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# 1 Anaerobic membrane bioreactors for wastewater treatment: Novel configurations, fouling 2 control and energy considerations

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## 18 Abstract

19 Water shortage, public health and environmental protection are key motives to treat wastewater. The  
20 widespread adoption of wastewater as a resource depends upon development of a technology.  
21 Anaerobic membrane bioreactor (AnMBR) technology has gained increasing popularity due to their  
22 ability to offset the disadvantages of conventional treatment technologies. However there are several  
23 hurdles, yet to climb over, for wider spread and scale up of the technology. This paper reviews  
24 fundamental aspects of anaerobic digestion of wastewater, and identifies the challenges and  
25 opportunities to the further development of AnMBRs. Membrane fouling and its implications are  
26 discussed, and strategies to control membrane fouling are proposed. Novel AnMBR configurations are  
27 discussed as an integrated approach to overcome technology limitations. Energy demand and recovery  
28 in AnMBRs is analyzed. Finally key issues that require urgent attention to facilitate global penetration  
29 of AnMBR technology are highlighted.

30 **Keywords:** Anaerobic membrane bioreactor, novel configurations, membrane fouling, energy  
31 recovery, wastewater treatment

## 1 **1. Introduction**

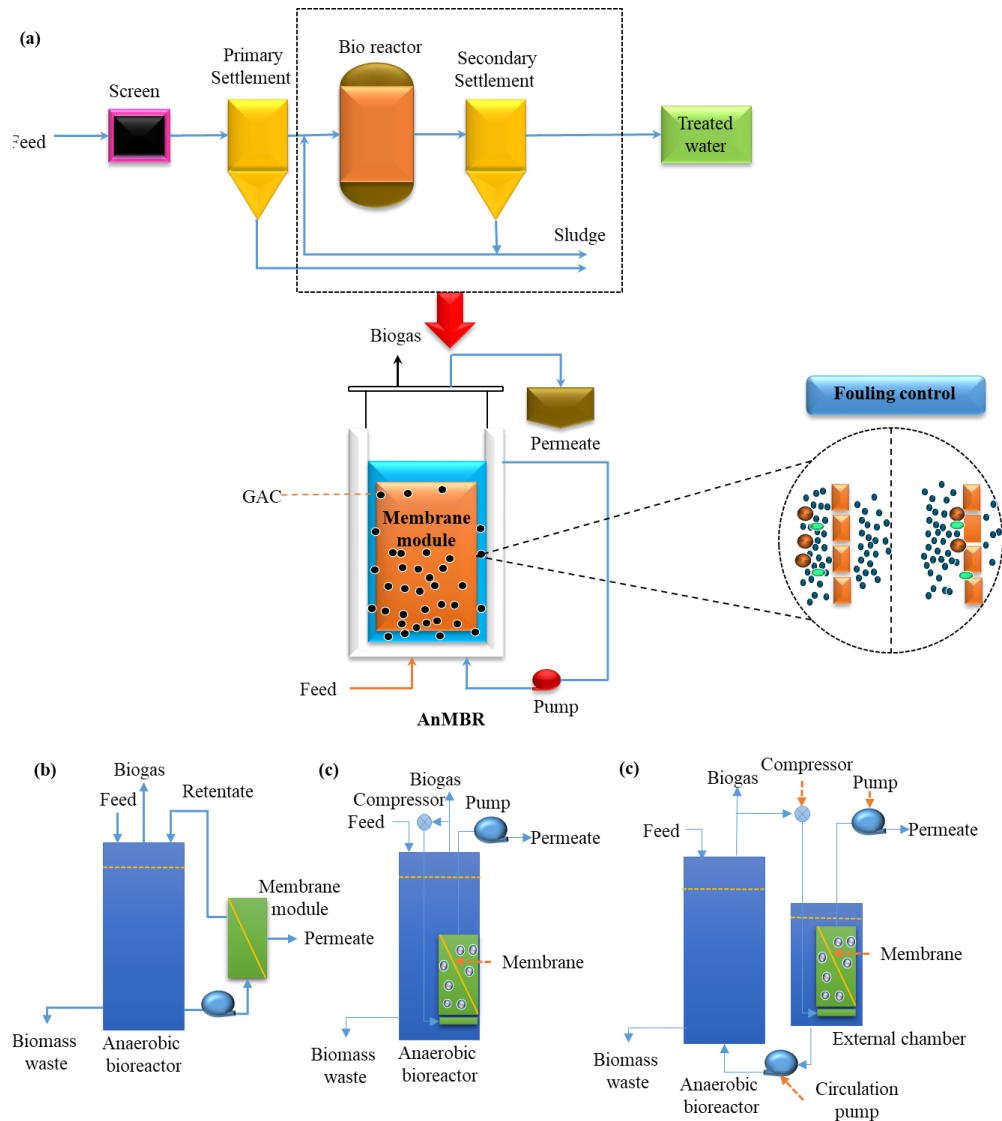
2           The scarcity of water resources and environmental consequences caused by the addition of  
3 wastewaters into main water bodies and soil has triggered the need of treating wastewater. Wastewater  
4 is considered as additional sources of water for domestic, agricultural and industrial use (McCarty et  
5 al., 2011; Smith et al., 2014). Since agricultural and industrial development relies on the availability of  
6 water, world is looking for additional water resources for which wastewater is readily available and  
7 abundant resource. Currently, wastewater serves as a source of clean water and a valuable resource for  
8 renewable energy (biogas) and nutrients (fertilizers) (McCarty et al., 2011). The practicality of using  
9 wastewater as a resource lies in the development of an efficient, economical and environmental  
10 friendly treatment technology. Aerobic wastewater treatment technologies have been in use since a  
11 century. However, these methods require high energy which offset the advantages of reusing  
12 wastewater (Krzeminski et al., 2017). Other disadvantages include production of high amount of  
13 sludge, need of larger space for plant operation, and higher maintenance cost. During the process, there  
14 is uncontrolled release of methane (CH<sub>4</sub>), carbon dioxide (CO<sub>2</sub>), and nitrogen oxide (N<sub>2</sub>O) which are  
15 potent greenhouse gases (Foresti et al., 2006; Ghauri et al., 2011). Hence, there is an immense need of  
16 developing alternate wastewater treatment technology. Recently, membrane separation processes  
17 gained significant attention to recover resources from waste streams, biofuels and especially  
18 wastewater to reduce potent greenhouse gases (Khalid et al., 2019; McCarty et al., 2011).

19           Anaerobic membrane bioreactors (AnMBR) are emerged as a promising alternative to aerobic  
20 wastewater treatment technologies. AnMBR have low energy input, and sludge production compared  
21 with aerobic processes. Furthermore, employing AnMBRs reduce: the operational space, and number  
22 of unit operations compared with conventional processes (as shown in Fig. 1). The also offer easy  
23 scale-up and selective separation and recovery of nutrients and resources (Aslam et al., 2018b; Liao et  
24 al., 2006). Technology is well established and some plants treating municipal and industrial wastewater  
25 are operational. However, widespread adaption of technology is still hindered due to several issues

1 including membrane fouling, dissolved CH<sub>4</sub> recovery, sulphide-induced low COD, and alkalinity.  
2 Among these, membrane fouling is a key bottleneck for their commercialization (Lin et al., 2013).

3 Membrane fouling gradually decrease the permeate flux and hence the life and efficiency of  
4 treatment process. Various strategies have been adapted to overcome membrane fouling. These include  
5 pretreatment of influent wastewater feed, improvement of membrane characteristics, and bioreactor  
6 operation under optimum conditions (Shoener et al., 2016). Novel AnMBR configurations are  
7 introduced to overcome the issues in scale-up of technology in general, and combat membrane fouling  
8 in particular. These reactor schemes have also reduced the energy input and increase the CH<sub>4</sub>  
9 production yield (Shin & Bae, 2018; Smith et al., 2014). To date, several review papers have been  
10 published on membrane fouling and biogas production in AnMBRs (Lin et al., 2013; Ozgun et al.,  
11 2013; Shin & Bae, 2018; Skouteris et al., 2012; Smith et al., 2012; Stuckey, 2012). The potentials of  
12 novel AnMBR configurations were not enlightened and the research works related with AnMBRs were  
13 only simply or partially articulated in the previous reviews. Moreover, there are several hurdles, yet to  
14 climb over, for wider spread, optimization of design configuration and economic feasibility, potential  
15 applications and scale up of the AnMBR technology. Hence, a critical review on recent technical  
16 innovations and emerging configurations to improve energy production potential to achieve energy  
17 neutrality or positivity is of crucial importance particularly in AnMBRs.

18 The review covers fundamental aspects of anaerobic digestion of wastewater and novel  
19 configurations, fouling control and energy considerations in AnMBRs. This study identifies the major  
20 challenges and opportunities in wastewater treatment using AnMBRs. Novel AnMBR configurations  
21 to combat membrane fouling are presented. Energy demand and recovery in AnMBRs is analyzed.  
22 Finally, key issues that require urgent attention to facilitate global adaption of AnMBR technology are  
23 highlighted.



1

2 **Fig. 1.** (a) Anaerobic membrane bioreactor technology in replacement of conventional activated sludge process; (b)  
 3 Side-stream AnMBR; (c) Submerged AnMBR; (d) External submerged AnMBR.

4 **2. Fundamentals of anaerobic membrane bioreactors**

5 *2.1. Technical overview*

6         AnMBR technology is a promising to treat wastewater and recovery of resources such as  
 7 bioenergy in the form of renewable methane, fertilizing elements it contains and water for reuse to  
 8 control the pollution in environment and energy sustainability. (Charfi et al., 2017b; Smith et al.,  
 9 2012). It is a hybrid system in which membranes are combined with anaerobic bioreactor. There are  
 10 different pathways selected for implementation of membrane in bioreactor or outside bioreactor.

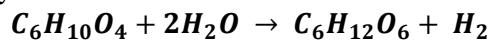
1 Membrane module is present outside reactor and permeate obtains under pressure provided by  
2 recirculation pump in external cross flow configuration (Fig. 1b). In this approach, filtration cake on  
3 membrane surface is disturbed by cross flow velocity (Skouteris et al., 2012). The drawback of this  
4 scheme comes in form of energy consumption by recirculation pump to maintain transmembrane  
5 pressure and elevated volumetric flow to keep cross-flow velocity at certain level. However,  
6 membrane cleaning is easy because membrane can easily be taken out for cleaning purposes  
7 (Szentgyörgyi & Bélafi-Bakó, 2010). An external membrane module system is easy to monitor and  
8 maintain. Moreover, this technique provides sufficient turbulence to increase back transport of foulants  
9 from membrane surface. However, more energy is required in this configuration with high hydraulic  
10 shear force which might damage the favorable anaerobic solid microbes into small sizes and causes  
11 membrane fouling. Operating TMP and cross flow velocity through membrane in external membrane  
12 module is relatively high. The range of cross flow velocity and TMP is 1 to 5 m/s and 207 to 690 kPa,  
13 respectively (Berube et al., 2006). In contrast, with submerged AnMBR, membrane module is directly  
14 immersed in bioreactor and treated water obtains under vacuum pump or gravity on permeate side of  
15 membrane and retained biomass presents in bioreactor. (Fig. 1c). In this approach, biogas is used to  
16 mitigate cake formation (Shin et al., 2016b). Submerged membrane bioreactor is more favorable for  
17 low strength organic loads like municipal wastewater (Musa et al., 2018). In this system cross flow  
18 velocity and TMP are relatively low about less than 0.6 m/s and 21 to 103 kPa, respectively. Gas is  
19 entered into the system at base of membranes. The gas bubbles carry liquid upward and causes cross  
20 flow along membrane (Berube et al., 2006). In external submerged configuration, membrane is present  
21 in separate tank outside bioreactor and needs pump to return retentate to bioreactor as shown in Fig. 1d  
22 (Liao et al., 2006). Membrane operates under vacuum in this configuration. In all these techniques,  
23 difference is in direction of flow that reverts and as result different transmembrane pressure is obtained  
24 (Singhania et al., 2012).

