

Review

# Wastewater Treatment and Biogas Recovery Using Anaerobic Membrane Bioreactors (AnMBRs): Strategies and Achievements

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Abstract: Anaerobic digestion is one of the most essential treatment technologies applied to industrial and municipal wastewater treatment. Membrane-coupled anaerobic bioreactors have been used as one alternative to the conventional anaerobic digestion process. They are presumed to offer the advantage of completely reducing or minimizing the volume of sludge and increasing biogas production. However, researchers have consistently reported different kinds of fouling that resulted in the reduction of membrane life span. Depending on the strength of the effluent, factors such as high suspended and dissolved solids, fats, oil and grease, transmembrane pressure (TMP) and flux were reported as major contributors to the membrane fouling. Moreover, extracellular polymeric substances (EPSs) are an important biological substance that defines the properties of sludge flocs, including adhesion, hydrophobicity and settling and have been found to accelerate membrane fouling as well. Extensive studies of AnMBR have been done at laboratory while little is reported at the pilot scale. The significance of factors such as organic loading rates (OLRs), hydraulic retention time (HRT), pH and temperature on the operations of AnMBRs have been discussed. Microbial environmental conditions also played the most important role in the production of biogas and the chemical oxygen demand (COD) removal, but adverse effects of volatile fatty acids formation were reported as the main inhibitory effect. Generally, evaluating the potential parameters and most cost effective technology involved in the production of biogas and its inhibitory effects as well as the effluent quality after treatment is technically challenging, thus future research perspectives relating to food to microorganism F/M ratio interaction, sufficient biofilm within the reactor for microbial attachment was recommended. For the purpose of energy savings and meeting water quality discharge limit, the use of micro filtration was also proposed.

Keywords: anaerobic digestion; biogas production; wastewater treatment; membrane bioreactors

# 1. Introduction

Over the past century, there has been quite a number of studies on the most economic, efficient and environmental friendly wastewater treatment technologies. Conventional aerobic methods have existed for over a century now, but they have major drawbacks that include sludge production, high energy use for aeration, large operating space and a higher maintenance cost. Moreover, the systems are characterized by uncontrolled release of potential atmospheric greenhouse gases



such as methane (CH<sub>4</sub>), carbon dioxide (CO<sub>2</sub>), and nitrogen oxide (N<sub>2</sub>O) that contribute immensely to deterioration of the environment [1], although in the aerobic process quality effluents are produced which comply with standards limits set by the different regulatory bodies, especially in developed countries. However, one obvious disadvantage is the lack of material and energy recovery [2,3]. Hence, interest continues to grow in finding the best alternative.

The advent of anaerobic digestion in the field of wastewater treatment marks the beginning of economic and efficient technology [4]. This is seen in the quality of the effluents discharged, material recovered and energy generated as well as the mode of sludge production, handling and processing [5]. One of the early constraints detected from the onset of this technology was the long hydraulic retention time coupled with the slow growing methanogenic bacteria [6]. However, towards attaining higher efficiency, research focus was moved towards coupling anaerobic bioreactors with membrane filtration units. These systems were seen to offer a more unique and prudent technique over conventional anaerobic method [7]. It simply combines the anaerobic process and membrane technology operating simultaneously. Table 1 presents a comparison of conventional anaerobic treatment and anaerobic membrane bioreactor (AnMBR).

 Table 1. Comparison of conventional anaerobic treatment and anaerobic membrane bioreactor (AnMBR) [8].

Feature	<b>Conventional Anaerobic Treatment</b>	AnMBR
Discharge quality	Moderate-Poor	High
Sludge volume	low	low
substrate loading concentration	High	High
Removal efficiency (Effluent)	High	High
Biomass retention	Low	Complete
Footprint	High-Moderate	Low
Alkalinity Requirement	Depends on microbial activity	Depends on microbial activity
Nutrient requirement	Low	Low
Startup period	2–4 Months	Less than 2 Weeks
Temperature requirement	Low-Moderate	Low-Moderate
Energy input	Low	Low
Pre-treatment requirement	Not necessary	Mostly for high solid substrates
Biogas recovery	Yes	Yes

The combination of anaerobic bioreactors with membranes is presumed to reduce the overall energy demand and to facilitate the retention of microorganisms so as to operate with high biomass concentration [9]. Depending on the intended use of the treated effluent, membranes such as microfiltration, ultrafiltration, nano-filtration and reverse osmosis are usually coupled to anaerobic reactors in both industrial and municipal wastewater treatment systems [10]. Buntner et al. [11] studied the combination of an upflow anaerobic sludge blanket (UASB) reactor with ultrafiltration membranes for dairy wastewater treatment at ambient temperature. The intent of this combination was to decrease the COD of the dairy wastewater whilst producing biogas rich in methane and diminish the overall sludge production as well as obtaining high quality effluent. About 150 L/kg COD of biogas was achieved out of which 73% was methane gas. The organic loading rate was up to 4.85 kg COD/m<sup>3</sup>·day and 95% COD removal was realized, reaching 99% during its stable operation period. However, this is lower compared to the work of Deowan et al. [12]. Both studies were reported at pilot scales, but one configured the membrane externally while the other was submerged within the bioreactor. The aim of this review paper is to gain insight knowledge on the strategies and achievements recorded in the recent past on the use of AnMBR technology, since the idea was presented as a possible solution or substitute that might overcome the disadvantages of conventional wastewater treatment in terms of space utilization, superior effluent quality, energy generation and the overall operation and maintenance cost.

#### 2. Fundamentals of AnMBR

An anaerobic reactor coupled with a membrane was first commercialized by Dorr-Oliver in the early 1980s [13]. It was constructed to treat high strength wastewater (whey) and was named anaerobic membrane bioreactor system. Since then, research on the feasibility and treatment efficiencies of AnMBRs continue, most especially on the type of material and configurations for treating low, medium, and high strength wastewater [14–17]. The two forms of membrane configuration includes internal and external, as shown in Figure 1, but the external configuration is the most widely reported.

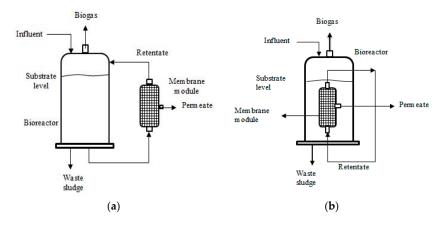


Figure 1. A schematic of AnMBR configurations—(a) Side or external membrane (b) submerged membrane.

#### Membrane Configuration Performance

A pilot scale study of municipal wastewater treatment with an external membrane configuration was reported by Huang et al. [14]. The system achieved COD removal efficiency close to 90%, but a slow and linear increase in the filtration resistance was observed under critical flux conditions and subsequently resulted in fouling due to solid accumulation on the surface of the membrane. However, it was observed that gas sparging and additional shear were needed to control membrane fouling. According to Chu et al. [10], high shear stress may interfere with the biological activities during anaerobic digestion of biomass. Furthermore, studies of Zhao et al. [15] showed that the external membrane configuration system is easier to maintain and monitor. However, these require more energy with high hydraulic shear force which might also disrupt anaerobic bio-solids favoring substrates with smaller particle sizes and this in turn causes membrane fouling. In another development, thorough investigations of submersible membrane module coupled to anaerobic bioreactors were reported in [16–19] and it is believed to overcome the numerous drawbacks of conventional methods of wastewater treatment. However, it also appears that the technology is more suitable for low strength organic loads, especially municipal wastewater as examined in the previous research of [20–22].

