroxyl and amine) and nitrogen in their molecular structure also showed high removal efficiency. By contrast, hydrophilic compounds with electron withdrawing functional groups (e.g. chloro and amide) were persistent to AnMBR treatment (Figure 8-3).

Insert Table 8-3

Insert Figure 8-2#

Although Tadkaew's framework for qualitative prediction of TrOC removal can be adapted to AnMBR, there are a few notable differences in the removal of these TrOCs between these two treatment processes. Of a particular note, hydrophilic TrOCs containing sulphur or nitrogen in

their molecular structure showed higher removal efficiencies by AnMBR than by aerobic MBR, except for atenolol, paracetamol and diclofenac (Table 8-3). Examples of these sulphur or nitrogen bearing TrOCs include sulfamethoxazole, carbamazepine, linuron, omeprazole, and atrazine. The observed higher removal of carbamazepine reported by Wijekoon et al., [55] is consistent with a previous study by Hai et al., [56] who also reported considerably higher carbamazepine removal under anoxic compared to aerobic conditions.

Hydrophilic TrOCs that do not containing sulphur or nitrogen in their molecular structure (e.g., ketoprofen, ibuprofen and gemfibrozil) showed significantly lower removal efficiency by AnMBR when comparing to aerobic MBR (Table 8-2). The removal efficiencies of sulphur or nitrogen bearing and hydrophobic TrOCs by AnMBR are also higher than that by aerobic MBR previously reported by Tadkaew et al., [54]. However, the effect of sulphur or nitrogen on AnMBR removal were less significant when the hydrophobicity of the TrOCs was high. For instance, linuron ($\text{Log } D_{\text{pH } 7} = 3.12$) and clozapine ($\text{Log } D_{\text{pH } 7} = 3.23$) removal efficiencies by AnMBR were remarkably high and the removal of triclocarbon ($\text{Log } D_{\text{pH } 7} = 6.07$) by AnMBR was similar to that by aerobic MBR removal (Table 8-3). On the other hand, the removal of the hydrophobic TrOC ($\text{Log } D_{\text{pH } 7} \geq 3.2$) triclosan from the aqueous phase by AnMBR was lower than that by aerobic MBR previously reported by Tadkaew et al., [54], probably because triclosan does not contain sulphur or nitrogen in its molecular structure and in the anaerobic condition triclosan was mainly removed by methanogens [64]. Thus, sulphur and nitrogen reducing bacteria can be a major factor contributing to the observed difference in TrOCs removal efficiency between anaerobic MBR and AnMBR [60-62].

2.4 Fate and transport of TrOCs in AnMBR

Wijekoon et al. [63] was probably the first who attempt to quantify the accumulation of TrOCs in sludge for a comprehensive investigation of the fate of TrOCs during AnMBR treatment. Their results show that biodegradation was the most important TrOCs removal mechanism by AnMBR despite the observed accumulation of some TrOCs in the sludge phase (Figure 8-3). Wijekoon et al. [63] postulated that adsorption significantly increases the retention time of TrOCs in the biological reactor. In other words, there is significantly more time to biodegrade these TrOCs. In term of mass distribution, for almost all of hydrophobic TrOCs, biodegradation accounted for more than 80% while adsorption accounted for less than 20%. Biodegradation also accounted for most of the removal of hydrophilic TrOCs, although the distribution varied significantly from 6 to 99%. Among these hydrophilic TrOCs, only those that are recalcitrant contaminants (containing electron withdrawing functional groups) showed discernible accumulation in the sludge phase (i.e., diuron, diazepam, carbamazepine, diclofenac, triamterene, amitriptyline and trimethoprim). However, adsorption was low and only considerable for amitriptyline (18%). Taking the sludge phase into account, results from Wijekoon et al., [63]. (Figure 8-3) are broadly consistent with the framework for qualitative prediction of TrOCs removal by aerobic MBR proposed by Tadkaew et al., [54].