## 1 2.2. Chain of processes in anaerobic membrane bioreactor

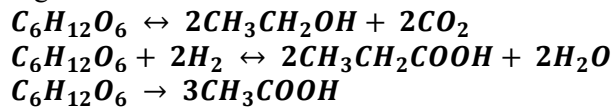
### 2 2.2.1. Hydrolysis

3 Anaerobic treatment of the wastewaters involves a logical sequence of processes starting with  
4 hydrolysis as shown in Fig. 2. During this process, small water soluble compounds are attained by the  
5 breakage of complex organic matter. The complex molecules contain carbohydrates, proteins and fats,  
6 which are converted into monosaccharaides, amino acids, and fatty acids by facultative and/or obligate  
7 anaerobic hydrolytic bacteria. During enzymatic hydrolysis extracellular enzymes are released to  
8 provide covalent bonds within substrate in chemical reaction with water (Lei et al., 2018). The  
9 hydrolytic bacterial species are unique and specifically degrade particular type of macromolecule. The  
10 hydrolysis rate depends on the pH, temperature, concentration of hydrolyzing mass and the size and  
11 type of particulate organic matter (Visvanathan & Abeynayaka, 2012). Stepwise chemical reactions  
12 involved till to methane production are (Bajpai, 2017; Zupančič & Grilc, 2012):

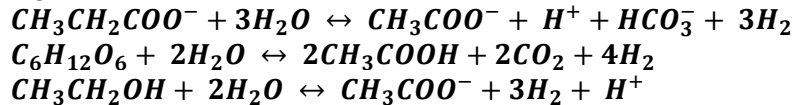
Hydrolysis



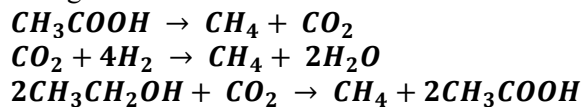
Acidogenesis



Acetogenesis



Methanogenesis



### 13 2.2.2. Acidogenesis

14 Acidogenesis is a second phase of anaerobic digestion. This process converts soluble sugars,  
15 amino acids, and fatty acids into short chain volatile fatty acids (VFAs: formic, butyric, propionic, and  
16 acetic acid), alcohols, aldehydes, hydrogen and carbon dioxide as shown in the Fig. 2. Environmental

1 conditions and bacterial type can vary the characteristics of acidogenic products. Sometimes hydrolysis  
2 and acidogenesis can be performed by the same facultative and/or obligate anaerobic bacteria e.g.  
3 *Clostridium*, *Micrococcus*, *Pseudomonas*, and *Flavobacterium* (Xie et al., 2014). Acidogenic bacteria  
4 are rapid growing micro-species and are affected by environmental changes (Visvanathan &  
5 Abeynayaka, 2012). The accumulation of VFAs during the process could lead to low pH conditions  
6 which are not suitable for the growth of methanogens during the next phase.

### 7 2.1.1 Acetogenesis

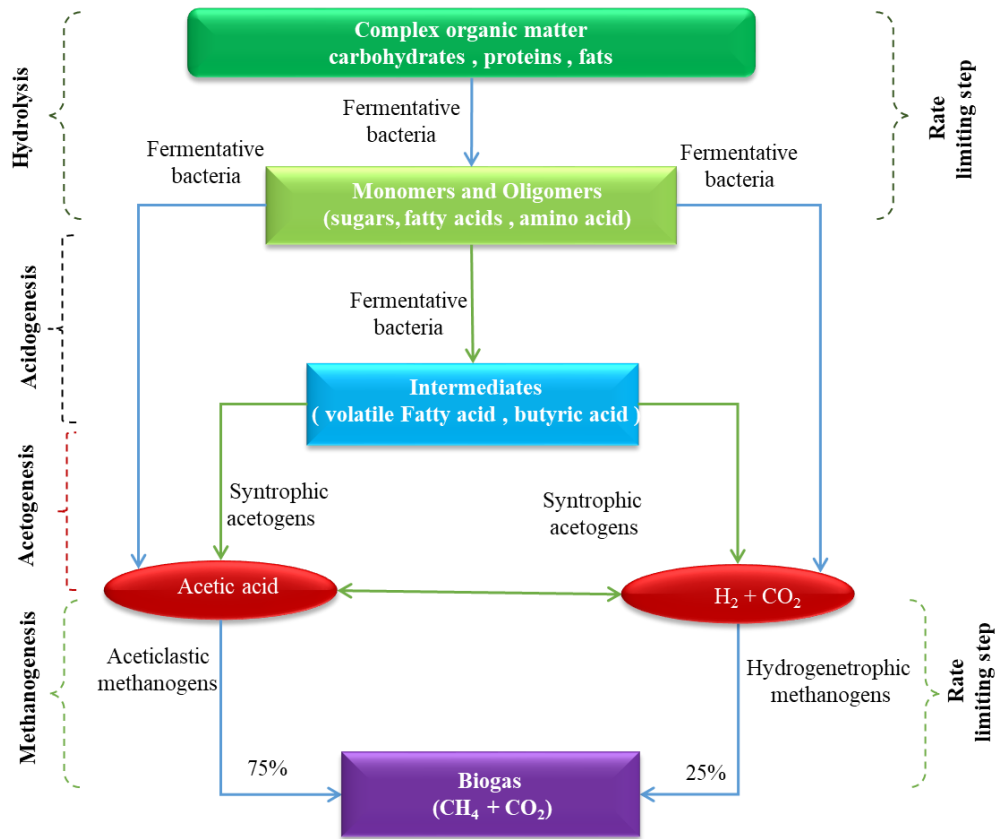
8 Acetogenesis is a third stage of anaerobic treatment process presented in the Fig. 2. This  
9 process is aided by acetogenic bacteria that convert VFAs, alcohols, and hydrogen into acetate. (Xie et  
10 al., 2014). The process of acetogenesis is led by two groups of bacteria: oxidation of propionate,  
11 butyrate, and long chain fatty acids to acetate (obligate H<sub>2</sub> producing bacteria), and intake of H<sub>2</sub> and  
12 CO<sub>2</sub> to produce acetate by homoacetogens (Ketheesan & Stuckey, 2015). Homoacetogens also  
13 consume sugar and alcohols, generating acetate and H<sub>2</sub>. Acetogenic bacteria include *Clostridium*,  
14 *Acetobacterium*, and *Sporomusa* (Ketheesan & Stuckey, 2015).

### 15 2.1.2 Methanogenesis

16 Last process during anaerobic digestion is methanogenesis in which methane is produced.  
17 Methane production is a two way process in the anaerobic digestion as shown in Fig. 2. In the first  
18 way, methane is produced from acetate by aceticlastic methanogens (*Methanosarcina* and  
19 *Methanosaeta*) (Ketheesan & Stuckey, 2015). Approximately, 70% methane is produced via  
20 aceticlastic methanogenesis. Second way of methane production is through hydrogenotrophic  
21 methanogens which converts carbon dioxide and hydrogen to methane and carbon dioxide (Xie et al.,  
22 2014). Methanogenic activity is the measurement of methane production rate which depends on  
23 availability of methane formers and substrate for methane yield. Any change in methanogenic activity  
24 presents obstacle or slow collection of degradable or non-degradable organic load. Methanogens are



1 responsible for methanogenesis (methane production pathway) which are group of archaea (Hussain &  
 2 Dubey, 2017).

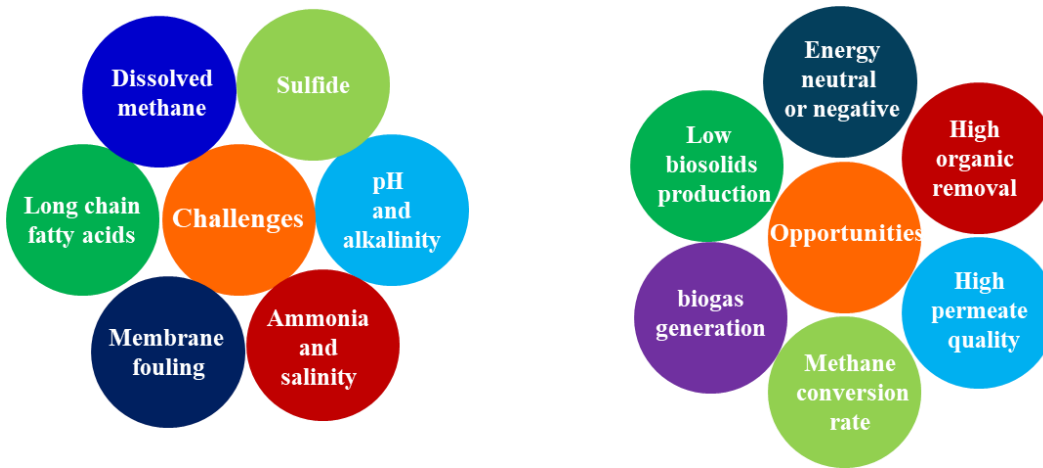


3  
 4 **Fig. 2.** Anaerobic treatment in AnMBRs

5  
 6 **3. Challenges and opportunities in AnMBRs development**

7 *3.1 Membrane fouling*

8 The main challenges associated with the scale-up and commercialization of AnMBRs and  
 9 achievable benefits by treating wastewaters using AnMBRs are presented in Fig. 3. Among them  
 10 fouling is considered as the most important one. This is due to the challenges in fouling control and  
 11 limited life time of membranes. Membranes are still the major contributor to capital and operational  
 12 costs of AnMBRs. Nevertheless, AnMBRs are more economical compared with conventional aerobic  
 13 treatment processes. In order to implement a proper fouling control system, it is important to  
 14 understand the type of membrane fouling discussed below.



**Fig. 3.** Challenges and opportunities in AnMBRs

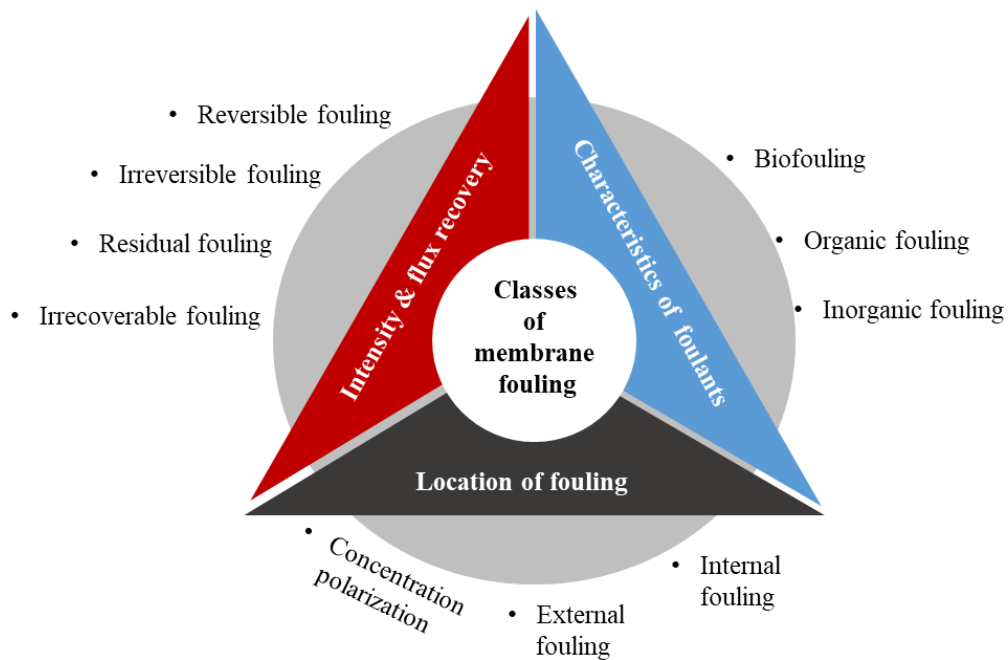
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External fouling occurs by the deposition of particles, colloids, and macromolecules with larger than pore sizes of membrane. It is divided into two types on the basis of layer formation. First one is cake layer which leads to agglomeration of grasp solids on membrane and has reversible character. The other is gel layer, formed by precipitation of soluble macromolecules, colloids, and inorganic solutes. It is characterized by irreversible fouling (Ruigómez et al., 2016; Wang et al., 2014). Different fouling types are presented in Fig. 4. Internal fouling occurs in the presence of fine particles, solutes, and undissolved matter that are submerged or retained inside the membrane pores. During internal fouling, porous membranes are blocked due to “pore blocking” by particles having an equal or smaller diameter compared with the membrane pores and generates irreversible fouling (Aslam et al., 2018a; Bagheri & Mirbagheri, 2018). During this continuous filtration process, direct flow path of foulants to membrane surface is higher than that due to backflow diffusion to bulk solution before steady state. This phenomenon in which accumulation of rejected components in the form of thin liquid layer occurs adjacent membrane surface called “concentration polarization” and this is a reversible and inherent phenomenon during membrane filtration (Wang et al., 2014). This can be mitigated by increasing cross flow velocity. This creates negative impact on transmembrane flux. Concentration polarization can be controlled by pulsation, velocity adjustment, ultrasound, or an electric filed (Bagheri & Mirbagheri, 2018).