Martinez-Sosa et al. [3] also examined a submerged anaerobic membrane bioreactor and found 97% COD removal. The final effluent COD was less than 20 mg/L at volumetric organic loading rate of 0.5 to 12.5 kg/m<sup>3</sup>·day. The system could be termed efficient even though, the temperature fluctuates between 12 °C and 26 °C. Likewise the membrane permeability was not outstanding due to intermittent suction mode and membrane flux that lead to frequent membrane cleaning. Furthermore, investigation of Deowan et al. [12] on a submerged anaerobic membrane bioreactor (SAMBR) treating textile wastewater revealed COD removal efficiency around 90% with negligible fluctuations in some phases. However, 60% color removal was reported in the first phase and subsequently dropped to between 20–50% after the system stabilized. This phenomenon was attributed to color adsorption on the surface of the membrane. In a similar configuration manner, the defects in COD removal were ascribed to the presence of high total nitrogen (TN) and total phosphorus (TP) [23]. In view of that, application of anoxic conditions or an aerobic process or a combination of the two could enhance removal of both TN and TP. Katayon et al. [24], experimented on the effect of vertical and

horizontal membrane configurations, in which two procedures consisting of low and high mixed liquor suspended solids (MLSS) concentrations were considered. The results showed that horizontally placed membrane was able to removed 99.2% total solids and 99.73% turbidity at lower MLSS concentration at a mean flux value of  $5.03 \text{ L/m}^2\text{h}$ . This is quite higher than those found in vertical modules with high MLSS concentrations and mean flux value of  $2.27 \text{ L/m}^2\text{h}$ . Hence, for higher efficiency, maintaining high biomass concentration and sufficient microbial activity with minimal energy utilization would enhance the process of anaerobic membrane bioreactor operation processes.

#### 3. Membrane Performance of Various Wastewater Treatments

#### 3.1. Industrial Wastewater

Industrial regulatory issues towards meeting the stringent water quality discharge permissible limits motivate researchers to study more on finding lasting solutions to the problems associated with the effluent released to the environment. Most industrial wastewaters are regarded as high strength wastewater, because they contain large amounts of settleable, dissolved and suspended solids or other elements such as heavy metals in greater proportion [25,26]. However, the strength of the wastewater could differ from one industry to another owing to the different types of operations. Generally, industrial wastewater comes from streams such as production lines, cooling towers or boiler and cleaning processes that contain diverse substances. These may include organic and inorganic compounds, viruses, bacteria and toxic compounds. Therefore, applying less energy and achieving high COD removal with minimum sludge production is of utmost priority to the water and wastewater treatment industry. For instance, a pilot scale study of SAMBR treating high strength wastewater (raw tannery wastewater) achieved higher COD removal efficiency up to 90% at organic loading rate OLR of 6 g/L·day and biogas yield (0.160 L/g COD removed) [27]. The system performed efficiently, but was strongly characterized by a high hydraulic retention time (HRT) (40 h) and as such, high energy was expended, although the permeate flux remained at (6.8 LMH) which was well below the critical flux (17.5 LMH) as determine in the earlier work of Hu et al. [28]. Similarly, Fush et al. [29] reported treatment of three different high strength wastewater effluents (artificial wastewater, animal slaughterhouse, and sauerkraut brine) using continuous stirrer tank reactor CSTR coupled with membrane filtration units. The COD removal in all the reactors were >90% at OLR of 20 g  $COD/L \cdot day$ , 8 g  $COD/L \cdot day$  and 6–8 g  $COD/L \cdot day$  respectively. The methane yields were in the range of 0.17–0.30, 0.20–0.34, and 0.12–0.32 Ln/g·COD fed. On the other hand, Saddoud et al. [30] showed that an anaerobic cross-flow ultrafiltration membrane bioreactor exhibited high efficiency removal of suspended solid (SS), biochemical oxygen demand BOD<sub>5</sub>, COD and microorganisms. It reached >100%, 90%, 88%, and 100% respectively. Biogas production got to 30 liters per day with average of  $0.27 \text{ L CH4/g} \cdot \text{COD}$  yield. Interestingly, this high yield of biogas was achieved at low organic loading rate (OLR) of 2 g COD/L·day.

Most recently, the filtration performance of an AnMBR treating high strength lipid-rich wastewater and corn-to-ethanol thin stillage was conducted by Dereli et al. [31]. The reactors delivered a high COD removal efficiency of up to 99% under stable operating conditions with an average OLR of 8.3, 7.8, and 6.1 kg COD/m<sup>3</sup>·day. However, the permeate quality turned out to be inferior in quality with increased in solid retention time (SRT). Table 2 present some examples of the biological and membrane performance of different AnMBR applications for treatment of industrial wastewater. Unfortunately, information regarding the membrane performance with respect to biogas production is quite limited in most of the studies.

#### 3.2. Municipal Wastewater

Effluents characterized by low organic strength and high particulate organic matter are mostly reported as municipal wastewater [32]. The ability of AnMBRs in retaining biomass within the reactor has made it a subject of research for municipal wastewater treatment as a possible alternative to the

conventional anaerobic treatment process. According to Ho et al. [33], Kocadagistan and Nazmi [34], bioreactors coupled with membranes used for the treatment of municipal wastewater (MWW) have shown excellent effluent quality that meets the stringent discharge standards in terms of COD, suspended solids and pathogen counts removals when compared to conventional anaerobic methods. Moreover, the use membrane bioreactor (MBR), for anaerobic treatment of municipal wastewater in high temperate climate is still a challenge. This is because, domestic wastewater is usually complex in nature and characterized by high fraction of particulate organic matter. It could be in the form of proteins, suspended solids of different origin, and fatty acids with large to moderate biodegradability portions [35]. Based on these characteristics, operation of MWW treatment reactor under psychrophilic temperature (<20 °C) might be some how difficult. Smith et al. [36] compared simulated and actual domestic wastewater (DWW) at the bench-scale using submerged flat-sheet microfiltration membranes. A psychrophilic temperature of 15 °C was set. An average removal efficiency of 92  $\pm$  5% COD was achieved, which corresponds to an average permeate COD of 36  $\pm$  21 mg/L in simulated DWW, while  $69 \pm 10\%$  was realized during actual DWW treatment. However, it is obvious the membrane in this study utilizes a lot of energy with relatively high concentration of dissolved methane. Although previous references [37–41] have reported the performance of simulated and actual DWW treatments, only a few of them were performed at psychrophilic temperature (15 °C and below). Table 3 summarizes some studies on the use of AnMBRs for domestic wastewater treatment.

Another investigation of Smith et al. [52] showed a simulated domestic wastewater using AnMBR at psychrophilic temperatures of 15, 12, 9, 6, and 3 °C. Remarkably, a total reduction of COD > 95% was realized at a temperature of 6 °C. The success was attributed to viable microbial activity in the membrane biofilm but subsequently give rise to high dissolved methane oversaturation in permeate and consequently fell to 86% at 3 °C. However, decreasing temperature resulted in increased soluble COD in the bioreactor. Thus, this signifies a reduction in suspended biomass activity. Dolejs et al. [53] conducted a research on the effect of psychrophilic temperature shocks on a gas-lift anaerobic membrane bioreactor (Gl-AnMBR) used for treating synthetic domestic wastewater. The stability of the system was measured by transiting between mesophilic and psychrophilic stages having several psychrophilic shocks (12–48 h). The result showed an average COD removal of  $94 \pm 2\%$  at mesophilic with an average methane yield of 0.19 L CH<sub>4</sub>/g COD removed (including psychrophilic shocks). More than 80% of the influent COD accumulated in the reactor under psychrophilic in comparison to 39% under mesophilic conditions.