Insert Figure 8-3#

2.5 Biogas production

The organic carbon content in wastewater can be converted to biogas by AnMBR. In general, CH₄ yield is proportional to the COD loading rate [65]. In theory, 98% of the influent COD can

be converted into biogas, which can produce seven times the energy required for AnMBR operation [18]. In practice, actual biogas production is far below the theoretical value, due to a range of inhibiting factors and CH₄ loss due to dissolution in the effluent. CH₄ solubility in water is about 23 mg/L. Thus, CH₄ loss in the effluent can be significant, particularly for low strength municipal wastewater [15].

2.6 Nutrient removal and recovery

Nutrient removal by AnMBR depends largely on microbial assimilation and is inherently limited due to low biomass yield. On the other hand, anaerobic treatment liberates nitrogen and phosphorus in the form of ammonium (NH₄⁺) and phosphate (PO₄³⁻), respectively, thus facilitating their recovery through subsequent precipitation. Thus, incorporation of additional processes with AnMBR is necessary for nutrient removal/recovery from AnMBR effluent. Jacob et al., [23] reported 90% removal of COD and ammonium nitrogen from AnMBR effluent by a direct contact MD process. In particular, Song et al., [66] also demonstrated the complementarity between AnMBR and MD for TrOC removal.

3. Factors underlying key challenges to further AnMBR development

AnMBR has demonstrated some significant potential for resource recovery from wastewater. Nevertheless, there remain several technical challenges, particularly for treating municipal sewage, including the low resource (organic carbon and nutrient) content in wastewater, low temperature of municipal wastewater, and salinity build-up when diluted wastewater is pre-concentrated, membrane fouling and stability, and inhibitory substances (e.g. free ammonia and sulphide), and low and uncertain TrOC removal efficiency (Figure 8-4). Thus, further studies

are necessary for developing effective strategies to address these technical challenges for commercial realisation of AnMBR for resource recovery.

Insert Figure 8-4#

3.1 Dilute nature of wastewater

Municipal wastewater has a organic carbon content for energy recovery and even lower concentration of nutrient for nitrogen and phosphorus recovery. A wastewater strength of more than 1000 mg COD/L is required to maintain effective activity of anaerobic digester for adequate biogas yield and removal of organic pollutants from wastewater [67]. For economically feasible nutrient recovery, ammonium and phosphate concentrations higher than 5 g NH₄-N/L and 50 mg/L, are necessary. Ammonium and phosphate contents in municipal wastewater are typically less than 0.1 g NH₄-N/L and 10 mg/L [68]. Therefore, it is necessary to pre-concentrate municipal wastewater prior to AnMBR for cost effective recovery of these nutrients.

Potential technologies for pre-concentrating municipal wastewater for subsequent resource recovery include direct membrane filtration [69] and FO [20]. Kimura et al., [69] successfully demonstrated a direct membrane filtration system that could recovery 75% of the dissolved organic carbon and pre-concentrate wastewater by 50 folds (based on volume). Frequent back washing was required to prevent membrane fouling. FO is another alternative for pre-concentrating wastewater for subsequent AnMBR treatment. Vu et al., [70] showed that most of organic carbon in wastewater can be retained by FO during the pre-concentration process. It

is also noteworthy that membrane fouling remains a major technical challenge to the practical applications of both direct membrane filtration and FO for wastewater pre-concentration.

3.2 Temperature

AnMBR can be operated under either thermophilic (50 – 60 °C) or mesophilic (30 – 40 °C) conditions [33, 71]. Psychrophilic condition (10 – 20 °C) is not suitable for municipal wastewater treatment. Thus, AnMBR treatment of municipal wastewater may not be feasible in cold regions, where the energy demand for heating to to a mesophilic condition can be excessive.