1 Membrane fouling can be distinguished in further two types such as reversible and irreversible  
2 fouling based on cleaning practice (Lin et al., 2013). Reversible fouling corresponds to cake formation  
3 defined as porous layer rejected on membrane surface. Reversible fouling tends to remove by physical  
4 means such as relaxation or backflushing (Calderón et al., 2011). Irreversible fouling is a fouling due  
5 to pore clogging/blocking or gel layer formation during long-term operation under sub-critical flux  
6 conditions, which cannot be diminished by physical cleaning methods. Irreversible fouling can cause  
7 by high molecular weight (MW) protein, carbohydrate compounds and EPS, as result inner pores are  
8 packed with these materials (Vyrides & Stuckey, 2009). Residual fouling corresponds to accumulation  
9 of fat, protein, and minerals that can be linked to different fouling mechanism. This type of fouling can  
10 be removed by recovery cleaning but it is difficult to clean residual fouling by chemically enhanced  
11 backwashing or maintenance cleaning (Wang et al., 2014). Irrecoverable fouling associated with long  
12 term experiment when if membrane is fouled once then original membrane permeability never  
13 achieved. This remaining resistance is termed as “irrecoverable fouling” and cannot be removed by  
14 typical chemical cleaning. It is also called as “long term irreversible fouling” or “permanent fouling”  
15 (Wang et al., 2014).

16 Biofouling is the interaction of components of biological treatment broth with the membrane  
17 surface. It is further divided into three types: pore clogging, sludge cake formation and adsorption of  
18 extracellular polymeric substances (EPS) (Lin et al., 2014). Pore clogging is caused by cell debris and  
19 colloidal particles (Meng et al., 2017). Sludge cake formation occurs when shear stress is not enough  
20 to remove solid contents which are mainly composed of biomass and struvite. The increased hydraulic  
21 resistance of membranes is due to formation of cake layer on the surface of membranes. Thickness of  
22 cake layer depends on concentration of suspended solids in the bulk phase, operating pressure applied  
23 and compressibility of the cake layer (Shin & Bae, 2018). Biofouling also occurred by accumulation of  
24 extracellular polymeric substances (EPS) and soluble microbial products (SMP) on membrane and  
25 pore surfaces (Aslam et al., 2018a). Organic fouling is caused by accumulation of macromolecular

1 species (biopolymer) and organic constituents on the membrane surfaces. Organic fouling in AnMBRs  
 2 may increase due to relatively high effluent COD concentration as compared to aerobic MBRs. In  
 3 other words, high organic loading rate (OLR) will cause high CODs and lower membrane fluxes  
 4 (Huang et al., 2011). In contrast, inorganic fouling usually refers to the scalants, inorganic colloids and  
 5 crystals on membrane and pore surfaces. The most common inorganic foulant is struvite  
 6 ( $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$ ) precipitates (Stuckey, 2012; Wang et al., 2014). It can occur on both organic and  
 7 inorganic membranes. Other than this, most common inorganic foulants are  $\text{K}_2\text{NH}_4\text{PO}_4$  and  $\text{CaCO}_3$   
 8 (Stuckey, 2012).



9  
10 **Fig. 4.** Classifications of membrane fouling in AnMBRs

11 **3.2 Sulfide**

12 Sulphide concentration cannot be neglected in AnMBRs application for wastewater treatment  
 13 because of its impact on COD to  $\text{CH}_4$  conversion (Chen et al., 2016). The total methane production can  
 14 be affected by influent sulphate content and thereby enhanced overall operating cost (Pretel et al.,  
 15 2014). The operating cost of AnMBR by using low or none sulphate content having municipal  
 16 wastewater was minimized upto 28%. AnMBR is more efficient technology in net energy production

1 for low sulphate content wastewaters in warm climates. High sulphur content has variations in biogas  
2 production and also increases H<sub>2</sub>S production during AnMBR operation. The presence of sulphate can  
3 precipitate non-alkaline metals (e.g. Fe and Co) in AnMBRs, which will limit the availability of  
4 micronutrients for methanogens. The other disadvantage of precipitation is membrane fouling. H<sub>2</sub>S is  
5 produced as result of reduction of sulphate which is toxic, corrosive, and malodourous gas (Park et al.,  
6 2014). H<sub>2</sub>S gas can easily pass through bacterial cell membrane and destroy native proteins inside  
7 cytoplasm. The performance of AnMBRs for wastewater treatment may be defected by competition  
8 between sulphate reducing bacteria (SRB) and methane producing archaea (MPA) for present carbon  
9 due to faster growth of SRB (Lei et al., 2018; Shin & Bae, 2018). SRB has low response to  
10 temperature change, and therefore at ambient temperature, SRB may have more strength for available  
11 carbon source. Generally, SRB competes MPA for hydrogen on the basis of available electron donors  
12 competition between SRB and MPA; and SRB complete more efficiently for hydrogen than that of  
13 MPA (Lei et al., 2018). So, SO<sub>4</sub><sup>2-</sup>-S induced low COD to methane yield due to competition between  
14 hydrogenotrophic SRB and MPA. Because SRB are oxidizers at both low and high COD/SO<sub>4</sub><sup>2-</sup>-S ratio  
15 and acetoclastic MPA is dominant at high ratio, so there is lack of hydrogen for hydrogenotrophic  
16 MPA and SRB. This will increase conversion of COD to methane (Lei et al., 2018). Sulphate content  
17 in wastewater minimized available COD for methanation because MPA species outcompeted by SRB  
18 for available substrate and biogas production per liter of treated wastewater is affected by COD/SO<sub>4</sub><sup>2-</sup>-  
19 S ratio (Giménez et al., 2012). Propionate is also used by SRB in hydrolysis process that has positive  
20 effect on system (Li et al., 2015). SRB has the advantage of keeping sulfate and sulfide concentration  
21 at low level and enhance biogas production at COD/SO<sub>4</sub><sup>2-</sup>-S>15 (Shin et al., 2014). In some cases, it  
22 was also observed that sulphate content is beneficial for degradation of propionic acid and promotes  
23 methane production when sufficient organic matter is supplied (Li et al., 2015). Generally, maintaining  
24 low sulphate concentration at optimum point is necessary in AnMBRs.

### 25 *3.3 pH and alkalinity*

1 An appropriate pH range is important factor for efficient methanogenic digestion system.  
2 Values outside this range can be harmful for system especially to methanogenesis, so it is important to  
3 control pH for AnMBR system. The operating temperature and nature of wastewater affect the pH and  
4 alkalinity in the system. Nevertheless at temperature below 20 °C, gaseous compounds become more  
5 soluble. So it means that reactor pH goes down due to dissolved amount of hydrogen sulphide and  
6 carbon dioxide under psychrophilic condition (Lei et al., 2018). Shin et al. (2014) operated staged  
7 fluidized membrane bioreactor (SAF-MBR) for primary settled domestic wastewater with temperature  
8 ranges from 8 to 30 °C. It was observed that pH range and alkalinity remain same at 6.6-6.8 and 220-  
9 260 mg CaCO<sub>3</sub>/L respectively in both staged reactors with 4.6-6.8 h hydraulic retention time (HRT)  
10 and average COD of 198-285 mg/L (Shin et al., 2014). Many studies indicated that methane  
11 production mostly occurs at pH ranges from 6.5 to 8.5 and the process is badly affected by pH below  
12 6.5 or rises above 8.5 (McCarty & Rittmann, 2001; Weiland, 2010). Many chemical species can also  
13 contribute to enhance alkalinity in anaerobic digestion which is HCO<sub>3</sub><sup>-</sup>, NH<sub>3</sub>, Ac<sup>-</sup>, S<sup>-2</sup>, OH<sup>-</sup>, HS<sup>-</sup>, and  
14 CO<sub>3</sub><sup>-2</sup>. The major controlling parameters in methanogenic process are carbonic acid related system,  
15 mainly HCO<sub>3</sub><sup>-</sup>, and its concentration equals total alkalinity (McCarty & Rittmann, 2001).

### 16 *3.4 Dissolved methane*

17 The major byproduct of AnMBR is the methane gas produced during biodegradation of organic  
18 matters (Khanal, 2008). Overall energy used during anaerobic treatment of wastewaters using  
19 AnMBRs is lower than the energy generated by methane which has a high energy content of 55,525  
20 kJ/kg (Khanal, 2008). Substrate composition has effective control on methane production rate (Khan et  
21 al., 2016). Methane gas is major constitute of greenhouse gases and has the potential of aggravating  
22 global warming 25 times more than carbon dioxide (McCarty et al., 2011). Due to this reason,  
23 emission of methane should be controlled. Methane in gas form is easy to capture, but it is difficult  
24 task to collect it in dissolved form. Methane solubility is more at 15 °C as compared to 35 °C with 70%  
25 methane content of biogas (Smith et al., 2012). It was reported that half of methane production (50%)

1 remained in dissolved form in liquid phase at 15 °C operational temperature (Smith et al., 2013). The  
2 dissolved methane in AnMBR permeate accounts for 25-67% of the total methane depending on the  
3 temperature from 15-25 °C (Smith et al., 2012). Several strategies have been reported to recover  
4 dissolved methane e.g. stripping of AnMBR effluent through post-treatment aeration, using degassing  
5 membrane reactor, and use of down-flow hanging sponge reactor (Crone et al., 2016; McCarty et al.,  
6 2011; Shin et al., 2016b). The releasing methane in biogas is low compared to dissolved methane in  
7 liquid phase in anaerobic processes. The dissolved methane is up to 38 mgL<sup>-1</sup> (Crone et al., 2016). The  
8 most feasible approach to recover methane from liquid phase is bubbling with air or another gas by  
9 using bubble column reactor. More than 30% of methane of collected gas is needed for electricity  
10 generation (Crone et al., 2016). In another approach there is a direct contact between liquid and gas  
11 phases in the hollow fiber membrane contactor in which methane diffused from liquid phase to gas  
12 phase. In this process more than 72% methane is recovered for power generation (Crone et al., 2016;  
13 Shin et al., 2016b). Based on the considerable potential for dissolved methane, future research are  
14 needed to evaluate trade-off between recovery efficiency and operation cost.

#### 15 **4. Strategies to control membrane fouling and system performance**

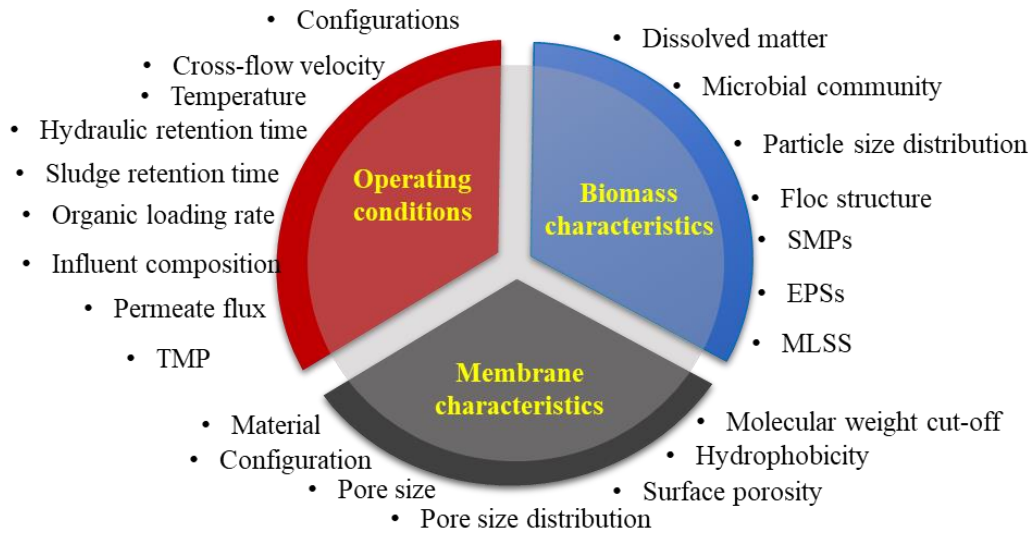
16 Fig. 5 presents key bottlenecks controlling membrane fouling in AnMBRs. Membrane fouling  
17 can either be controlled by optimizing the operating conditions or surface modifications of membrane  
18 or developing membranes less prone to fouling or controlling biomass properties.