A pilot scale study of anaerobic urban wastewater treatment in a submerged hollow fiber membrane bioreactor was reported by Gimenez et al. [20]. They assessed the effect of a number of operational variables on both biological and physical separation process performance. Mesophilic temperature of 33 °C, at 70 days SRT, and HRT ranging from 20 h down to 6 h. COD removal stood almost at 90%, with no trace of irrevocable fouling observed but yet the methane yield was very low and this was mainly attributed to influent COD/SO<sub>4</sub>-S ratio. The work of Gouveia et al. [54] on a pilot scale AnMBR coupled to an external ultrafiltration treating MWW also attained COD removal efficiency of  $87 \pm 1\%$  with specific methane yield of 0.18 and 0.23 Nm<sup>3</sup> CH<sub>4</sub>/kg COD removed at a lower temperature of 18  $\pm$  2 °C. More recently, a toxicity reduction in wastewater (synthetic wastewater) aiming at reuse possibilities was performed at relatively psychrophilic temperature (25 °C) using submerged anaerobic membrane bioreactor with forward osmosis membrane (FO-AnMBR) [55]. The FO-AnMBR process exhibited >96% removal of organic carbon, nearly 100% of total phosphorus and 62% of ammonia-nitrogen respectively. This suggests better removal efficiency than the conventional AnMBR. The average COD removal was 96.7% corresponding to the influent COD concentration of 460 mg/L and a methane production of 0.21 L CH4/g COD. Thus, the system demonstrates high feasibility of energy recovery.

Type of System/Module Configuration/Membrane Configuration	Membrane Type/Material/Characteristic	Wastewater Treated	Operating Condition	Reactor Working Volume + Scale	Influent COD mg/L	Effluent COD Mg/L	Maximum COD Removal (%)	Biogas Production (L CH <sub>4</sub> /g COD removed	Reference
AnMBR/External cross flow membrane/Tubular	-/ceramic (ZrO <sub>2</sub> -TiO <sub>2</sub> )/ Pore size = 0.2 µm Area = 0.25 m <sup>2</sup>	Industrial wastewater	Flux = 8.4 L/m <sup>2</sup> ·h, HRT = 1.7–5, 3.5, 3–3.5, SRT = 120–450 day, Temp = 35–37 °C, MLSS = OLR = 2.5 g, COD L/day	V = 50 L S = P	4300	830	78	0.5 and 0.6	[42]
SAMBR/Submerged/Flat sheet and hollow fiber	MF (Polipropilen-PP)/Chlorinated polyethylene/ Pore size = 0.05 μm Area = 0.66 m <sup>2</sup>	Synthetic industrial wastewater	Flux = 3-0.9 L/m <sup>2</sup> ·h, HRT = 390, 167, 168, SRT = ∞, Temp = 35 °C, MLSS = OLR = 0.3-0.54 g, COD/L·day	V = 4 L S = L	20,000–23,000	15.2	85–90	-	[43]
Submerged anaerobic membrane bioreactor (SAnMBR)/Submerged/ Hollow fiber	MF/Curtain-type/ Pore size = 0.2 μm Area 5.4 m <sup>2</sup>	Synthetic industrial wastewater	Flux = 6 L/m <sup>2</sup> ·h, HRT = 2.2 h, SRT = -, Temp = 35 °C, MLSS =10.9 g/L, OLR = 3.0 kg COD/m <sup>3</sup> ·day	V = 25 L S = P	223 ± 111	50 ± 22	87	0.12	[44]
SAnMBR/Submerged/ Hollow fiber	MF/Curtain-type/ Pore size = 0.4 µm Area = 0.040 m <sup>2</sup>	Paper mill wastewater	Flux = 7.2 L/m <sup>2</sup> ·h, HRT = 35 h, SRT = 40 day, Temp = 21 °C, MLSS = 12.90 g/L, OLR = 7.0 kg COD/m <sup>3</sup> ·day	V = 10 L S = L	11,415 ± 15	228.3 ± 5	98	-	[45]
AnMBR	UF/Hollow fiber/Polyvinylidene fluoride (PVDF) Pore size = - Area = 20 m <sup>2</sup>	Synthetic anti-biotic solvent	Flux = 20 L/m <sup>2</sup> ·h, HRT = 48, 36, 24, and 18 h, Temp = $35 \pm 1$ °C, MLSS = 16.5–12.4 g/L, OLR = 3.9–12.7 kg, COD/m <sup>3</sup> ·day	$V = 4.4 \text{ m}^3$ $S = P$	7892–21,986	8056.5 1218.2 1523.5 1828.3 2207.7	93.6 and 98.7	94 and 130.7	[46]
AnMBR	UF/Hollow fiber/ Polyvinylidene fluoride (PVDF) Pore size = - Area = 20 m <sup>2</sup>	Synthetic anti-biotic solvent	Flux = 20 L/m <sup>2</sup> ·h, HRT = 48 h, Temp = 37 °C, MLSS = 52.2 g/L, OLR = 3.79 kg , COD/m <sup>3</sup> ·day	$V = 4.4 \text{ m}^3$ $S = P$	15,000–25,000	4000	96.5	-	[47]
AnMBR	UF/Hollow fiber/ Polyvinylidene fluoride (PVDF) Pore size = - Area = 20 m <sup>2</sup>	Synthetic anti-biotic solvent	Flux = 20 L/m <sup>2</sup> ·h, HRT = 48–24 h, Temp = $(35 \pm 3 \ ^{\circ}C, 25 \pm 3 \ ^{\circ}C, 15 \pm 3 \ ^{\circ}C, 25 \pm 3 \ ^{\circ}C),$ MLSS = 16.5 g/L, OLR = 10.0 kg, COD/m <sup>3</sup> ·day	$V = 4.4 \text{ m}^3$ $S = P$	1000–25,000	-	95	-	[48]
AnMBR	UF/Hollow fiber/ Pore size = 0.04 μm Area = 0.047 m <sup>2</sup>	Brewery wastewater	Flux = 8.64 ± 0.69, L/m <sup>2</sup> ·h, HRT = 44 h, Temp = (35 °C), MLSS = 2.8 g/L, OLR = 3.5–11.5 g , COD/L·d	V = 15 L S = L	19,100	171	99	$0.53\pm0.015$	[49]

# Table 2. References on anaerobic membrane bioreactors (AnMBRs) for industrial wastewater.

Type of System/Module Configuration/Membrane Configuration	Membrane Type/Material/Characteristic	Wastewater Treated	Operating Condition	Reactor Working Volume + Scale	Influent COD mg/L	Effluent COD Mg/L	Maximum COD Removal (%)	Biogas Production (L CH <sub>4</sub> /g COD removed	Reference
AnMBR and B-AnMBR	UF/Hollow fiber/ Polyvinylidene fluoride (PVDF) Pore size = 0.02 μm Area = 0.07 m	Bamboo wastewater	Flux = $33.4-16.2 L/m^2 \cdot h$ , HRT = $3 d$ , Temp = $32 \pm 2 °C$ , MLSS = $16 g/L$ , OLR = $6 kg$ , COD/m <sup>3</sup> ·day	V = 15 L S = L	17,160 ± 814	278.9 ± 4.2 mg/L and 125.3 ± 3.2 mg/L	$94.5\pm2.9$ and $89.1\pm3.1$	$\begin{array}{c} 13.2 \pm 1.2, \\ 10.3 \pm 0.8 \end{array}$	[50]
C-AnMBR and B-AnMBR	UF/Hollow fiber/ Polyvinylidene fluoride (PVDF) Pore size = $0.04 \ \mu m$ Area = $0.047 \ m^2$	Pharmaceutical wastewater	$\label{eq:hardward} \begin{split} Flux &= 6 \ L/m^2 \cdot h, \\ HRT &= 30.6 \ h, \\ Temp &= 27 \ \pm 1.0 \ ^\circ C, \\ MLSS &= 9500 - 10,200 \ and \ 10,000 \ mg/L, \\ OLR &= 13.0 \ \pm 0.6 \ kg, \ COD/m^3 \cdot day \end{split}$	V = 10 L S = L	16,249 ± 714	$8723\pm593$ and $6432\pm445$	$46.1\pm2.9$ and $60.3\pm2.8$	$24.5 \pm 2.1$ And 17.6 $\pm 1.5$	[51]

AnMBR = anaerobic membrane bioreactor, COD = chemical oxygen demand, HRT = hydraulic retention time, MLSS = Mixed liquor suspended solids, OLR = organic loading rate, P = pilot scale, - = not reported.