AnMBR operation at low temperature can also result in several operational issues including aggravated membrane fouling, slow contaminant biodegradation, and CH₄ loss due to high solubility in the treated effluent. Hydrolysis of particulate matter into dissolved organics is also limited at low temperature, resulting in the accumulation of suspended solids in the reactor and a decrease in methanogenic activity. Martinez-Sosa et al., [33] observed an increase in the total suspended solids content and soluble COD in the bioreactor when the temperature of AnMBR was reduced from 35 to 20 °C, leading to severe membrane fouling and decreased CH₄ production. Decreased CH₄ production could also be attributed to its increased solubility in the effluent when the temperature decreased to 20 °C. Low temperature also leads to an increase in the mixed liquor viscosity, thus requiring more energy for mixing and pumping.

3.3 High salinity wastewater

AnMBR performance can be negatively affected by salinity with respect to biogas production and contaminants (possibly including TrOCs) removal when treating highly saline wastewater from cheese production and seafood processing [72]. High salinity could result in enzyme inhibition, reduce biological activity decline, and cause plasmolysis to anaerobic microbes, thereby negatively affecting the anaerobic digestion process [73]. Ng et al., [74] reported that under a saline concentration the CH₄ yield of AnMBR was reduced to less than 160 L/kg COD_{removed} when treating pharmaceutical wastewater due to the disruption of normal metabolic functions and degradation kinetics. Song et al., [75] also reported the adverse effects of salinity increase (up to 15 g/L NaCl) on biogas production and TrOCs removal by AnMBR.

3.4 Inhibitory substances

AnMBR can be susceptible to some inhibitory substances, particularly sulphate and free ammonia and sulphate, in wastewater. Ammonia is produced from the biodegradation of nitrogenous organics, mostly proteins in wastewater, during anaerobic digestion [73]. Ammonia toxicity (> 3500 mg/L) within an anaerobic digester could be attributed to direct inhibition to the activity of cytosolic enzymes as well as an increase in the intracellular pH and/or the concentration of other cations, such as potassium [76]. The observed inhibition was due to free ammonia in solution rather than ammonium ions. It is noted that in the aqueous phase, there is an equilibrium between free ammonia and ammonium ion depending on pH and temperature [73]. Free ammonia is much more toxic than ammonium ion, because free ammonia can readily penetrate through the cell membrane and thus result in the disruption of cellular homeostasis,

potassium deficiency and/or proton imbalance. High temperature and pH value can also exacerbate free ammonia inhibition since the equilibrium is shifted toward free ammonia [77].

The AnMBR process can also be inhibited by a high sulphate content in wastewater. Such inhibition can be explained by the competition between sulphate reducing bacteria ($2 \text{ g COD/g SO}_4\text{-S}_{\text{removed}}$) and methanogens for available carbon [22]. Furthermore, sulphate can induce the precipitation of non-alkaline metals in anaerobic reactors, reducing their availability as micro-nutrients for methane producing microbes [78, 79]. In the anaerobic condition, sulphate is reduced to hydrogen sulphide (H_2S), which is a corrosive, malodourous, and toxic gas [80-82]. Similar to free ammonia, H_2S can also penetrate through microbial cell membrane and denature native proteins inside the cytoplasm producing sulphide and disulphide cross-links between polypeptide chains [79].

The inhibition of AnMBR by free ammonia and sulphate can be mitigated by organic carbon addition to the wastewater. Meabe et al., [77] demonstrated that longer SRT could allow for sufficient acclimatization of biomass to resist ammonia inhibition during AnMBR treatment. Thus, Meabe et al., [77] did not observe any critical ammonia inhibition threshold for both mesophilic and thermophilic AnMBR in their study. Tian et al., [83] developed a stepwise acclimation strategy to allow anaerobic communities to adapt to $10 \text{ g NH}_4^+\text{-N/L}$ in mesophilic CSTR anaerobic system. The negative impact of sulphate is also insignificant provided that the ratio of COD and SO_4^{2-} is above 10 [84]. It is noteworthy that in a few cases, sulphate addition may be beneficial to methane production by boosting the degradation of propionic acid [85]. Song et al., [86] examined the effect of sulphate concentration on the performance of AnMBR

and reported that basic biological performance of AnMBR was not impacted by the increasing sulphate concentration when the influent COD/SO₄²⁻ ratio was maintained at higher than 10. Nevertheless, H₂S content increased significantly in the produced biogas and membrane fouling was aggravated with sulphate addition [86]. Thus, physicochemical techniques (e.g. stripping, pH adjustment, and precipitation) are recommended to reduce sulphate load to AnMBR to ensure good biogas quality and sustain membrane performance [87].