##### 19 *4.1 Optimization of operational conditions*

###### 20 *4.1.1 Temperature*

21 Viscosity of liquid decreases with rise in temperature, which improves flux through membrane.  
22 To obtain same shear stress, a lower shear rate will enough for low viscosity sludge and hence  
23 demands lower energy (Dereli et al., 2012). Methanogenic activity is significantly controlled by  
24 temperature change. Different experimental studies revealed that methanogenic activity reduced at  
25 lower temperature (15 °C) as compared to higher temperature (25 °C) (Ho & Sung, 2009; Jain, 2018).

1 The temperature of AnMBR could be higher (32 °C) as compared to temperature of aerobic process  
 2 (29 °C) (Berube et al., 2006; Zhao et al., 2018). The effect of temperature when permeate flux  
 3 increased over 30% by increasing temperature from 40 to 47 °C (Berube et al., 2006). When two lab-  
 4 scale AnMBRs were run at 25 and 15 °C, COD removal efficiency was more at high temperature (25  
 5 °C) as compared to lower temperature (15 °C) (Hu & Stuckey, 2007). Mostly, anaerobic processes are  
 6 run at mesophilic (35 °C) and thermophilic (55 °C) temperatures. These temperatures are used where  
 7 streams consist of high contents of solids in municipal wastewater sludge. In these streams, high  
 8 reaction rates with increase in temperatures and decrease reactor sizes (Khanal, 2008; Liao et al.,  
 9 2006). From economical point of view, AnMBR is to be treated at ambient temperature.



10  
11 **Fig. 5.** Membrane fouling: controlling parameters

12 **4.1.2 Organic loading rate (OLR)**

13 OLR is another operating parameter for fouling control as shown in Fig. 5. Organic loading  
 14 causes filtration resistance. Fluctuations in organic loading can be entertained by AnMBR. In AnMBR  
 15 for biogas production, OLR ranges from 0.23 to 33.7 kgCODm<sup>-3</sup>d<sup>-1</sup> (Chen et al., 2016). OLR indicates  
 16 the quantity of volatile solids that treated in reactor per day. More biogas production is achieved by  
 17 increasing organic loading rate. Wijekoon et al. (2011) reported this fact that biogas production rate  
 18 was proportional to organic loading rate. When organic loading rate was increased from 5 to



1 12 kgCODm<sup>-3</sup>d<sup>-1</sup> then biogas production rate also increased from 5 to 35 Ld<sup>-1</sup> in two stage thermophilic  
2 AnMBR. Higher the OLR, higher will be activity of methanogens that increase the biogas production.  
3 Disadvantage of increasing OLR is accumulation of VFA that requires irreversible acidification and  
4 will deteriorate biogas production (Wijekoon et al., 2011).

#### 5 4.1.3 HRT

6 HRT play a vital role in the performance of AnMBR. HRT is a time interval in which waste to  
7 be treated remained in contact with microbial biomass to achieve certain degree of treatment (Khanal,  
8 2008). Values range for HRT could be from a few hours i.e., 2h to a few days i.e., 20 days (Jeong et  
9 al., 2010). HRT is controlling parameter from economic perspective. In this sense, it allows smaller  
10 biogas producing AnMBR for shorter HRTs. Depending on feed characteristics, system hydraulics,  
11 and sludge properties etc., the HRT values can be changed from as low as 1 h to as high as 30 d (Chen  
12 et al., 2016; Jeong et al., 2010; Stuckey, 2012). Ho and Sung et al. (2009) highlighted the effect of  
13 HRT on methane recovery by reducing HRT from 12 to 6 h. The methane production from municipal  
14 wastewater decreased by 13% as result of increasing accumulation in AnMBR (Ho & Sung, 2009).  
15 Therefore, methane yield can be enhanced by optimum HRT. It was reported that methane production  
16 decreased by increasing HRT due to lack of COD availability as substrate for methane production. As  
17 HRT lowers, the biogas production increased due to increasing organic loading rate in SAnMBR.  
18 However, it was not recommended to close HRT value too short due to high biomass concentration  
19 that could deteriorate membrane fouling (Khanal, 2008). Gao et al. (2014b) observed an increase in  
20 methane production by lowering HRT from 8 to 6 h, that might connected to increasing OLR.  
21 Meanwhile, the biogas productivity decreased with much shorter HRT due to more VFAs  
22 accumulation. There could be an optimized value of HRT for each case depending on feed  
23 characteristics, system hydraulics, reactor design, substrate types etc. (Gao et al., 2014). Disadvantage  
24 of shorter HRT may lead to membrane fouling, VFA accumulation, reduction of biogas production for  
25 wastewater treatment process.

#### 1 4.1.4 *Upflow velocity*

2 The key parameter that plays a vital role in performance of AnMBR is upflow velocity that has  
3 two opposing effects on biological removal efficiency. A better mixing of substrate-biomass may be  
4 achieved by increasing upflow velocity. On the other hand, disadvantage of increasing upflow velocity  
5 may come in form of detachment of captured solids due to high hydraulic shear force and damage  
6 removal efficiency by increasing settling velocity. By using effluent recirculation, the results revealed  
7 better COD removal efficiency at higher upflow velocities. Filterability in AnMBRs is increased by  
8 applying upflow velocity that increased shear stress, resulting in lower fouling. The effect of upflow  
9 velocity and permeability increased with increase in upflow velocity (Ozgun et al., 2015). However,  
10 some studies contradicted that upflow velocity provided strong shear impact, which broke up sludge  
11 aggregate into small/fine particles in mixed liquor and enhance amount of microbial byproducts,  
12 leading to more fouling. Permeate flux can be reduced by size reduction of biosolids and concentration  
13 of microbial byproducts due to pore clogging and cake formation on the surface of membrane (Berube  
14 et al., 2006).

#### 15 4.1.5 *SRT*

16 Like HRT, SRT is also a critical parameter for controlling irreversible membrane fouling. At  
17 high value of SRTs, the internal pore blocking increases due to large concentration of responsible  
18 foulants. Larger SRTs also cause less flocculation of particles due to large concentration of  
19 carbohydrates and proteins in SMP and can result in smaller particle sizes which can lead to membrane  
20 fouling (Liao et al., 2006). Combination of longer SRT with shorter HRT, cause high concentration of  
21 mixed liquor suspended solids (MLSS) and SMP within AnMBR tanks, which results in cake  
22 formation due to particle deposition. Many researchers investigated values of SRT ranged from 20 d to  
23 infinite days (Dereli et al., 2014a; Dereli et al., 2014b). AnMBRs with high values of SRTs produce  
24 the larger amount of biogas because lower values of SRTs reduce the extent of reactions required for  
25 stable digestion. Longer SRTs have advantage of low disposal cost and minimal sludge production, but

1 can have adverse effect on methanogenic activity due to reduction of viable biomass concentration.  
2 Longer SRTs also have a disadvantage of membrane fouling due to small particle size and  
3 accumulation of SMP. Inorganic fouling can also occur due to inorganic solids at high SRTs (Skouteris  
4 et al., 2012).

#### 5 *4.1.6 pH*

6 The pH of effluent is usually examined by the concentration of VFA. Bacterial growth occurs  
7 by applying different range of pH from 4.0 to 8.5 but pH level of 6.8 to 7.2 is also beneficial for  
8 methanogenic bacteria. A pH range between 5.5 and 6.5 is mostly favorable for hydrolysis and  
9 acidogenesis (Ozgun et al., 2013). Maximum biogas production of 16,607 mL was obtained at pH 7.0  
10 but generation of biogas decreased at pH 6.0 and 8.0 (Musa et al., 2018). It was noted that pH 6.5 and  
11 7.5 had similar fouling and was less severe than pH 5.5 and 8.5. Mostly neutral pH is used for AnMBR  
12 operation. Feasible range for methane fermentation is within pH 6.5-8.5 with optimal range from 7.0 to  
13 8.0 (Weiland, 2010). Such pH range is usually controlled through neutralization process in which  
14 excessive amounts of chemicals such as sodium carbonate/bicarbonate or calcium carbonate are used.  
15 Extreme pH conditions have adverse effects on performance of AnMBRs in form of disturbing  
16 biological performance and methane generation and also affect lifespan and membrane permeability  
17 (Weiland, 2010).

#### 18 *4.2 Membrane characteristics*

19 The other principal factor that affects membrane fouling is the membrane characteristics that  
20 include morphology, materials, pore size, zeta potential, and hydrophobicity/hydrophilicity affinity  
21 (Kochkodan & Hilal, 2015). The modification of membrane surface is done to increase hydrophilic  
22 character of surface by altering the physical or chemical interactions with membrane surface (Meng et  
23 al., 2017). Membrane fouling associated with hydrophilicity/hydrophobicity of membrane surface  
24 when two contacting surfaces are infinite planar. It was reported that interaction between membrane  
25 surface and sludge foulants remarkably affected by surface roughness and zeta potential than

1 membrane hydrophilicity/hydrophobicity. Membrane fouling was reduced by using high zeta potential  
2 and certain roughness (Kochkodan & Hilal, 2015).

### 3 *4.2.1 Membrane configuration*

4 AnMBR consists of bioreactor coupled with membrane filtration for separation of liquid and  
5 solid. There are two types of filtration methods: (a) dead end filtration that has high accumulation rate  
6 and low cost; and there is no requirement of backwashing and chemical cleaning but it has high  
7 frequency for membrane change; (b) cross flow filtration where membrane filter exchange frequency is  
8 low and can be reused after backwashing and chemical cleaning (Aslam et al., 2017a). It has  
9 disadvantage of high cost and low collection rate. Three main configurations of membrane filtration  
10 that commonly used in an AnMBR are (a) external cross flow (b) internal and (c) external submerged  
11 as shown in Fig. 1 and discussed in section 2.1. In external cross flow membrane configuration,  
12 pressure is used for operation because membrane is not inserted in bioreactor and pump is required to  
13 force effluent into membrane unit. Because the membrane is situated outside from reactor so it is easier  
14 to clean and makes replacement simple. The drawback of this technique is that sludge adheres to  
15 membrane surface easily. It requires high power and energy cost (Smith et al., 2014). In internal  
16 submerged configuration, the membrane filtration is placed in AnMBR from where effluent draws out  
17 through membrane by vacuum. It has advantage of low energy consumption. While in external  
18 submerged assembly, the membrane is fitted in separate reactor. It has higher fluxes and is easier to  
19 replace (Lin et al., 2013).

### 20 *4.2.2 Membrane pore size and type*

21 Other than membrane configuration, there are some properties that characterize membrane.  
22 These are nominal pore size and type of membrane material. Nominal pore size has significant effect  
23 on permeate flux (Berube et al., 2006). It is clear that membrane pore size is function of microbial  
24 mixed liquor being treated. On the basis of membrane pore sizes, AnMBRs are mostly packed with MF  
25 and UF. Hollow fibers can be inserted in submerged reactors (Kim et al., 2011a; Liu et al., 2013; Yoo

1 et al., 2012). Nevertheless, flat sheets have been examined due to high stability, easy to clean and  
2 exchange (Lin et al., 2011). Tubular membranes can also be used because these have low membrane  
3 fouling (Calderón et al., 2011). On the other hand, it requires high energy input because it operates at  
4 high pressure for filtration (Stuckey, 2012). There is higher rate of fouling for large membrane pore  
5 size as compared to small pore size. AnMBR wastewater treatment process consists of membrane pore  
6 sizes of 0.02-0.5  $\mu\text{m}$  (Ozgun et al., 2013). On the basis of material, membranes can be divided into  
7 three types as: polymer, metallic and ceramic (inorganic). The advantage of using polymeric  
8 membranes is of lower cost as compared to others (Stuckey, 2012). The disadvantages of polymer  
9 membrane are lower permeability and reduced stability towards chemical cleaning. Backwashing of  
10 ceramic membranes providing with high resistance to corrosion, abrasion and fouling can make it more  
11 announced as compared to others.

#### 12 *4.3 Biomass characteristics*

##### 13 *4.3.1 Mixed liquor suspended solids (MLSS)*

14 Permeate flux has been significantly affected by concentration of suspended solids in mixed  
15 liquor. The effect of MLSS concentration of 35 g/L was investigated that required TMP two times  
16 greater than at MLSS concentration of 7 g/L (Stuckey, 2012). More accumulation of colloids, sludge  
17 particles, macromolecular matter, and microbial products in bioreactors was observed at higher MLSS.  
18 Higher sludge viscosity will lower rise velocity of larger biogas bubbles, lower effect of gas bubbling  
19 on membrane fouling mitigation and bothered membrane fiber vibration in bioreactors (Deng et al.,  
20 2016). Moreover, high sludge viscosity also damaged back transport effect and in consequence  
21 increases attraction of sludge flocs, biopolymers, and smaller particles to membrane surface. Thus,  
22 sludge with high MLSS and viscosity would damage membrane performance in the form of reduce  
23 membrane filterability, rapid flux decline, higher membrane fouling resistance and TMP (Meng et al.,  
24 2017).