Table 3. References on anaerobic membrane bioreactors (AnMBRs) for municipal wastewater treatment.

Type of System/Module Configuration/Membrane Configuration	Membrane Type/Material/Characteristic	Wastewater Treated	Operating Condition	Reactor Working Volume + Scale	Influent COD mg/L	Effluent COD mg/L	Maximum COD Removal (%)	Biogas Production (L CH4/g COD removed)	Reference
AFMBR/Submerged/ Hollow fiber	-/Non-woven Fibrous (chlorinated polyethylene)/ Pore size = 0.1 μm Area 0.022 m <sup>2</sup>	Raw municipal wastewater	Flux = 30 L/m <sup>2</sup> ·h, HRT = 4 h, SRT = $\infty$ , Temp = 23 $\pm$ 1 °C, MLSS = -, OLR = 3-6	V = 2.7 L S = L	-	23.5	97.07	2.11	[56]
SAMBRs/Submerged/ Hollow fiber	MF/Polyvinylidene fluoride/ Pore size = 0.02 μm Area = -	Municipal (wastewater from the ethanol fermentation of food waste)	Flux = 9.72 L/m <sup>2</sup> ·h, HRT 110 h, SRT = -, Temp = 23–25 °C, MLSS = 8–12 g/L, OLR =2.3–3.6 g/L·day	V = 0.628 L S = L	15,000 ± 1000	$202\pm23$	98.2	-	[57]
UASB/Submerged/Tubular	UF/Polyvinylidene fluoride/ Pore size = - Area = 0.2375 m <sup>2</sup>	Raw municipal wastewater	Flux = 2.5 L/m <sup>2</sup> ·h, HRT = 8 h, Temp = 18–21 °C, MLSS = -, OLR = -	$V = 0.7 \text{ m}^3$ $S = P$	$525 \pm 174, \\657 \pm 235$	$222 \pm 61, \\ 130 \pm 55$	68.6	-	[58]
CG-AnMBR and SG-AnMBR/Submerged/ Hollow fiber	-/A polyvinylidence (PVDF)/ Pore size = 0.22 μm Area = 0.06 m <sup>2</sup>	Synthetic domestic wastewater	Flux = 5.3 L/m <sup>2</sup> ·h, HRT = 12 h, SRT = 25–30 day, Temp = 20 °C, MLSS = $20.50 \pm 1.53$ g/L, OLR = -	V = 3 L S = L	330–370	-	90	$156.3\pm5.8$	[59]
MBR/Submerged/Flat sheet	MF/-/ Pore size = 0.4 μm Area = 16 m <sup>2</sup>	Real municipal wastewater	Flux = 7.8 L/m <sup>2</sup> ·h HRT = 35 h, SRT = 25–30 day Temp = 6.5–21 °C/7.0–20 °C MLSS = 5300–9800 mg/L OLR = -	$V = 3 m^3$ $S = P$	896 GC mL <sup>-1</sup>	18.7 ± 2.9	93	-	[60]

# Table 3. Cont.

Type of System/Module Configuration/Membrane Configuration	Membrane Type/Material/Characteristic	Wastewater Treated	Operating Condition	Reactor Working Volume + Scale	Influent COD mg/L	Effluent COD mg/L	Maximum COD Removal (%)	Biogas Production (L CH <sub>4</sub> /g COD removed)	Reference
AnCMBRs/Submerged/ Flat sheet	-/Ceramic membrane/ Pore size = 80 nm Area = 0.08 m <sup>2</sup>	Domestic wastewater	$Flux = 8 L/m^{2} \cdot h, HRT = 5.8 h, SRT = 60 day,$ Temp = 25 °C, MLSS = -, OLR =10.0 kg COD/m <sup>3</sup> · day	V = 3.6 L S = L	$417\pm 61$	-	87	-	[61]
SAnMBR/Submerged/Flat-sheet	-/Polyethylene terephthalate/ Pore size = 0.2 μm Area = 0.116 m <sup>2</sup>	Synthetic municipal wastewater (alcohol ethoxylates)	Flux = -, HRT = 42–12 h SRT = -, Temp = 25 ± 1 °C MLSS = -, OLR = 3.0–6.0 kg COD/m <sup>3</sup> ·day	V = 6 L S = L	-	17.1	95.5–98.8	2.30-4.25	[62]
SAnMBR/Submerged/Flat-sheet	UF/Polyvinylidene fluoride/ Pore size =0.2 μm Area 0.735 m <sup>2</sup>	Domestic wastewater	Flux = 8.3–9.5 L/m <sup>2</sup> ·h and 6.0–6.7 L/m <sup>2</sup> ·h, HRT = 5.8–4.8 and 8.0–7.1 h, SRT = 50 day, Temp = 35 °C and 25 °C, MLSS = -, OLR = 0.43–0.90 kg COD/m <sup>3</sup> ·day	V = 15 L S = L	400	-	90	276 ± 13	[63]
AnMBR/External/Tubular	UF/Polyethersulfone/ Pore size = 30 μm Area 0.11 m <sup>2</sup>	Synthetic municipal wastewater	Flux 12.3 L/m <sup>2</sup> ·h, HRT = 6 h, SRT = 126 day, Temp = 25 °C and 15 °C, MLSS = -, OLR = 2 kg/m <sup>3</sup> ·day	V = 7 L S = L	530 ± 30	42 and 52	92 and 90	-	[64]
AnMBR/External/Tubular	UF/Polyethersulfone/ Pore size = 30 µm Area 0.0038 m <sup>2</sup>	Synthetic municipal wastewater	Flux 12.3 L/m <sup>2</sup> ·h, HRT = 6 h, SRT = 126 day, Temp = 25–15 °C, MLSS = -, OLR = 2 kg/m <sup>3</sup> ·day	V = 7 L S = L	530 ± 30	$149 \pm 5.9$ to $42 \pm 4.4$ ,	92	-	[65]

AnMBR = anaerobic membrane bioreactor, COD = chemical oxygen demand, HRT = hydraulic retention time, MLSS = Mixed liquor suspended solids, OLR = organic loading rate, P = pilot scale, - = not reported.

#### 3.3. Synthetic Wastewater

Compounds such as starch, glucose, molasses, peptone, yeast, and cellulose are usually used as synthetic substrates to test new concept of AnMBR. The results of a number of studies are summarized in Table 4. The COD removal efficiencies of those investigations were generally >90%, with OLR less than 10 kg COD/m<sup>3</sup>·day. Effect of HRT and SRT on treatment performance of submerged AnMBR for synthetic low-strength wastewater reveals a total COD removal efficiencies higher than 97% at all the operating conditions with a maximum biogas production in terms of mixed liquor volatile suspended solid (MLVSS) removals (0.056 L CH4/g MLVSS) at infinite SRT [66]. Though increasing in OLR with short hydraulic time HRT and long SRT boosted the methanogenic environment, but membrane fouling was worsened due to a decrease in HRT which heightened the growth of biomass and accumulation of soluble microbial products (SMP). The experiment performed by Jeison et al. [67] on synthetic wastewater using submerged anaerobic membrane bioreactor showed a reversible cake layer formation on short term basis. Additionally, cake consolidation was detected during a long-term operation at a flux close to the critical point. Remarkably, increasing OLR from 50–60 g COD/L·day towards the end of the operation presented high COD removal efficiency greater than 90%.