3.5 Membrane fouling

Membrane fouling is still a vexing challenge to the advancement of AnMBR due to the significantly high cost of fouling control and membrane cleaning. Fouling is caused by the accumulation of inorganic and organic foulants internally in membrane pores and externally on the membrane surface. Membrane fouling can lead to water flux reduction, transmembrane pressure increase, and consequently necessitate chemical cleaning and even membrane replacement. The primary foulants of interest in AnMBR include suspended biomass, colloidal solids, SMP, EPS, attached microbes, and inorganic precipitates (e.g. struvite) [15]. Jun et al., [88] conducted a long-term study of continuous operation of AnMBR over two years and encountered frequent, sudden irreversible fouling. They ascribed the observation to biologically induced mineral scaling and concluded that intense chemical cleaning was required to recover membrane permeability.

Membrane fouling during AnMBR treatment is governed mainly by membrane properties and operational conditions (e.g. water flux, operating temperature, HRT, and SRT), hydrodynamics, and wastewater characteristics. Lin et al., [42] reported that the filtration resistance in

thermophilic AnMBR was 5 – 10 times higher than that of the mesophilic system when both systems were operated under similar hydrodynamic conditions. The observation by Lin et al., [42] was due to more SMP, biopolymer clusters, and fine flocs ($< 15 \mu\text{m}$) being formed under the thermophilic conditions. Huang et al., [89] observed that a decrease in HRT enhanced biomass growth and SMP accumulation and at the same time longer SRT reduced the flocculation of particulates and particle size, thereby exacerbating membrane fouling. These previous studies suggest that membrane fouling in AnMBR can be mitigated to some extent by optimising the operational conditions.

Several techniques to control membrane fouling during AnMBR operation have been reported. In side-stream AnMBR, high cross-flow velocity can be applied to reduce foulant build-up on the membrane surface; while fouling control can be accomplished through biogas sparging for the submerged configuration. Stuckey [17] reported that the addition of powdered or granular activated carbon could effectively reduce membrane fouling in AnMBR, however, their long-term effects on membrane integrity have yet been demonstrated. Pre-treatment, membrane relaxation, and sub-critical flux operation can also be effective to control membrane fouling for AnMBR.

In many cases, even if fouling control is effective, membrane cleaning may still be necessary. Membrane cleaning can be categorised as physical, chemical, and biological cleaning. Physical membrane cleaning include backwashing, surface flushing, and ultrasonication [31]. When physical cleaning alone is not adequate, chemical cleaning is necessary to further remove fouling layers using suitable agents, such as sodium hypochlorite, acids (e.g. hydrochloric acid,

nitric acid, citric acid), caustic (e.g. sodium hydroxide), and chelating agents (e.g. EDTA) to target specific membrane foulants. Mei et al., [90] developed a chemical enhanced backflush cleaning method for both ex-situ and in-situ membrane cleaning during AnMBR operation using NaOH solution at certain concentrations (12 mmol/L).

3.6 TrOCs removal

As discussed in section 2.3, current understanding of the removal of TrOCs by AnMBR is still limited. Research to date that the removal of certain TrOCs by AnMBR may not be sufficient for water reuse application and post treatment of AnMBR effluent is necessary. Further research into the role of microbial community in governing biodegradation of TrOCs during AnMBR can yield new and useful insight to improve the process performance.