##### 25 *4.3.2 Floc size*

1 Permeate flux is positively affected by sludge floc size. Large flocs might be pulled away from  
2 membrane by high inertial force and high shear induced diffusion as well as low Brownian diffusion.  
3 More porous and permeable cake layer formed with larger and loosely deposited flocs, thus reduced  
4 fouling resistance. The motions of small flocs were controlled by Brownian diffusion at lower shear  
5 stress (Skouteris et al., 2012). When investigates effect of floc size on membrane fouling, it is also  
6 important to put an eye on membrane pore size. Shen et al. (2015) reported that there were no more  
7 pore clogging with membrane pore size of 0.3  $\mu\text{m}$  and flocs size greater than 1  $\mu\text{m}$ . According to the  
8 thermodynamic analysis, decreasing floc size increases specific energy barrier. Hence, smaller flocs  
9 retain on membrane surface and form less porous cake layer with smaller pore size and thus increased  
10 hydraulic cake resistance and osmotic pressure induced resistance (Deng et al., 2016; Shen et al.,  
11 2015).

#### 12 4.3.3 Soluble microbial products (SMP) and Extracellular polymeric substances (EPS)

13 Microbial byproducts such as SMP and EPS had strongest engagement with membrane fouling  
14 when compared to other sludge characteristics such as MLSS, floc size, particle size distribution etc.  
15 High Brownian diffusion and permeation drag might be occurred due to retention and adsorption of  
16 SMP and EPS on or into membrane. This engagement of these components to membrane can be  
17 achieved by high interaction energy and by overcoming repulsive energy barrier (Deng et al., 2016;  
18 Meng et al., 2017). Thus, this will cause membrane pore blocking by gel formation and seep into pores  
19 and spaces of cake layer. SMP and EPS with macromolecular property assembled major soluble  
20 organic substances in gel and cake layers. Gel layer clutched more SMP/EPS as compared to cake  
21 layer and thus had more filtration resistance than cake layer. These effects directed negative impact on  
22 permeate flowrate by membrane fouling (Hong et al., 2014). Recent studies showed that membrane  
23 fouling was impacted through molecular weight (MW) dissemination of these microbial byproducts.  
24 The pore blocking and flux decline was reported with large molecular weight fraction of SMP (>10  
25 kDa) (Lin et al., 2013). The compositions have also strong effect on membrane fouling especially in

1 form of proteins/polysaccharides ratio. Internal or irreversible fouling decreased as ratio increases and  
2 contributed cake layer formation on membrane surface (Ozgun et al., 2013). However, another study  
3 showed that filtration resistance increased with increasing ratio and had negative impact on cake layer  
4 (e.g. lower porosity, small floc size, higher thickness) during industrial wastewater treatment (Gao et  
5 al., 2013). Biopolymers had transformable concerning size distribution and presented appropriate size  
6 distribution in mixed liquor (Aslam et al., 2015).

#### 7 *4.4 Pretreatment of feed*

8 The characterization of feed has advantageous impact on membrane fouling. The refuse or  
9 debris present in some industrial wastewater can plug coarse bubble diffusers. The unbalanced  
10 conditions of pH of wastewater have adverse effect on membrane permeability and lifespan. It can also  
11 alter the biological activity of microbes. It was observed that cake layer on membrane surface was  
12 populated with elements such as Mg, Al, Ca, Si and Fe (Stuckey, 2012). The contact of these elements  
13 with biopolymer materials had strong impact due to cake formation and compactness of cake layer.  
14 Due to this reason, these elements should be removed from wastewater through pretreatment  
15 approaches like filtration, pH adjustment, establishment of local wastewater treatment. Kim et al.  
16 (2007) for  $\text{NH}_4^+$  removal used dialyzer/zeolite unit in the influent and achieved 90% removal  
17 efficiency. This pretreatment reduced struvite precipitation on ceramic membrane in AnMBR (Kim et  
18 al., 2007). Pre-sedimentation has also been observed as a feasible and economic way for pre-treatment  
19 of feed. The results declared that pre-sedimentation has significant effect than fine screening.

#### 20 *4.5 Membrane scouring methods*

21 Membrane fouling is controlled by applying shear stress through three different approaches:  
22 biogas sparging, particle sparging, and rotating/vibrating membrane. Biogas sparging is commonly  
23 used for controlling membrane fouling in AnMBRs. In this technique, the uplifting gas bubbles enter  
24 back from downside of membrane module and create turbulence. Turbulence of liquid has scouring  
25 effect on membrane surface by back movement of foulants from membrane surface. Specific gas

1 demand per unit membrane area ( $SGD_m$ ) is indication of biogas sparging intensity. Gas sparging with  
2 sludge recirculation in pilot scale AnMBRs provide better mixing and enhance turbulence on  
3 membrane surface (Shin & Bae, 2018). Shear force can be applied by liquid recirculation and/or gas  
4 sparging. Xie et al. (2014) observed the effect of biogas sparging rate from 0.3 to 0.75  $Lmin^{-1}$ . By  
5 increasing rate, fouling rate decreased and flux increased (Xie et al., 2014). Membrane fouling controls  
6 by periodic washing/cleaning remains a headache during wastewater treatment in AnMBRs. This way  
7 of combating fouling increases membrane replacement cost, decreases system productivity and reduces  
8 membrane lifespan. The absorption and attachment of components of sludge suspension, organic and  
9 inorganic foulant onto or into membrane structure causes the reduction of flux and enhance TMP.  
10 Mitigation of membrane fouling in vibrating AnMBR occurs with different motions/mechanical forces  
11 i.e., longitudinally, transversely, torsionally or their combination that produces the shear effect at  
12 membrane surface. Different types of vibrating MBR that have been studied: transverse vibration  
13 system, vertical movement, magnetically induced membrane vibration and high frequency power  
14 vibration (Bilad et al., 2012; Eliseus et al., 2018; Kola et al., 2014). Another study investigated the  
15 operations of transverse vibration of submerged hollow membrane with permeate flux, where  
16 anaerobic bioreactor effluents enhanced and efficiently worked at high concentration of mixed liquor  
17 suspended solid (Kola et al., 2014).

18 In particle sparging, AnMBR also called anaerobic fluidized membrane bioreactor (AFMBR),  
19 granular activated carbons (GACs) act as fluidized media having scouring action on membrane surface  
20 (Charfi et al., 2017a). GACs are helpful in biofilm growth. This technique has low energy consumption  
21 than biogas sparging (Ahmad et al., 2018; Aslam et al., 2019; Aslam et al., 2014; Kim et al., 2011a;  
22 Shoener et al., 2016). Hu and Stuckey (2007) investigated the effect of GAC and powder activated  
23 carbon (PAC) as fluidized media on membrane fouling under biogas sparging. A positive effect was  
24 observed in the shape of reduction in TMP whereas flux increased in both case (Hu & Stuckey, 2007).  
25 In contrast, Kim et al. (2011a) operated AFMBR using GAC with recycling liquid instead of biogas



1 sparging and resulted in control membrane fouling. Fluidized GAC as scouring agent provided  
2 scouring (mechanical) action on membrane surface and also provided large surface area for biofilm  
3 growth. It had low energy consumption and was highly effective to control membrane fouling (Charfi  
4 et al., 2018a; Kim et al., 2011a). Gao et al. (2014b) also reported the effect of GAC as carrier on  
5 membrane fouling by using integrated anaerobic fluidized bed membrane bioreactor (IAFMBR)  
6 system for wastewater treatment. It reduced membrane fouling and also had long term operational  
7 period. Membrane filtration performance increased by high dosage of GAC as more protein absorbed  
8 by GAC (Gao et al., 2014). PAC, zeolite, or coagulants were attempted to reduce membrane fouling.  
9 The most effective inhibitor was polyaluminum chloride (Aslam et al., 2017a; Zhang et al., 2017).

10 AnMBRs with rotating flat sheet, tubular, hollow fiber or helical membrane modules have been  
11 examined (Jiang et al., 2013; Wu et al., 2008). Fouling can be controlled by increasing rotation speed.  
12 Wu et al. (2008) reported influence of rotation speed of 60 rpm on membrane fouling which has  
13 significant effect on cleaning efficiency. Jiang et al. (2013) stated that fouling rate in rotating flat sheet  
14 MBR was lower as compared to conventional MBRs when consuming same energy. In rotating  
15 membrane technique, turbulence is created by rotation of membrane around axial axis to mitigate  
16 deposition of foulants on membrane surface. In rotating membrane bioreactor, the rotation of  
17 membrane itself around its axis generates shear stress which helps in reducing membrane fouling. This  
18 technique was granted to improve membrane fouling control by increasing shear action with rotation.  
19 In this strategy, fouling contribution is lower than gas scouring application (56-18 versus 60-53%,  
20 respectively) (Ruigómez et al., 2017). Several studies have predicted that flux increases with speed of  
21 rotating membrane that result in enhancing shear forces. Greater shear forces produce uniform  
22 gradients in axial direction that increasing mass transfer (Liao et al., 2006).

#### 23 *4.6 Filtration cycles*

24 Filtration cycles technique consisted of filtration and relaxation with optional backwashing.  
25 Relaxation, duration of time with no permeation, is broadly applied for membrane fouling control.

1 Membrane fouling is also controlled by backwashing in which reverse flow of permeate remove  
2 foulants on membrane surface. Backwashing is useful with low pressure membrane systems e.g.  
3 microfiltration and ultrafiltration. Backwashing is typically significant for cleaning of accumulated  
4 particles over membrane surface, which mainly causes reversible fouling; disentangle loosely adherent  
5 aggregates sludge from surface of membrane (Ozgun et al., 2013). Some studies revealed that  
6 backwashing could be effective for removing not only external fouling but also internal/pore clogging.  
7 Moreover, biogas combined with backwashing can sufficiently control internal fouling. It was also  
8 reported that pore blocking was not fully reversible by backwashing and partially leads to irreversible  
9 fouling (Lei et al., 2018).

#### 10 *4.7 Chemical cleanings*

11 Membrane performance can be restored by chemical cleaning. Membrane permeability is  
12 improved periodically through maintenance cleaning (MC). On the other hand, recovery cleaning (RC)  
13 is used to recover membrane permeability. But chemical cleaning could cause damage to membrane  
14 material and reduce lifespan of membrane module. Chemical cleaning is occurred through six steps as:  
15 bulk reaction of cleaning reagents, transportation of cleaning reagent to interface, penetration of  
16 cleaning reagent into fouling layer, solubilization and detachment of foulants, back transportation to  
17 interface of waste cleaning agent and detached foulants; and transportation of waste into bulk solution  
18 (Bagheri & Mirbagheri, 2018). Chemical cleaning is carried out under following processes: In-situ and  
19 ex-situ. Under in-situ chemical cleaning process, membrane module is not transferred and membrane  
20 permeability is recovered more easily. When membrane is more fouled then ex-situ chemical cleaning  
21 process is used to extract out membrane module from bioreactor and by inserting it into separate  
22 cleaning tank with chemical reagents. Chemical cleaning may include acids, bases, oxidants,  
23 surfactants and chelates (Meng et al., 2017; Wang et al., 2014)

24 Cleaning of fouled membrane through acids is more effective and low cost approach.  
25 Precipitated salts are removed from membrane surface and pores by using acids cleaning. Acids

1 cleaning can be used to remove precipitated inorganic matters and mineralization that exert between  
2 biopolymer and salts (Malaeb et al., 2013b). Oxalic, citric, nitric, hydrochloric, phosphoric and sulfuric  
3 acids are commonly used for membrane cleaning. Citric acid has advantage of decreasing risk of pH  
4 damage and it also provides buffering (Wang et al., 2014). Organic foulants can be removed by using  
5 bases such as sodium hydroxide deposited on membrane surface in pH range from 11 to 12 (Bagheri &  
6 Mirbagheri. 2018). Weakly acidic organic matters can be removed by using hydroxide with carboxylic  
7 and phenolic functional groups. Bases are used to hydrolyzed and solubilized proteins and  
8 carbohydrates into small molecules. Fats and oils are decomposed by bases such as sodium hydroxide  
9 and produced water soluble micelles through saponification process (Bagheri & Mirbagheri, 2018).  
10 Membrane cleaning through bases includes the following processes: hydrolysis, solubilization, and  
11 saponification. It has advantage of neutralization of acidic organics and reduced number of bonds  
12 existed between foulants and membrane surface (Krzeminski et al., 2017). Oxidants are also used to  
13 clean organic and biological foulants through oxidation and disinfection. Functional groups of organic  
14 foulants are oxidized by oxidants to ketonic, aldehydic and carboxylic groups. Major examples of  
15 oxidants are sodium hypochlorite, hydrogen peroxide, and polyvinylpyrrolidone (PVP)-iodine. Among  
16 all of these, sodium hypochlorite is the most used oxidant to clean fouled membranes (Meng et al.,  
17 2017). The rhamnolipids are also used as chemical cleaning reagents against membrane fouling. They  
18 do not face problem of high cost, low solubility, and toxicity. The presence of rhamnolipids during  
19 membrane cleaning have high degree of biofilm disengagement and effective biofilm minimization.  
20 They reported that membrane fouling decreased due to hydrophobic character of surface exerted by  
21 rhamnolipids through deliverance of lipopolysaccharides and EPSs (Kim et al., 2015b).