Fallah et al. [68] reported a significant COD removal of (<99%). In their studies, they considered two hydraulic retention times (24 h and 18 h) using a MBR to remove styrene from a synthetic wastewater having a chemical oxygen demand and styrene concentration of 1500 mg/L and 50 mg/L. nonetheless, reduction in HRT to 18 h caused a release of extracellular polymeric substance (EPS) from the bacterial cells that led to the rise in soluble microbial product (SMP) and sludge deflocculation. Moreover, the dramatic rise in transmembrane pressure TMP which was operating fairly low and constant for a number of days give rise to severe membrane fouling. This trend was attributed to the rise in SMP concentrations and the decrease in mean floc size. Similar research was reported in Ho et al. [69]. The HRT were varied during treatment of synthetic municipal wastewater. The permeate quality was outstanding, irrespective of HRT differences with over 90% COD removal at HRT of 6 h. Methane produce was 0.21 to 0.22 CH<sub>4</sub>/g COD removed. Conversely, the fraction of methane recovered from the synthetic municipal wastewater declined from 48 to 35% with the reduction of HRT from 12 to 6 h. Subsequently, the result of the increased mixed-liquor soluble COD which was precluded and accumulated in the AnMBR drastically affects the performance.

Type of System/Module Configuration/Membrane Configuration	Membrane Type/Material/Characteristic	Wastewater Treated	Operating Condition	Reactor Working Volume + Scale	Influent COD mg/L	Effluent COD mg/L	Maximum COD Removal (%)	Biogas Production (L CH <sub>4</sub> /g CODremoved)	Reference
SAMBR/ Submerged/ Flat sheet	-/Non-woven fibrous (chlorinated polyethylene)/ Pore size = 0.2 µm Area = 0.116 m <sup>2</sup>	Synthetic municipal wastewater (linear alkyl benzene sulfonate concentration in sewage)	Flux = -, HRT = 24–12 h, SRT = Temp = 25 ± 1 °C MLSS = -, OLR = 3–6 kg COD/m <sup>3</sup> ·day	V = 6 L S = L	-	23.5	97.07	2.11	[74]
SAMBRs/ Submerged/ Flat sheet	MF/Polymethyl methacrylate/ Pore size = 0.2 µm Area = 0.116 m <sup>2</sup>	Synthetic sewage	Flux = 15 L/m <sup>2</sup> ·h, HRT = 12, 8, 6, 4 and 2 h, SRT = 200 day, Temp = 35 ± 1 °C, MLSS = 6000 mg/L and 7000 mg/L, OLR = -	V = 3 L S = L	$544 \pm 22$	$14\pm2$	>97	$\begin{array}{c} 252\pm 27,\\ 236\pm 27,\\ 249\pm 29,\\ 264\pm 46,\\ 134\pm 23 \end{array}$	[75]
SMBR/-/-	MF/Polymethyl methacrylate/ Pore size = 0.04 μm Area = 0.047 m <sup>2</sup>	Synthetic wastewater	Flux = 10.64L/m <sup>2</sup> ·h, HRT = 8 h, SRT = 140 day, Temp = 18 $\pm$ 15 °C, MLSS = 6000 mg/L, OLR = 1.77 $\pm$ 0.03 g COD/L·day	V = 4 L S = L	-	-	91 and 98	-	[76]
AnOMBR/submerged/-	Cathode/Stainless steel mesh/ Pore size = - Area 1.5 m <sup>2</sup>	Synthetic wastewater	Flux = 3.532 LMH HRT = -, SRT = - Temp = 35 ± 1 °C MLSS = 16.31 g/L OLR = -	V = 6.8 L S = P	2000	-	71.1	0.254	[77]
SAMBRs/Submerged/Flat sheet	MF/Non-woven fibrous (chlorinated polyethylene)/ Pore size = 0.2 µm Area = 0.116 m <sup>2</sup>	Synthetic sewage	$\label{eq:Flux} \begin{array}{l} Flux = 5 \ L/m^2 \cdot h, \\ HRT = 12 - 6 \ h, \\ SRT = , Temp = 35 \ ^{\circ}C, \\ MLSS = -, OLR = 3 - 6 \ kg \ COD/m^3 \cdot day \end{array}$	V = 3.2 L S = L	-	$29\pm8$	93.2 ± 6.6	$1.7 \pm 0.3$	[78]
Integrated anaerobic fluidized bed membrane (IAFMBR)/-/-	-/Hollow fiber/ Pore size = $0.4 \ \mu m$ Area = $0.21 \ m^2$	Synthetic benzothiazole wastewater	Flux = 11.3 L/m <sup>2</sup> ·h, HRT = 24 h, Temp = 35 °C, MLSS = -, OLR = -	V = 6.1 L S = L	-	230	96	$\begin{array}{c} 0.31 \pm 0.02 \text{,} \\ 0.32 \pm 0.03 \text{ and} \\ 0.31 \pm 0.01 \end{array}$	[79]
SAMBRs/Submerged/Flat sheet	MF/Chlorinated polyethylene/ Pore size = 0.2 μm Area = 0.116 m <sup>2</sup>	Synthetic sewage	Flux = -, HRT = 48, 24, 12, 6 h, Temp = 25, 15, 10 °C, MLSS = -, OLR = -	V = 6 L S = L	-	134	94	-	[80]
AnMBRs/External/-	-/Polyvinylidence (PVDF) hollow fiber/ Pore size = 0.22 μm Area = 0.06 m <sup>2</sup>	Synthetic sewage	$ \begin{array}{l} Flux = 20.2 \pm 8.5 \ L/m^2 \cdot h, HRT = 3.5 \pm 1.1, \\ 2.2 \pm 0.5, 2.3 \pm 0.3 \ day, Temp = 35 \ ^\circ C, \\ MLSS = 4.78 \pm 1.9 \ g \ L^{-1}, \\ OLR = 3.4 \ kg \ COD/m^3 \cdot day \end{array} $	V = 3.5 L S = L	$6752\pm 663$	$\begin{array}{c} 0.095 \pm 0.15 \\ 0.14 \pm 0.23 \\ 0.28 \pm 0.35 \end{array}$	96.7 ± 2.7	$0.50 \pm 0.17$	[81]
EG-AnMBR and SG-AnMBR	-/Polyvinylidence (PVDF) hollow fiber/ Pore size = 0.22 μm Area = 0.06 m <sup>2</sup>	Synthetic sewage	Flux = 7 L/m <sup>2</sup> ·h, HRT = 12 h, Temp = 20 °C, MLSS = 22.34 $\pm$ 0.41 g/L, OLR = 0.53-0.59 kg COD/m <sup>3</sup>	V = 4 L S = L	-	-	<90%	160	[82]

# Table 4. References on anaerobic membrane bioreactors for synthetic wastewater treatment.

AnMBR = anaerobic membrane bioreactor, COD = chemical oxygen demand, HRT = hydraulic retention time, MLSS = Mixed liquor suspended solids, OLR = organic loading rate, P = pilot scale, - = not reported.

#### 4. Effect of Microbial Activity on Anaerobic Membrane Performance

During microbial activity, the concentration of volatile fatty acids (VFA) usually reflects the state of anaerobic digestion performance, specifically in the acetogenic and methanogenic phase. According to Wijekoon et al. [70], 85–96% of high strength molasses-based synthetic wastewater was removed as total chemical oxygen demand (COD) at optimum organic loading rate of  $8 \pm 0.3$  kg COD/m<sup>3</sup>·day. Though it was at higher temperature, but biogas production of 15, 20 and 35 L/day at OLR 5.1  $\pm$  0.1, 8.1  $\pm$  0.3 and 12.0  $\pm$  0.2 kg COD/m<sup>3</sup>·day respectively was achieved. It was seen from the results of the treatment, increasing loading rate amounts to increase in hydrolytic and methanogenic activities. However, it was also observed that the process performance reached its maximum level with continuing increase in loading rate which subsequently reduce the biological activities of the system.