4. Conclusion

AnMBR can offer a promising platform to achieve simultaneous wastewater treatment and resource recovery. Nevertheless, the uptake of AnMBR at an industrial scale is still limited due to a host of technical challenges that are to be resolved by future research. Utilising high retention membrane separation processes such as FO and MD for AnMBR is a promising approach to complement the low contaminant removal performance of anaerobic treatment and the diluted nature of wastewater. It is however noteworthy that FO technology is also at an early stage of development. Moreover, wastewater pre-concentration can result in the accumulation of inhibitory substances (e.g. salts, free ammonia, and sulphate) in the AnMBR process. Thus, it is necessary to develop new techniques to remove these inhibitory substances to ensure stable AnMBR performance for treating concentrated wastewater.

Membrane fouling in AnMBR is often more severe compared to aerobic MBR due to the absence of aeration and lower sludge filterability. Thus, in addition to the optimisation of operational parameters, it is also necessary to develop advanced techniques to control membrane fouling during AnMBR operation. Using an alternative membrane process with a low fouling propensity, such as FO, is a promising strategy, which can also enhance contaminant removal in comparison to MF and UF membranes that are commonly used for AnMBR.

CH₄ loss due to dissolution in effluent is also a hurdle for the development of AnMBR, particularly for municipal wastewater treatment. Several approaches including air stripping and membrane separation have been proposed for the recovery of CH₄ from AnMBR effluent. Nevertheless, their feasibility has not yet been fully evaluated in terms of economic viability and process safety. Thus, continued efforts should be devoted to this aspect to address this technical hurdle for a broad application of AnMBR.

There remains some uncertainties regarding the removal of TrOCs by AnMBR. For water reuse applications, post treatment of AnMBR effluent may be necessary. In particular, further research into the role of microbial community in governing biodegradation of TrOCs during AnMBR can yield new and useful insight to further develop this technology for resource recovery and water reuse applications.

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Table 8-1: Comparison between AnMBR and aerobic MBR for wastewater treatment.

Feature	AnMBR	MBR	Reference
Energy consumption (kWh/m ³)	0.03 – 5.7 ^a	~ 2 ^b	[14]
Biomass concentration (g/L) ^c	10 – 40	5 – 20	[12]
Organic loading rate (kg COD/L)	0.17 – 35.5	0.25 – 0.8	[5, 31]
Organic removal efficiency (%)	> 90	> 95	[15, 32]
Hydraulic retention time (hours)	> 8	4 – 8	[12, 19]
Sludge retention time (day)	> 100	5 – 20	[12, 30]
Operational temperature (°C)	20 – 50	20 – 30	[33, 34]
Sludge yield (VSS/g COD _{removed})	0.04-0.09	0.25-0.4	[35, 36]
CH ₄ production (L/kg COD _{removed})	130-460	NA	[35]

^a Energy consumption was calculated for submerged AnMBR treating wastewater with strength between 0.27 and 10 g COD/L;

^b Energy consumption was calculated for submerged MBR treating wastewater with strength between 0.3 and 1.0 g COD/L;

^c Biomass concentration was on the basis of mixed liquor suspended solids content.

Table 8-2: Examples of AnMBR performance regarding organic removal and methane production (From [38]).

Wastewater	Bioreactor configuration	Membrane integration	Organic removal (COD, %)	Methane yield (L/kg COD)	Reference
Meat packing wastewater	N.A	Submerged MF	88 – 98	130 – 180	[41]
Kraft evaporator condensate	UASB	Submerged MF	97 – 99	290 – 310	[42]
Raw tannery	N.A	Submerged MF	90	160	[43]
Real municipal	CSTR	Side-stream MF	86 – 88	300	[44]
Pre-concentrated synthetic wastewater	N.A	Submerged MF	96	223	[45]
Food wastewater	N.A	Side-stream MF	81 – 94	136	[46]
Landfill leachate	N.A	Submerged UF	90	460	[47]

* UASB: up-flow anaerobic sludge reactor; CSTR: continuous stirred-tank reactor; N.A: information is not available.

Table 8-3: Removal efficiencies (mean \pm standard deviation of 10 – 16 measurements) of nitrogen or sulphur bearing TrOCs (From Wijekoon et al., [63] and Tadkaew et al., [54]).