#### 22 *4.8 Comparison of different fouling control strategies*

23 There are different pathways through which membrane fouling can be controlled to some  
24 extent and in some methods could be removed to maximum extent. Such strategies have advantages  
25 and drawbacks in different aspects while treating different types of wastewaters in AnMBRs.

1 Deposition of particles on membrane surface can be controlled by cross flow velocity along  
2 membranes. This technique has potential to mitigate filtration resistance resulted by concentration  
3 polarization (CP) and cake formation through higher cross flow velocity (Skouteris et al., 2012).  
4 However, cost related to higher cross flow velocities and formation of fine/colloid particles at high  
5 velocity is a major concern. So high shear force has negative impact on membrane permeate flux in  
6 AnMBR. Ultrasonic irradiation is another path to control membrane fouling. But it becomes unfeasible  
7 at high sludge concentration that requires more ultrasonic irradiation energy for long period of time  
8 (Meng et al., 2017). In addition, high power density and low frequency are required to improve the  
9 cleaning efficiency.

10 Biogas sparging is most commonly used to mitigate membrane fouling through recycling of  
11 biogas at bottom of module in AnMBRs. Biogas sparging rate will definitely define process efficiency  
12 as membrane fouling decreases by increasing biogas sparging rate (Stuckey, 2012). It is an efficient  
13 technique; however, energy expenditure is high. Relaxation and backflushing are two widely employed  
14 hydraulic cleaning protocols. Mechanical cleaning by combining biogas sparging and particles/carriers,  
15 vibration and rotation has also positive effect on membrane fouling control. Adding carriers/particles  
16 provide solid support for biomass growth and causes reduction in floc rupture (Vyrides & Stuckey,  
17 2009). It provides sufficient shear force on membrane surface by mechanical cleaning and reduces  
18 TMP and membrane fouling. However, the possible damage of membrane surface should be taken into  
19 consideration. Physical cleaning is used only to remove cake layer and reversible fouling (temporary  
20 fouling) (Lin et al., 2013). It is a fast process as compared to chemical cleaning and produces no  
21 chemical waste. This cleaning process does not involve membrane degradation. In contrast, chemical  
22 cleaning is used to remove organic, inorganic, and irreversible fouling by using different chemical  
23 cleaning agents such as bases, acids and oxidants (Lin et al., 2013). But these chemicals may be  
24 corrosive or caustic and can damage membrane. The combination of chemical and physical cleaning is  
25 a powerful strategy to enhance cleaning efficiency, for example, chemical-enhanced backwash (CEB)

1 and ultrasound assisted chemical cleaning are two applicable ways. Different novel configurations e.g.  
2 anaerobic fluidized membrane bioreactor, anaerobic dynamic membrane bioreactor, anaerobic rotating  
3 membrane bioreactor, anaerobic bioelectrochemical membrane reactor and anaerobic osmotic  
4 membrane bioreactor has been developing rapidly in recent years to control membrane fouling in an  
5 efficient manner. Particle sparging through bulk liquid recirculation is a promising technology to  
6 mitigate cake layer formation and control membrane filtration resistance (Aslam et al., 2017a).  
7 However, novel cleaning strategies and configurations should be paid attention. Their pilot-scale and  
8 full-scale feasibility should be further verified. Novel chemical foam cleaning, self-cleaning membrane  
9 development and bioelectrochemical cleaning are also worth investigating.

## 10 **5 Novel configurations**

### 11 *5.1 Anaerobic fluidized-bed membrane bioreactor*

12 Recently biogas sparging was combined with biofilm carriers or fluidized media to reduce  
13 biogas flow rates for fouling reduction as shown in Fig. 6a (Vyrides & Stuckey, 2009). However,  
14 energy consumption was still higher because gas sparging accounts for largest fraction of energy  
15 requirement. Anaerobic fluidized bed membrane bioreactor (AFMBR) is a newly developed novel  
16 liquid recirculation particle-sparging system for wastewater treatment that coupled with membrane  
17 filtration without biogas sparging as shown in Fig. 6b (Kim et al., 2011a). Good mass transfer  
18 characteristics and high SRT with high concentration of active microbial species indicate singularity of  
19 this process on other processes. Membrane fouling in this configuration was controlled through  
20 scouring action of GAC, also known as fluidized media, on membrane surface (Aslam & Kim, 2019).  
21 This process has low energy demand also during cleaning of fouled membrane (Aslam et al., 2017b;  
22 Aslam et al., 2018c; Charfi et al., 2018b; Shin et al., 2014). The maximum methane recovery from  
23 wastewater treatment is also acquired by two stage fluidized bed bioreactor. Municipal wastewater  
24 from primary clarifier effluent was investigated by Kim et al. (2011) in coupled system of anaerobic  
25 fluidized bioreactor and anaerobic fluidized-bed membrane bioreactor for methane rebate. The

1 generation of methane from this system was  $4.11 \text{ molCH}_4\text{m}^{-3}$ . The composition of this methane was  
2 86% at less than 5h HRT. Total energy required for fluidization obtained from only 30% gaseous  
3 methane which indicated AFMBR as more propitious bioenergy production technology (Kim et al.,  
4 2011a). Same favorable points for AFMBR system were also derived from study of Yoo et al. (2012)  
5 and Aslam et al. (2018) as it was highly efficient, had low energy demand, and ability of large  
6 bioenergy production.

7         Researchers indicated major problem in methane production during winter season because it  
8 was significantly affected by dissolved methane and temperature. Another simplified and smaller  
9 footprint system was reported by Gao et al. (2014b) which consisted of integrated anaerobic fluidized  
10 bed membrane bioreactor (IAFMBR). According to this study,  $80\pm 2\%$  methane content acquired in  
11 biogas production and conversion of influent COD in methane was about 50% in which 25% of  
12 produced methane went to liquid phase. Shin et al. (2014) reported that enriched acetoclastic culture  
13 grew rapidly on GAC as compared to acetoclastic methanogenic activity (SAMA). Due to this reason,  
14 further research should deal with growth of syntrophic acetogens (VFA-degrading) and acetoclastic  
15 methanogens on GAC for maximum biogas production. Recently, Seib et al. (2016) developed a new  
16 configuration of anaerobic cross-flow particle sparging MBR using GAC fluidized media as presented  
17 in Fig. 6c (Düppenbecker et al., 2017; Seib et al., 2016). Another study investigated glass beads as  
18 fluidized media (Düppenbecker et al., 2017). The fluidized media in cross-flow system provided 55-  
19 120% longer membrane operation between cleanings under reduced cross-flow velocity (CFV),  
20 resulting in 98-99% reduced energy requirements than that of conventional tubular membrane  
21 operational techniques under higher cross-flow velocities of 3-5 m/s (Seib et al., 2016).

## 22 *5.2 Anaerobic submerged rotating membrane bioreactor (AnSRMBR)*

23         The most emerging and frustrating issue in anaerobic membrane bioreactor is membrane  
24 fouling which causes reduction in flux, membrane replacement period, membrane efficiency, and  
25 enhance operation cost. Membrane fouling can be controlled through many attempts such as

1 chemical/physical cleanings to applications of scouring media fluidization and transverse vibration  
2 (Kola et al., 2014; Wu et al., 2017). This issue can be overcome by using novel configuration of  
3 membrane in bioreactor. This novel approach consists of submerged rotating membrane bioreactor as  
4 shown in Fig. 6d (Ruigómez et al., 2017; Wu et al., 2008). In this technique, immersed and rotatable  
5 membrane is used fixed on axis and rotatory movement occurs through electric motor. This strategy  
6 has improved filtration capacity and fouling prevention ability. In submerged rotating membrane  
7 bioreactor, cross flow velocity is function of gas intensity and membrane module configuration (Wu et  
8 al., 2008). Mainly cross flow velocity is produced by rotation of membrane module. A rotary filtration  
9 system is an economically feasible technique than conventional cross flow systems. Another advantage  
10 of this technique is its antifouling action proposed by rotating membrane that increases shear action  
11 between membrane and floating media. This system has large permeate flux in ultrafiltration and  
12 provides strong shear impact that results in reduce cake on membrane surface (Jørgensen et al., 2014).

### 13 *5.3 Anaerobic dynamic membrane bioreactor (AnDMBR)*

14 Problems in conventional AnMBR processes include high cost of membrane configuration, low  
15 membrane flux, and rapid membrane fouling that can be overcome by using dynamic membrane (DM)  
16 technology. In an AnDMBR, supported cake layer configuration is used for solid-liquid separation.  
17 Materials for supporting are meshes and fabrics with macropores (Ersahin et al., 2014). This  
18 technology of DM commutes major problem of AnMBR, namely membrane fouling, into advantage.  
19 After complete fouling of dynamic membrane, cake layer removal and cleaning can occur through easy  
20 way and can be replaced with new deposited layer. By this way, membrane cost is reduced. Chemical  
21 resistance of fillers and dynamic membrane material are helpful in selection of cleaning process and its  
22 frequency (Ersahin et al., 2012; Ma et al., 2013). A large pore size mesh of 200  $\mu\text{m}$  was used by  
23 Alibardi et al. (2014) in lab-scale anaerobic dynamic MBR. They reported the effect of variation in  
24 COD removal and HRT on changing in biogas production and methane content that was 1.0 L/d  
25 maximum and 50-79% respectively. Sustainable aspect of AnDMBR occurs by low CFV (due to large

1 pore size) that had low energy input and improved methanogenic activity by reducing shear stress on  
2 biomass. Maximum amount of biogas was produced due to higher methanogenic activities of cake  
3 layer (Alibardi et al., 2014).

4 Application of dynamic membrane on monofilament woven support was also investigated by  
5 (Ersahin et al., 2014). They treated synthetic concentrated wastewater in AnDMBR. They observed  
6 different average production of methane at 20 and 40 days SRTs which was  
7  $0.31 \pm 0.02 \text{ LCH}_4/\text{gCOD}_{\text{removed}}$  and  $0.34 \pm 0.04 \text{ LCH}_4/\text{gCOD}_{\text{removed}}$ , respectively. Permeate had  
8 solubilized methane. The study also reported shear stress of biogas sparging on syntrophic anaerobes  
9 coupled with methane forming. In the study of Xie et al. (2014), raw leachate, high heavy metal  
10 concentration, and high total ammonium concentration  $>3000 \text{ mg/L}$  were treated in AnDMBR using  
11 Dacron mesh of pore size  $40 \mu\text{m}$  and achieved methane production of  $0.34 \text{ LCH}_4/\text{gCOD}_{\text{removed}}$  ( $0.30$   
12  $\text{LCH}_4(\text{STP})/\text{gCOD}_{\text{removed}}$ ). Archeal taxonomic identified more activity of acetoclastic methanogens  
13 functional group than hydrogenotrophic methanogens that were removed from system when  
14 ammonium inhibition reported (Xie et al., 2014).