In anaerobic digestion, rapid methane formation is mostly attributed to the presence of acetic acid and butyric acid. However, acetic acid contributes 70% of the total acidic portion. This might be due to the fact that, all the volatile acids are converted into acetate during metabolism. The presence of propionic acid tends to upset anaerobic digestion processes due to its toxicity among the VFAs, but this effect could be counteracted under thermophilic conditions. The experiments of Speece et al. [71] showed that propionic acid degradation rate to other intermediate was very poor, largely because of lower partial pressure (H<sub>2</sub>) demand, but it also supports process start up and stabilization under strict anaerobic conditions.

Moreover, methanogenic activity might be inhibited with propionic acid concentrations greater than 1–2 g/L. It could also withstand acetic and butyric acid concentrations up to 10 g/L. Ahmed et al. [72] have found that, microbial respiratory quinones are essential parts of the bacterial respiratory chain that perform a vital role in electron transfer during the period of microbial respiration. However, an investigation of Hiraishi et al. [73] demonstrated that alterations in microbial community structure in a mixed culture of microbes could effectively be quantified using quinone profiles.

Recent research on the effects of temperature and temperature shock on the performance and microbial community structure of a submerged anaerobic membrane bioreactor (SAnMBR) was demonstrated by Gao et al. [21]. The result obtained indicates that not only the diversity, but also the species richness of microbial populations are affected by temperature variation. It further proves that submerged AnMBR performance under temperature shock conditions have no effect on the COD removal ability of the reactor. The biogas production rates were  $0.21 \pm 0.03$ ,  $0.20 \pm 0.03$  and  $0.21 \pm 0.02$  L/g COD removed. Moreover, no major change is observed in the production and composition of biogas. Nevertheless, temporary production of biogas occurred at temperature shock which affected the abundance and diversity of microbial populations.

According to Iranpour et al. [83], the similarities that exist among the microbial community working under variable temperatures may be connected to the development of thermomesophiles rather than thermophiles. During microbial activity in anaerobic digestion, the release of soluble organic compounds called SMP in normal biomass metabolism is of great interest, not only in terms of achieving discharge standard limits, but also in setting the lower limit for treatments [84]. The significance of SMP in all kinds of wastewater treatment is at the moment objectively well recognized, but complications still come about in trying to measure SMP and draw conclusions when they are present in effluents from plants treating highly complex feeds. However, researchers like Sciener et al. [85] and Chidoba et al. [86] have studied this and conclude that large portions of the soluble organic matter in the effluent from the biological treatment processes are actually SMP.

The work of Judd [87] and Li et al. [88] reveals that microbial activity decreases under prolonged SRT in AnMBR. This is because microbes take a long time to degrade the inorganic matter and require a high concentration of biomass to ensure all the organics are totally degraded. Still, the application of high SRT is advantageous, since it favors biomass growth which is responsible for biodegradability of organic pollutants. It also provides room for higher MLSS in AnMBR that establish starvation conditions to achieve good quality effluent and create a low F/M ratio [89,90].

#### 5. Operational and Performance Parameters

#### 5.1. Temperature

In anaerobic digestion (AD) processes, temperature is a major factor that plays an important role in the stabilization and performance of the whole system [91,92]. Under strict anaerobic conditions, some bacteria thrive well under psychrophilic (<25 °C), mesophilic (25–40 °C) and thermophilic (>45 °C) conditions [93]. According to El-Mashad et al. [92] and Duran et al. [94], thermophilic conditions offer more advantage in terms of specific growth and metabolic rates with comparatively less ammonia inhibition than mesophiles. However, it could also cause higher microbial death rates, poor supernatant quality and reduced process stability due to chronically high propionate concentrations than mesophilic bacteria. Studies on biogas production and wastewater treatment at varying temperature such as psychrophilic (0–20 °C), mesophilic (20–42 °C), and Thermophilic (42–75 °C) were well investigated in [10,12,14,94].

The diversity and activities of microbial communities coupled with thermodynamic equilibrium of the biochemical reactions are adversely affected by temperature [95]. It usually shifts the abundance and activities of specific microbial populations and determines the roles of specific taxa in the AD food chain. It all begins with substrate hydrolysis, followed by acidogenesis, then acetogenesis, and ends with methanogenesis [96]. Study on temperature adaptation was examined by Chidoba, 1985 [86]. It was initially set at mesophilic condition (37 °C) and gradually switched to thermophilic (55 °C). A drastic reduction in biogas production by 15% from the original state of mesophilic was observed. This was attributed to the change in temperature along with increase in VFA to 3000 mg/L. More detailed studies were reported by Song et al. [97] on thermophilic and mesophilic temperature co-phase anaerobic digestions. Their examination exclusively focused on the sewage sludge using the exchange process of digesting sludge between spatially separated mesophilic and thermophilic digesters and compared with single-stage mesophilic and thermophilic anaerobic digestions. It was confirmed that the system stability, effluent quality, specific methane production during single-stage operation was greater in mesophilic than thermophilic., but volatile solids (VS) reduction and total coliform destruction were much higher in single-stage thermophilic than mesophilic digestion.

In the past, study on the effect of temperature range between 40–64  $^{\circ}$ C using cow manure as the main substrate was well studied by Angelidaki, 1994 [98]. Two different ammonia concentrations (2.5 and 6.0 g N/L) were continuously fed to the lab-scale reactor. It was observed at some stage, precisely (HRT 15 days); the high temperature and ammonia loading resulted in poor process performance. Consequently, a significant change in the amount of biogas production and process stability was seen when the ammonia load was high and temperature reduces to below 55  $^{\circ}$ C.

### 5.2. pH

Volatile fatty acid (VFA) concentration usually determines the pH of the effluent, which is one of the influential factor in anaerobic digestion (AD) processes [99]. Different range of pH is required for bacterial growth in AD, comprehensively between 4.0 to 8.5 [100]. However, a pH level of 6.8 to 7.2 suitably favors methanogenic bacteria [101,102]. On the other hand, hydrolysis and acidogenesis thrive well in the pH range between 5.5 and 6.5 [103,104]. It was also shown that, excessively alkaline pH may result in microbial granules disintegration and subsequent failure [105]. The pH adjustment studies in [101] favorably increased methane yield. The maximum cumulative biogas production reached 16,607 mL at pH 7.0 (0.4535 L methane/g VS). However, the yield decreased to 6916 and 9739 mL at pH 6.0 and 8.0, equivalent to 0.1889 L methane/g VS and 0.2659 L methane/g VS, respectively.

Recent research on novel biogas-pH automation control strategy using a combined gas-liquor phase monitoring was developed by Yu et al. [106] using an AnMBR treating high starch wastewater COD (27.53 g/L). The biogas pH progressed with threshold between biogas production rate >98 NmL·h<sup>-1</sup> preventing overload and pH > 7.4 preventing under load. The OLR and the effluent COD

was doubled to 11.81 kg COD/m<sup>3</sup>·day and halved as 253.4 mg/L, respectively. In another development, a ternary contour was performed by Mao et al. [107] to picture the pH dissimilarities in ternary buffer system. The variation was controlled by a system composing of VFAs, ammonia and carbonate. However, the accurate simulation of pH variation in AnMBR during methanogenesis is extremely challenging due to enormous magnitude of electrolytes, and as such the ternary macro-quantity buffer salts were chosen for the visualization which denotes the critical pH buffer capacity to during methanogenic stage.