Compound	Log <i>D</i> at pH 7	Number of N/S in molecular structure	Removal (%)	
			AnMBR- Wijekoon et al. (2015)	Aerobic MBR- Tadkaew et al. (2011)
Atenolol	-2.09	2N	76.5 \pm 10.5	96.9 \pm 0.2
Caffeine	-0.63	4N	90.4 \pm 3.6	49.6 \pm 4
Sulfamethoxazole	-0.22	3N	99.6 \pm 0.2	91.9 \pm 0.6
Trimethoprim	0.27	4N	97.5 \pm 0.7	16.6 \pm 3.7
Paracetamol	0.47	1N	85.9 \pm 5.2	95.1 \pm 3.4
Primidone	0.83	2N	16.6 \pm 11.5	12.4 \pm 4.3
Triamterene	1.03	7N	75.3 \pm 9.9	27.9 \pm 6.3
Diclofenac	1.77	1N	2.8 \pm 1.7	17.3 \pm 4.2
Carbamazepine	1.89	2N	39.2 \pm 21.2	13.4 \pm 4.3
Amitriptyline	2.28	1N	99.6 \pm 0.2	97.8 \pm 0.8
Omeprazole	2.35	3N and 1S	99.9	62.1 \pm 3.5
DEET	2.42	1N	19.5 \pm 24.2	4.6 \pm 2.7
Atrazine	2.64	2N	56.9 \pm 19.6	4.4 \pm 3.7
Linuron	3.12	2N	88.1 \pm 3.2	21.1 \pm 4.1
Clozapine	3.23	4N	99.6 \pm 0.4	84.8 \pm 5.4
Triclocarbon	6.07	2N and 2Cl	95.6 \pm 5.7	> 98
Simazine	2.28	5N	54.1 \pm 5.3	–
Diuron	2.68	2N	43.1 \pm 23.5	–
Diazepam	2.8	2N and 1Cl	61.6 \pm 18.2	–
Diazinon	3.77	2N and 1S	93.0 \pm 2.4	–
Bisphenol A	3.64	–	99	90.4
Triclosan	5.28	–	70	>91.8
4-n-nonylphenol	6.14	–	94	99.3
Ketoprofen	0.19	–	27 \pm 21	70.5
Naproxen	0.73	–	74 \pm 14	40.1 \pm 2.8
Ibuprofen	0.94	–	25 \pm 24	96.7 \pm 0.7
Gemfibrozil	2.07	–	12 \pm 11	98.9 \pm 0.1