#### 15 *5.4 Anaerobic electrochemical membrane bioreactor (AnEMBR)*

16 Recently, development of multifunctional conductive, catalytic, porous, hollow-fiber cathodes  
17 (CCPHF) in anaerobic bio-electrochemical membrane reactor (AnEMBR) has gained promising  
18 attention and presented in Fig. 6e (Katuri et al., 2018; Katuri et al., 2014; Werner et al., 2016). Katuri et  
19 al. (2014) recommended a novel configuration of a microbial electrolysis cell (MEC) with anaerobic  
20 filtration that incorporated porous, conductive, nickel-based hollow fiber membranes (Ni-HFM). This  
21 configuration recovered energy as biogas from low organic strength wastewater ( $300 \text{ mg/L}$  of COD).  
22 The benefit of Ni-based porous conductive membrane was recovery of energy and treated water in  
23 single stage. The problem of low specific surface area of cathode was also defeated by increasing  
24 packing density of cathode (Katuri et al., 2014). The other problem in this technology was membrane  
25 fouling. Therefore to become more attractive and advantageous choice for low organic strength



1 wastewater treatment, efficiency of energy recovery and fouling of AnEMBR system are to be under  
2 consideration. The performance of MECs is improved by applied voltage. Hydrogen evolution rate and  
3 energy balance can be affected by applied voltage (Ding et al., 2018). The hydrogen evolution rate  
4 increases as applied voltage increases and as rate of gas bubble formation increases at membrane  
5 surface, the scouring action increases and membrane fouling decreases (Pant et al., 2010).

6 Membrane fouling in bioelectrochemical system also reduces by using fluidized bed membrane  
7 bioelectrochemical reactor in which granular activated carbon is used as scouring media as shown in  
8 Fig. 6f (Cusick et al., 2014; Li et al., 2014; Li et al., 2016). The contaminants from wastewater or  
9 synthetic solution were removed with electricity generation in MBER during 150 days operation (Li et  
10 al., 2014). The granular activated carbon minimized transmembrane pressure (TMP) and its work as  
11 anode was minor. When this system (MBER) coupled with regular microbial fuel cell (MFC), energy  
12 recovery and contaminants removal occurred mostly in MFC section. Fluidized media aided in  
13 removing inorganic scaling on cathode surface in MEC (Cusick et al., 2014). The MFCs and MECs  
14 alone are not able to produce high quality effluent that is required for water reuse applications.  
15 Different studies have investigated the system of MFCs coupled with Anaerobic membrane bioreactors  
16 to improve effluent quality as shown in Fig. 6g (Li et al., 2014; Li et al., 2016; Ren et al., 2014; Tian et  
17 al., 2014; Wang et al., 2013). Malaeb et al. (2013) examined a hybrid system of MFC-MBR that  
18 consolidated with ultrafiltration, flat-sheet, conductive bio-cathode membrane. This porous bio-  
19 cathode membrane provided benefits of filtration for effluent and cathode for reduction reaction. But  
20 this type of membrane has low specific surface area means to say low cathode surface area per reactor  
21 volume (Malaeb et al., 2013a). Microbial fuel cells have advantage of low cost but they are of low  
22 effluent quality. (Logan, 2010; Rabaey & Rozendal, 2010). Effluent quality can be improved by  
23 inserting membrane filtration in MFCs. Wang et al. (2011) reported new technology of bioreactor  
24 namely, anaerobic membrane bioelectrochemical reactor in which membrane worked as cathode and  
25 filter media in MFC. The advantage of this process was high effluent quality and large bio-energy

1 production. The membrane fouling would be minimized due to anode oxidation of substrates that has  
2 low organic loading during membrane filtration (Wang et al., 2011).

### 3 *5.5 Anaerobic osmotic membrane bioreactors (AnOMBRs)*

4 AnOMBR is a new configuration for wastewater treatment integrating forward osmosis (FO)  
5 and AnMBR. FO membrane allows water to pass through semi-permeable membrane from feed  
6 solution having lower osmotic pressure to draw solution with higher osmotic pressure in AnOMBR  
7 (Chen et al., 2016; Gu et al., 2015). This process does not require any energy input to run filtration  
8 process in comparison to traditional pressure AnMBRs. Gu et al. (2015) treated low strength  
9 wastewater treatment in AnOMBR for recovering of energy in form of biogas at mesophilic  
10 temperature. A significant amount of methane was achieved in the range of  
11 0.25-0.3 LCH<sub>4</sub>/gCOD<sub>Removed</sub> (Gu et al., 2015). Chen et al. (2014) investigated methane recovery ability  
12 of FO-AnMBR system and found that methane of 0.21 LCH<sub>4</sub>/gCOD<sub>Removed</sub> produced with 65 to 78%  
13 methane content. Though, the yield of methane was 58% of theoretical yield due to dissolved methane  
14 in effluent and low methanogenic activity under salinity environment (Chen et al., 2014). Therefore,  
15 future research is needed to investigate the activity of methanogens and microbial kinetics to enhance  
16 biogas generation under salinity conditions.

### 17 *5.6 Anaerobic bio-entrapped membrane reactors (AnBEMRs)*

18 AnBEMRs have now become an emerging technique to overcome conventional AnMBRs  
19 especially for high concentration of biomass. The major advantage of this technique was simultaneous  
20 deduction of nitrogen and carbon within a single bioprocess. It had enough capability to treat higher  
21 dissolved and complex organics (Ng et al., 2014). Because of its ability to produce low soluble and  
22 suspended biomass, so membrane fouling can be effectively minimized in integrated entrapped  
23 bioprocess with membrane (Chen et al., 2016). Ng et al. (2014) investigated efficiency of lab-scale  
24 AnBEMR in biogas generation during treatment of pharmaceutical wastewater and 15% more biogas  
25 was produced in AnBEMR (0.159±0.035 mLCH<sub>4</sub>/gCOD<sub>Removed</sub>) than conventional AnMBR

1 (0.142±0.034 mLCH<sub>4</sub>/g COD<sub>Removed</sub>) after starting period of 70 days. However, production of methane  
2 was inhibited by high salinity conditions, over organic loading, and buildup of toxic organics in both  
3 systems at high OLRs (34±2.7 kgCODm<sup>-3</sup>.d<sup>-1</sup>). Moreover, AnBEMR had longer period of membrane  
4 operation than that of conventional AnMBR due to small concentrations of SMP, EPS, and suspended  
5 biomass (Ng et al., 2014). Low-cost polyurethane sponge was also used for bacterial attachment,  
6 growth and bio-entrapment. Sponges were used as a feasible moveable shipper because of high  
7 durability and sponginess, which can retain microorganisms and remove nutrients and organics. Kim et  
8 al. (2014) examined performance of rotary disk membrane bioreactor with single and two stage  
9 submerged AnBEMR in the absence of membrane replacement and cleaning. The disk rotation boosted  
10 mass transfer and shear force caused by media and motivated collision between membrane external  
11 and sponge. Thus it improved membrane filterability and fouling in anaerobic rotating disk membrane  
12 bioreactor (Kim et al., 2014). The composition and yield of methane in single process were 13 and  
13 12%, which was greater than staged systems. So the single stage process was more efficient because of  
14 the higher energy potential (Chen et al., 2016). However, still there is a gap present corresponding with  
15 impacts of biocarriers/sponge shape, density and size, and shear force of disk rotation on activation of  
16 biomass and biogas yield and more research is needed.

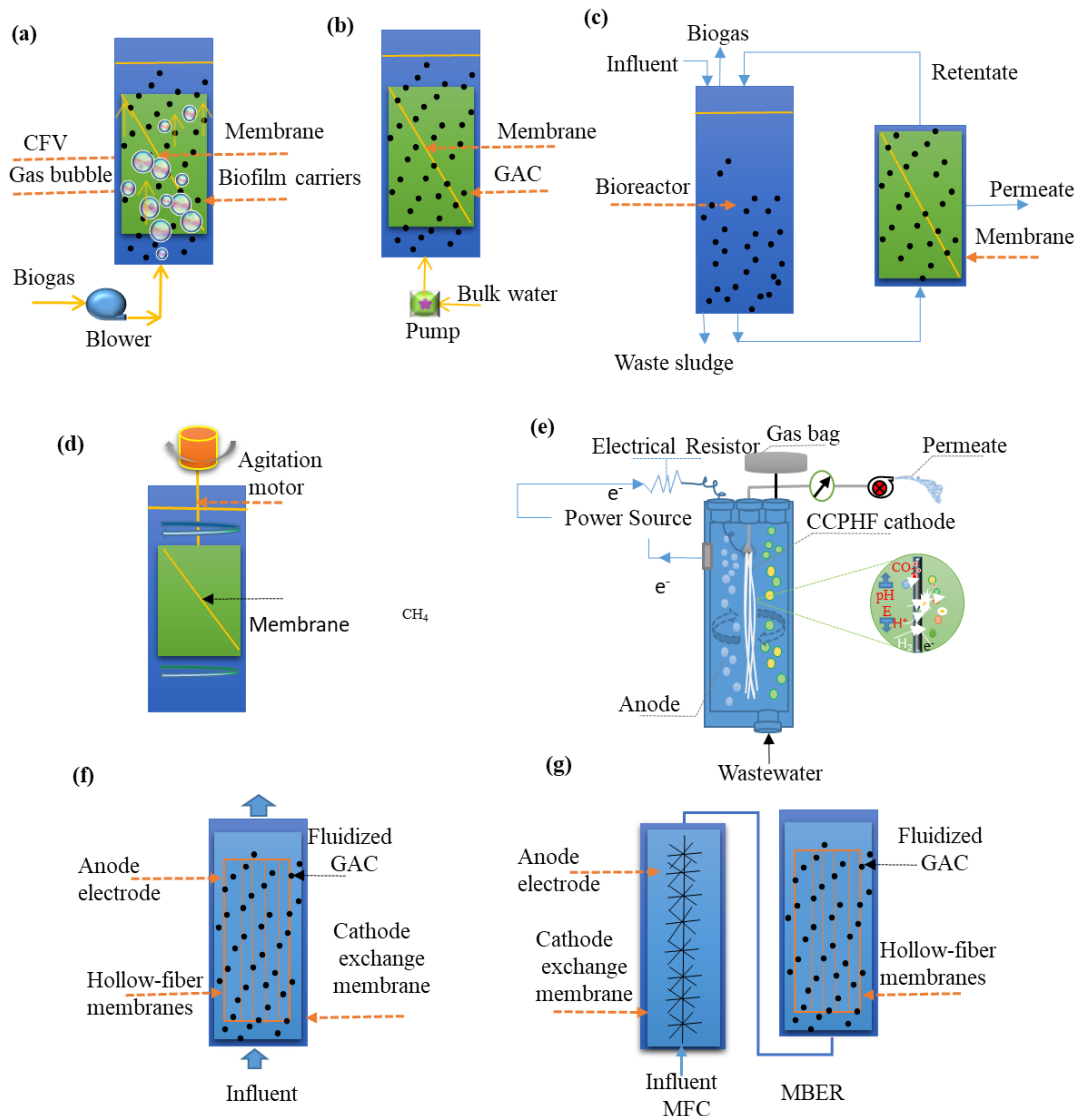
### 17 *5.7 Anaerobic membrane distillation bioreactors (AnMDBRs)*

18 Bioreactors that engaged with membrane distillation process operate at thermal difference  
19 phenomenon. Membrane is characterized as hydrophobic and microporous in process in which water  
20 vapors pass through membrane to form water on other side (Goh et al., 2015; Kim et al., 2015a).  
21 Coupling of anaerobic process with membrane distillation (MD) can be used as competitive  
22 technology for methane production as mesophilic or thermophilic operation (Kim et al., 2015a).  
23 AnMDBR is used as a biogas source which can be applied to mitigate membrane fouling through gas  
24 sparging technique. Spare biogas can be employed for heating purpose and energy applications (Goh et  
25 al., 2015). However, space is available for further treatment e.g. post-treatment to obtain dissolved

1 methane and ammonium nitrogen in permeate. It was revealed that dissolve methane was more in  
2 AnMBR as 30% and 50% at 35 and 15 °C, respectively (Smith et al., 2012). The reason behind this is  
3 inverse relation between temperature and solubility of methane gas which decreases with temperature  
4 increase. While methane gas remains exist in gas phase in AnMDBR and easier to extract and recover  
5 (Chen et al., 2016). Therefore, permeate obtains from AnMDBR has lower dissolved methane than  
6 from other AnMBRs (Goh et al., 2015). Xie et al. (2014) treated domestic wastewater in hybrid  
7 anaerobic MBR-MD system. Results demonstrated that system produced biogas with 58-72% contents  
8 of methane while no fraction of methane obtained from MD process (Xie et al., 2014). AnMDBRs  
9 overcome other systems in terms of small footprints, complete retention of organics and biomass for  
10 efficient conversion of organics into biogas (Goh et al., 2015; Kim et al., 2015a; Phattaranawik et al.,  
11 2008). AnMDBR has also strength for complete deletion of total phosphorous to control eutrophication  
12 (Kim et al., 2015a). However, further study is still needed with respect to biogas generation and  
13 challenges associated with AnMDBRs.