Kim et al. [104] demonstrated that one major problem faced with COD removal especially for starch wastewater is the provision of sufficient carbonate alkalinity. The effect of high pH shocks (pH 8.0, 9.1 and 10.0) on the performance of submerged AnMBR was well reported in [105]. It was found that; pH 8.0 had slight influence, while pH 9.1 and 10.0 shocks have put forth vigorous impact on biogas production, COD removal, and membrane filtration performance. In addition, colloids and solutes accumulation in the sludge suspension further accelerates deterioration of membrane performance. Interestingly, when neutral pH (7) was taken up again, it cost the reactor approximately 1, 6, and 30 days to recover from the previous shocks.

An experiment on the effect of mixed liquor (pH 5 and 9) in the removal of trace organics in both acidic and basic environment was also reported in [106]. A reduction in total organic carbon TOC and total nitrogen TN removal efficiencies was detected with ionisable trace organic contaminants. Hence, the removal efficiencies were extremely pH dependent. Conversely, the biological performance was favorable at near optimum pH (i.e., approximately pH 6–7) with respect to TOC and TN removal efficiencies. As result of the differences in attainment of optimum pH by acidogenic bacteria (5.5–6.5) and methanogens (6.5–8.2), several studies have been conducted on phase separation of acidogenesis and methanogenesis using AnMBRs. For example, Mao et al. [107] demonstrated a two-stage reactor configuration and optimizes each one phase as an entity. VFA accumulation was drastically reduced and the system stability improves to the extent of forbearing greater loading rate and toxicity. Therefore, maintenance and adherence to these features would positively escalate the chance of achieving a high methane yield.

#### 5.3. Effect of OLR

The amount of volatile solids VS fed to a bioreactor at an interval of time under continuous operation is termed as the OLR. Under normal operating conditions, it is expected that an increase in organic loading rate would also increase the quantity of biogas. AnMBR processes have the advantage of tolerating changes in organic loading similar to tolerance to fluctuations in temperature. Organic loadings ranging from 0.5 to  $12.5 \text{ kg/m}^3$  day was applied to AnMBR for the treatment of domestic wastewater. The system achieved 97% (COD) removal with less than 20 mg/L effluent COD [108]. In a similar experiment by Vincent et al. [109], less than 50 mg/L soluble COD in the effluent was obtained at organic loading rate 0.25 kg/m<sup>3</sup>·day and 0.7 kg/m<sup>3</sup>·day using AnMBR. This is not the case in the work of Qiao et al. [110]. The system exhibited very poor effluent quality after a long HRT. Study of the performance of AnMBR by Yingyu et al. [111] showed that biogas yield rose linearly with increasing organic loading. A similar trend was observed by Wijekoon et al. [70] using a two-stage thermophilic AnMBR with continuous increasing loading rate from 5 to 12 kg COD/m<sup>3</sup>·day. Bornare et al. [112] achieved biogas production increased from 159 to 289 L/day, but they observed a decreased in yield from 0.48 to 0.42 L biogas/g CODremoved when OLR was increased from 0.62 to 1.32 kg COD/m<sup>3</sup>·day. However, Dereli et al. [113] indicated that OLR could not be an independent parameter and therefore should be assessed along with SRT.

High performance integrated anaerobic–aerobic fixed-film pilot-scale reactor with an arranged media for treating slaughter house wastewater showed a removal efficiency of 93% at OLR of 0.77 kg COD with methane yield of 0.38 m<sup>3</sup> CH<sub>4</sub>/kg COD [114]. High mixing as a result of system integration causes low extension of the anaerobic process and consequently affects the methanogenic activities. A fact known to most researchers in the field of anaerobic digestion is that an increase in OLR

could result in excessive VFA formation which may inhibit microbial activities and deteriorate the system. For example, the research of Saddoud et al. [115] confirms that accumulation of VFA was the main reason for methanogenic inhibition, thus, resulting in a decrease in methane yield at OLR of 16.3 kg COD/m<sup>3</sup> in one-phase AnMBR. To counteract this effect, they proposed the use of a two-stage AnMBR coupled with an anaerobic filter as acidogenic reactor while a jet flow AnMBR was used as methanogenic reactor at high OLR and realized a substantial improvement in biogas production in the subsequent stage. Jeison et al. [116] presented removal efficiency below 50% using continuous stirrer tank reactor CSTR fed with acidified and partially acidified substrate. Even though the organic loading rates reached 10–17 kg COD/m<sup>3</sup>·day, the low removal efficiency might be connected to soluble microbial product level within the AnMBR and these could cause irreversible pore fouling in the membranes.

## 5.4. Effect of HRT and SRT

Operational parameters such as hydraulic retention time (HRT) and solid retention time (SRT) are two factors that play a vital role in the treatment performance of AnMBRs. A study by Mei et al. [61] demonstrated using submerged anaerobic membrane bioreactors (SAnMBRs). SRTs of 30, 60 and infinite and HRT of 12, 10 and 8 h were used. A total COD removal efficiency >97% was observed at all operating conditions. Biogas production rate reached 0.056 L CH4/g MLVSS day at an infinite SRT. However membrane fouling occurred as a result of shorter HRT and infinite SRT. It was also seen that, longer SRT was the main cause of higher SMP production. The presence of SMP subsequently introduces more nutrients onto the membrane surface that caused the blockage of the pores and enhanced biocake formation. Dong et al. [117] reported the influence of SRT and HRT on bioprocess performance of pilot and bench scale AnMBRs using municipal wastewater as substrate. The SRT and HRT applied were 40–10 day and 2.5 to 8.5 h. A good permeate quality with COD concentration (40 mg/L) and BOD<sub>5</sub> (10 mg/L) was observed in all conditions. The range of values tested for SRT and HRT have not considerably interfered with COD and BOD<sub>5</sub> removal efficiencies. Moreover prolonged SRTs caused a reduced sludge production and increase methane yield.

Similarly, in the research of Ozgun et al. [65], reduction in permeate COD concentrations from 16.5 to 5 mg/L resulted in an increased methane yield from 0.12 to 0.25 L CH<sub>4</sub>/g COD. The improvement in the quality of effluent and the methane was attributed to prolonged SRT from 30 days to infinite. Salazar et al. [39] and Hu et al. [28] revealed a permeate COD concentration increase from 10 to 50 mg/L with decreasing HRT. However, Liao et al. [35] strongly suggested that, lower HRTs give room for shorter contact time between microorganisms and substrate and therefore might pave way for a part of influent COD leaving the reactor without proper treatment. One very important aspect of AnMBRs is the enabling environment that allows SRT to be completely independent from HRT irrespective of the sludge properties. Conversely, experience and frequent practice shows that longer SRTs operation yield more quantities of biogas. This is because any reduction in the SRT may decrease the extent of reactions required for stable digestion. For instance Huang et al. [66] clearly reported a methane yield of (0.670  $\pm$  0.203 L CH<sub>4</sub>/day), (0.906  $\pm$  0.357 L CH<sub>4</sub>/day), (1.290  $\pm$  0.267 L  $CH_4/day$ ) at longer SRT of 30, 60, and infinite days respectively. Therefore, it is apparent that, longer SRT in AnMBRs operations give room for minimal sludge production, and hence cuts disposal cost significantly. Based on the studies on the effects of HRT and SRT conducted so far, it could be seen that prolonging HRT may result in inadequate utilization of AnMBRs' volume and its reduction may lead to rapid VFA accumulation which may hinder methanogenic activities. Prolonged SRT might also result to membrane fouling. It could also encourage the release of soluble microbial products (SMP) as well as rapid cake formation and excessive decline in flux.

## 6. Inhibitors

Inhibitory substances are the primary cause of disparity in anaerobic reactors. This might also lead to entire failure of the digestion processes when the concentration is beyond tolerable limits. Anaerobic

digestion usually presents different variations in the level of inhibition and toxicity. Mechanisms such as synergism, acclimation, and complexity substrate could considerably upset the phenomenon of inhibition. Nevertheless, bioreactor failures are regularly reported as the result of high ammonia inhibition which directly affects the microbial activity [118].