LIST OF FIGURES

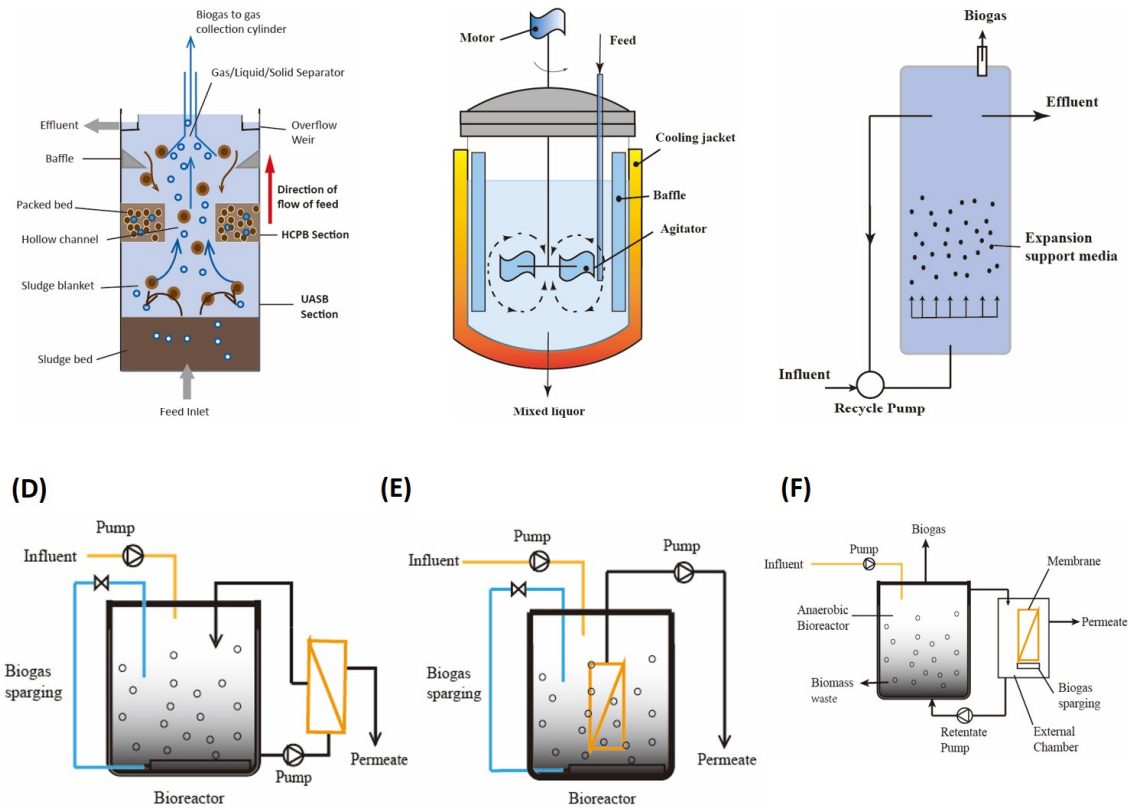


Figure 8-1: Typical anaerobic bioreactors (A: up-flow anaerobic sludge reactor; B: continuous stirred-tank reactor; C: anaerobic fluidized bed reactor) and their integration with membrane separation process in the (D) side-stream, (E) submerged and (F) external chamber modes (Modified from: [38]).

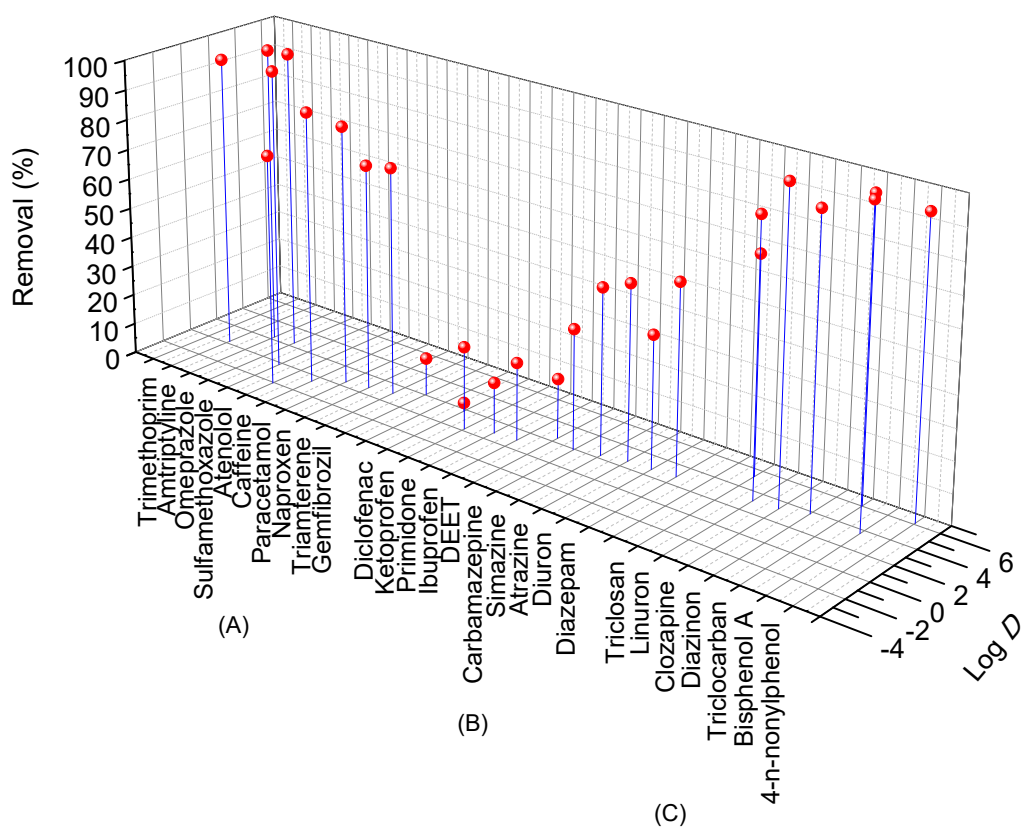


Figure 8-2: Aqueous phase removal of TrOCs according to hydrophobicity and functional groups (A) hydrophilic contaminants with EDGs only (B) hydrophilic contaminants with EWGs, and (C) Hydrophobic contaminants. Removal efficiency represents the average value of duplicate samples taken once a week for five weeks. Log D denotes the values at pH 7 (From [63]).

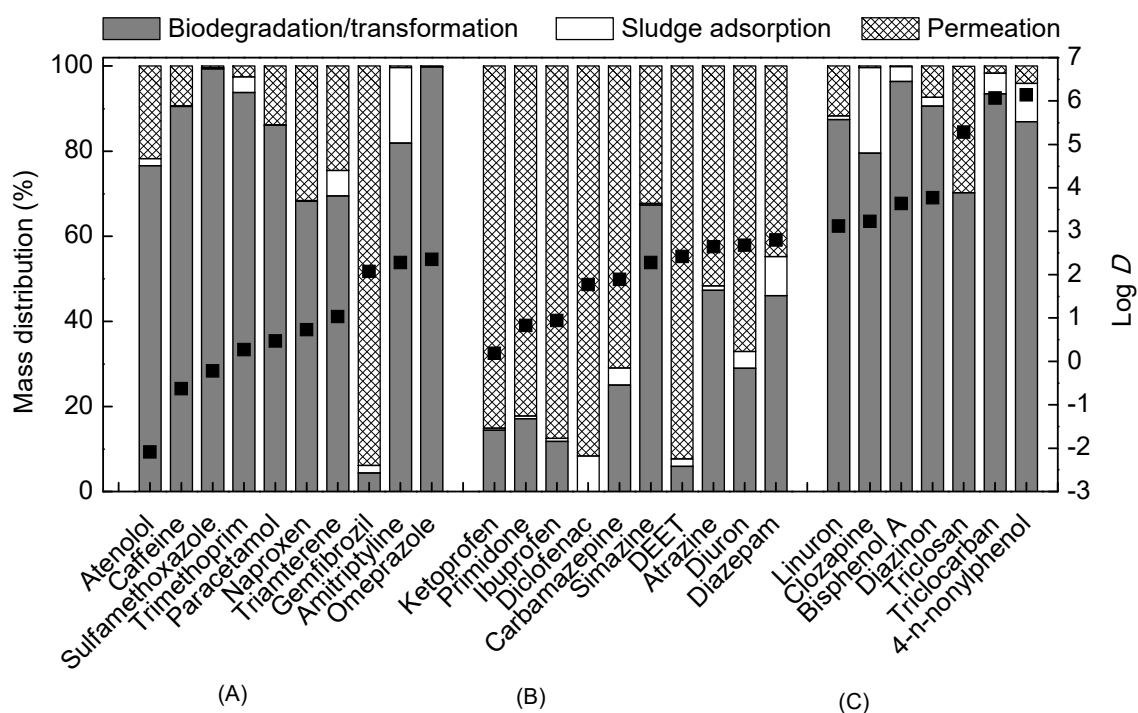


Figure 8-3: Removal mechanisms of TrOCs during AnMBR treatment according to hydrophobicity and functional groups; (A) hydrophilic contaminants with EDGs, (B) hydrophilic contaminants EWGs, and (C) hydrophobic contaminants (From [63]).



Figure 8-4: Key challenges to the development of AnMBR for wastewater treatment and resource recovery (From: [38]).