Kayhanian et al. [119], reported ammonia as one of the inhibitory substances found in anaerobic digestion. It is mainly as a result of biological degradation of nitrogenous matter in the form of proteins and urea. Inorganic ammonia nitrogen like ammonium ion  $NH_4^+$  and free ammonia (FA) are usually found in aqueous form. FA is considered as the primary cause of inhibition since it is freely membrane-permeable [120,121]. Calli et al. [122] reveal an apparent COD removal of 78–96% in a study of the effect of high free ammonia concentrations in synthetic wastewater using UASB reactor. The reactor was fed at OLR of 1.2 kg COD m<sup>3</sup>/day with total ammonia nitrogen concentration total ammonia nitrogen (TAN) increasing from 1000 to 6000 mg/L. However, Tchobanoglous et al. [123] showed that, the accumulation of propionic acid along with reduction in eubacterial suggest a sensitivity of propionate degrading acetogenic bacteria to free ammonia than methanogenic archaea. A theoretical stoichiometric relationship of estimating the quantity of ammonia that could be generated from organic substrate in anaerobic biodegradation is shown in Equation (1):

$$CaHbOcNd + ((4a - b - 2c + 3d)/4)H_2O \rightarrow ((4a + b - 2c - 3d)/8)CH_4 + ((4a - b + 2c + 3d)/8)CO_2 + dNH_3$$
(1)

According to Kayhanian [124], methanogens are the most sensitive to ammonia inhibition among the four bacteria types that exist in anaerobic digestion processes. The effect might even cause the bacteria to cease to grow. Study on the effects of ammonia on propionate degradation and microbial community in bioreactors showed that, using propionate as a sole carbon source result to reactor failure after four hydraulic retention times [125]. A total ammonia nitrogen (TAN) concentration of 2.5 g  $NL^{-1}$ at OLR of 0.8 g propionic acid (HPr)/L·day were observed and 95% of the degraded HPr was converted to methane. On the average, the degradation rate of HPr is below 53%, likewise an average of 74% and 99% HPr degradation and methane recovery rates was also recorded during the last HRT. Thus, these behaviors demonstrate an alteration of the microbial community. Frequent maintenance, change in intracellular pH of methanogens and inhibition of a specific enzyme reactions are some of the numerous suggested pathways for overcoming ammonia inhibition. Thorough understanding of the ammonia toxicity occurrence which ammonia may affect methanogenic bacteria is not readily available but, is the few available studies with unadulterated cultures this was revealed to influence the treatment in two ways: (i) direct inhibition of methane- producing enzymes by ammonium ion and/or (ii) the hydrophobic nature of ammonia molecules which may diffuse passively into bacterial cells causing proton imbalance [126].

Another important anaerobic digestion inhibiting parameter is sulfate. Sulfate is a common constituent of many industrial wastewaters that is converted in to sulfide by the sulfate-reducing bacteria (SRB) [127]. Major SRB includes complete and incomplete oxidizers. Complete oxidizers convert acetate to  $CO_2$  and  $HCO_3^-$  completely. Compounds such as lactate are reduced to acetate and  $CO_2$  by incomplete oxidizers. Furthermore, primary and secondary inhibitions are the two stages that exist during sulfate reduction [128]. Methane production is suppressed by primary inhibitors as a result of competition for common organic and inorganic substrate by the SBR, but secondary inhibition is caused by various bacterial groups due to the toxicity of sulfide [129,130].

#### 7. Membrane Fouling

Membrane fouling is the result of gradual accumulation of suspended solids (SS) and dissolved solids (DS) mostly in the form of fats, oil and grease onto the surface of the membrane [131]. Mechanisms that propel fouling occur via: (1) deposition of sludge flocs onto the surface, (2) adsorption of solutes within the membrane, (3) surface cake layer formation, and (4) the shear force that exists between membrane surface, soluble microbial products (SMPs) and extracellular polymeric substances

(EPS) [132]. The surface blockage or clogging phenomenon is always attributed to the type of membrane itself, biomass and operating conditions. Fouling due to the membrane itself could be attributed to the nature of the material, configuration, hydrophobicity, porosity and pore size of the membrane [8]. Moreover, biomass characteristic such as MLSS, EPS or SMP, flock structure and dissolved matter contribute immensely to causing fouling [4]. Conditions at which membrane bioreactor operates might also increase fouling if not properly monitored. Other factors like cross flow velocity, HRT or SRT, aeration and transmembrane pressure could also contribute significantly to fouling [133].

Zhang et al. [134] further revealed that, the use of adsorbent/flocculants in (AnMBR) could rarely overcome the effect of fouling completely. However, their research revealed that, among the combination of eight additives (three powdered activated carbon PACs, two granular activated carbons, one cationic polymer, and two metal salts), 400 mg/L of PAC was able to reduce transmembrane pressure rise from 0.94 to 0.06 kPa/h. This outcome signified an outstanding fouling reduction technique. Still, the effect of increasing in OLR and reduction of HRT (24-18 h) is clearly confirmed as the main factor responsible for severe membrane fouling, but research by Jeison et al. [67] showed that, extracellular polymeric compounds (EPS) from microbial cells was responsible for the release of SMP and a drastic increase in TMP during the long term operation that led to the occurrence of fouling. Filtration performance of membrane bioreactors is largely dependent on the mixed liquor suspended solids. In the experiments performed by Meng et al. [135], filtration resistance including membrane resistance (12%), cake resistance (80%), blocking and irremovable fouling resistance (8%), depicts that the formation of cake layer is the main cause of membrane fouling. Lee et al. [136] pointed out that cake layers are not uniformly distributed on the entire surface of all of the membrane fibers. It could be wholly covered by a static sludge cake that could not be removed by the sheer force due to aeration, and partially by a thin sludge film that might frequently washed away due to aeration turbulence. A study of Jeison [67] concluded that cake layer formation could be removed despite longer operation period (>200 days). However, during these periods, cake consolidation was seen close to the critical flux. A normal backwashing cycle was unable to remove the consolidated cake and as such physical external treatment was applied. Similarly, Cho et al. [131] and Di-Bella et al. [137] showed that cake layer formations on AnMBRs coupled with external aeration zone could easily be removed. This study suggest that AnMBRs operating with aerobic zones external to the membrane module could achieve higher cake layer removal as compared to cake layer formed on membrane embedded within an anaerobic bioreactor.

## 8. Conclusions

They has been a quite significant development of technologies towards meeting the stringent environmental regulatory discharge requirements and biogas production. These can be seen in the manner, in which MBRs are configured, variations of operational parameters, and the different techniques adopted towards provision of the basic living conditions for microorganisms to strive. However, membrane lifespan is still the main concern of stakeholders in the water and wastewater treatment industries. The efficiency of physical, chemical and biological methods of reversing fouling on membrane surface is being exploited. Though physical and chemical methods have been sufficient, the disadvantages are huge. A lot of energy is consumed during aeration and considerable amount of chemicals are utilized which does not favor the players in this field with regards to cost and environmentally-wise. Research is still ongoing on the most economical biological methods of producing quality effluent and biogas production using AnMBRs, and future studies should take in to account the following factors:

 Food to microorganism F/M ratio interaction and HRT of bioreactors operating at thermophilic, mesophilic or psychrophilic temperature. These will offer more information on biogas production and effluent quality.

- 2. Provision of lining or rough surfaces within the bioreactor (biofilm) would facilitate the retention of large microbial populations preventing them from exiting the reactor along with the effluent if shorter HRT is to be used.
- 3. If the purpose of the treatment is to meet discharge standard limits, and subsequently discharge the effluent to water bodies, then the use of microfiltration should be encouraged rather than ultrafiltration. This is because; smaller size membrane surface area would require more energy for water to pass through and as such applying ultrafiltration for such treatment would be costly.

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