## 14 **6. Energy demand and recovery in AnMBRs**

15 The overall energy balance analysis was purposed by two aspects: energy that was obtained  
16 from methane production and energy consumption during operation of AnMBRs. Energy  
17 independency ratio is ratio of measured energy potential to total energy demand. Energy balance  
18 analysis predicts that net energy which can be recovered (Mei et al., 2016). Major part of energy in  
19 AnMBRs is consumed in fouling control in membrane tanks which is more than 70% of energy  
20 consumption of AnMBRs (Lin et al., 2013; Pretel et al., 2014; Smith et al., 2014). The energy that is  
21 used in fouling control mainly depends on fouling mitigation method applied. In contrast, the  
22 production of electrical energy can be obtained from combustion of produced methane (Kim et al.,  
23 2011a).



1

2 **Fig. 6.** Different configurations of AnMBR (a) Biogas-particle sparging; (b) Liquid recirculation  
 3 particle-sparging; (c) Anaerobic cross flow-particle sparging MBR; (d) Anaerobic rotating MBR (e) Anaerobic  
 4 electrochemical MBR; (f) Individual Anaerobic EMBR; (g) Hybrid MFC-AnEMBR system

5 *6.1. Energy demand*

6 Total energy demands in AnMBRs mainly consider two aspects: energy requirement in fouling  
 7 control and others. The energy demand in others include energy requirements for the rest of AnMBR  
 8 systems including energy consumption for operation of reactor, permeate pumps, and biogas/liquid  
 9 circulation in AnMBRs (Shin & Bae, 2018). Energy requirements associated with membrane operation  
 10 in AnMBRs consists of two operations: permeate pumping and fouling control. Membranes are  
 11 immersed in mixed liquor in submerged AnMBRs and gas is introduced below membrane module in

1 form of biogas. The other configuration consists of membrane module in outside bioreactor in side  
2 stream pumped cross flow system and mixed liquor is forced through pump to membrane module and  
3 recycled back to bioreactor. This system is operated at constant pressure with pump and performs two  
4 operations: liquid cross flow and driving force for permeation. Energy required for permeate flow can  
5 be obtained theoretically. Energy demands for fouling control can be further subdivided into two parts  
6 in systems: energy for pumping in cross flow and energy for biogas sparging in submerged  
7 configurations (Martin et al., 2011). Energy demands in submerged AnMBRs were calculated from  
8 0.03 to 5.7 kWhm<sup>-3</sup>. While in side stream AnMBRs, energy demand was observed from 0.23 to 16.52  
9 kWhm<sup>-3</sup> due to cross flow velocity and bioreactor MLSS on flux and pressure losses (Martin et al.,  
10 2011). As strength of wastewater increases from 0.24 to 10 gCODL<sup>-1</sup>, energy obtained from biogas  
11 also increases ranges from 0.62 to 34.8 kWhm<sup>-3</sup> (Martin et al., 2011). Energy demands in different  
12 configurations are summarized in Table 1.

### 13 6.2. Energy recovery

14 Biogas produced from AnMBR has usually composition of: 50-90% methane, 3-15% carbon  
15 dioxide, and 0-15% nitrogen (Lin et al., 2013). Methane rich biogas may have demand in digester  
16 heating, electricity generation, or recycle for fuel generation. It was investigated that 2.02  
17 kWh/kgCOD<sub>Removed</sub> obtained in AnMBR treating synthetic wastewater (Lin et al., 2013). Methane  
18 production is significantly affected by temperature variation. Methane production increases as  
19 temperature increases due to high specific growth and consumption rates of substrate (Lei et al., 2018;  
20 Lin et al., 2013). Anaerobic rotating membrane bioreactor has energy potential of 0.92 kWh/m<sup>3</sup> packed  
21 with hollow fiber membrane (Lei et al., 2018; Shin & Bae, 2018). Methane production in anaerobic  
22 bioreactor with submerged forward osmosis membrane recorded as 0.21 LCH<sub>4</sub>/gCOD (Chen et al.,  
23 2014). Methane production in upflow anaerobic sludge blanket (UASB) reactor was observed in ranges  
24 from 0.16 to 0.2 LCH<sub>4</sub>/gCOD (Chen et al., 2014). Energy production from municipal organic waste  
25 treated through anaerobic fermentation process was recorded as 4.54 kWh/kgCOD (Chen et al.,

1 2017d). Methane generation potential in AnDMBR was reported in range from 0.28 to 0.31 LCH<sub>4</sub>/g  
 2 COD<sub>Removed</sub> (Ersahin et al., 2016). Anammox-AnMBR produced methane of 0.223 LCH<sub>4</sub>/gCOD<sub>Removed</sub>  
 3 (Chen et al., 2016). AnOMBR was found to have methane production potential ranges from 0.25 to 0.3  
 4 L CH<sub>4</sub>/g COD<sub>Removed</sub> (Chen et al., 2016). Table 2 summarizes the methane production and system  
 5 performance of different AnMBRs.

6 Energy potential in AnMBRs for wastewater treatment is mainly affected by influent sulfate  
 7 concentration. Energy potential in biogas sparging AnMBRs recorded from 0.08 to 0.50 kWhm<sup>-3</sup> and  
 8 0.14 kWhm<sup>-3</sup> for particle sparging bioreactor (Lei et al., 2018; Shin & Bae, 2018). Energy production  
 9 in MBER was recorded about 0.011 kWhm<sup>-3</sup>. The coupled MFC plus MBER system recovered total  
 10 energy of 0.047 kWhm<sup>-3</sup> (Li et al., 2014). It was also observed that in AFMBRs with bulk liquid  
 11 recirculation, energy requirement was sufficiently low than energy produced from methane as 0.23  
 12 kWhm<sup>-3</sup> (Aslam et al., 2017b). Energy potential can be increased by using staged-anaerobic fluidized  
 13 bed membrane bioreactor (SAF-MBR) for treatment of effluents generated by anaerobic fluidized bed  
 14 bioreactor (AFBR). Net energy produced from this system was investigated about 0.238 kWhm<sup>-3</sup>. This  
 15 was 9.8 times more than energy required to operate SAF-MBR system (Aslam et al., 2018c).

16 **Table 1.** Energy consumption with different configurations

AnMBR configuration	Energy consumption (kWh/m <sup>3</sup> )	References
Cross flow AnMBR	3-7.4	(Aslam et al., 2017a; Lei et al., 2018)
Particle-sparging cross flow AnMBR	0.02-0.05	(Seib et al., 2016)
Biogas-sparging submerged AnMBR	0.25-3.41	(Lei et al., 2018; Liao et al., 2006; Martin et al., 2011)
Biogas particle-sparging AnMBR	0.01-1.0	(Aslam et al., 2017a; Lei et al., 2018)
Liquid circulation particle-sparging submerged AnMBR	0.0087-0.23	(Shin & Bae, 2018) (Aslam et al., 2018c; Kim et al., 2011b; Shin et al., 2014)
Rotating membrane submerged AnMBR	0.23	(Shin & Bae, 2018)
Anaerobic Membrane bioelectrochemical reactor (AnMBER)	0.043-0.09	(Li et al., 2014; Li et al., 2016)
Hybrid MFC-MBER system	0.0186-0.046	(Ren et al., 2014)

1 In anaerobic process, biodegradation occurs in absence of oxygen or nitrate as electron  
2 acceptor. The final products are mainly methane and carbon dioxide. Here methane can be recovered  
3 as biogas or dissolved in effluent. Energy production in AnMBRs is associated with biogas production  
4 mainly contains methane. The highest methane yield observed ranges from 0.29 to 0.33 LCH<sub>4</sub>/gCOD  
5 when treating low strength wastewater (Hu & Stuckey, 2007). Lowest methane yield also reported as  
6 0.12 and 0.08-0.09 LCH<sub>4</sub> g<sup>-1</sup> COD when operated at 11-25 °C and 25-30 °C, respectively (Chu et al.,  
7 2005). These values indicate significance of temperature in production and recovery of biogas. The  
8 energy production potential is significantly affected by influent COD concentrations and SO<sub>4</sub><sup>2-</sup>S  
9 particularly for low-strength wastewater having COD lower than 300 mg/L. Recently, seven pilot scale  
10 AnMBRs were found to be energy positive or energy neutral, suggesting that AnMBRs not only  
11 accomplish electrical energy balance but also achieve energy output (Lei et al., 2018).

### 12 *6.3. Strategies to reduce energy expenditures*

13 Energy consumption and related cost are both connected to membrane fouling issues. The  
14 major focusing points in energy reduction are associated with module configuration, control systems,  
15 low energy membrane cleaning methods, or other fouling control methods such as membrane  
16 vibration, scouring agent with granular medium, electric field, and others (Meng et al., 2017). Many  
17 researchers suggested energy efficient AnMBRs in the form of optimal flow conditions and hydraulic  
18 capacity utilization of membranes. Since high energy consumption is associated with operation run at  
19 below optimal flow conditions (design flow rate is approximately equal to the hydraulic load)  
20 (Krzeminski et al., 2017). The major aspects that correspond to energy reduction are: Combined effect  
21 of number of membrane modules, interval of filtration and relaxation/backwash, use of flux enhancer,  
22 pump configuration and recirculation rate (Krzeminski et al., 2017). Moreover, operating condition  
23 that is lower than critical flux is successive turn to achieve hydraulic and energy efficiency. As  
24 membrane flux has direct link to the hydraulic utilization of membrane, so it plays a positive effect in  
25 energy efficient system. Palmowski et al. suggested energy efficient system in the form of increasing



1 filtration at high flux in limited/short time interval. In another approach, intermittent electric field is  
2 also used as fouling control for energy saving purpose. Electric field process may consist of  
3 electrophoresis, electro-coagulation in which sludge size increases and zeta potential reduces, and  
4 electrostatic repulsion/rejection against particles (Meng et al., 2017).

5 As aforementioned, energy consumption in AnMBRs can be minimized by implementation of  
6 higher flux. This target can be achieved by using high permeable membrane and introducing periodic  
7 chemical cleanings. Chemical cleaning is the most feasible and reliable approach to overcome  
8 membrane fouling. Aslam et al. (2017) compared flux rate of two different configured AnMBRs with  
9 particle sparging. Hydrophilic ceramic membrane inserted in first reactor with periodic MCs. The other  
10 one had hydrophilic polyvinylidene fluoride (PVDF) with no MCs. Results declared that flux rate was  
11 1.7 times higher ( $17 \text{ Lm}^{-2}\text{h}^{-1}$ ) in first reactor (Aslam et al., 2017b). The selection of optimal MLSS  
12 conditions can also reduce energy expenditure. This is done by maintaining concentration of MLSS at  
13 low level in membrane tank. It causes reduction of specific gas demand ( $\text{SGD}_m$ ). Robles et al. (2012)  
14 showed that as MLSS concentration increased from 23 to 28 g/L,  $\text{SGD}_m$  also increased from 0.25 to  
15  $0.5 \text{ Nm}^3\text{m}^{-2}\text{h}^{-1}$  at critical flux of  $14 \text{ Lm}^{-2}\text{h}^{-1}$  in anaerobic gas sparging hollow fiber membrane  
16 bioreactor. Pretel et al. (2015) reported the path of usage of primary clarifier to reduce MLSS  
17 concentration from 10-18 g/L to 5-10 g/L in membrane tank. Low MLSS concentration requires low  
18 energy demand in AnMBRs but will be engaged with increase in sludge production (Pretel et al., 2015;  
19 Robles et al., 2012). Therefore, more explanatory and detail work is required on this approach. The  
20 implementation of coagulant or fluidizing GAC can also reduce energy consumption. The drawback of  
21 particle sparging AnMBR is reduced lifespan of membrane by scouring action of particles on  
22 membrane surface and need for replacement of membrane (Shin et al., 2016a). The use of coagulant as  
23  $\text{FeCl}_3$  also caused replacement of membrane twice for 536 days where TMP increased with  
24 enhancement of operating flux and resulted in irreversible fouling (Dong et al., 2015). Further research  
25 is needed as frequent replacement of membrane increases economic aspects.

1 **Table 2.** Summary of methane production and system performance of AnMBRs

Scale	Wastewater	configuration	Reactor capacity (L)	Membrane characteristics	Temperature (°C)	HRT (h)	Influent COD (mg/L)	Methane yield (LCH <sub>4</sub> /gCOD <sub>Removed</sub> )	Methane content/ conversion rate	COD removal (%)	Reference
Pilot	Synthetic molasses wastewater	External	72	Silver coated PVDF, PES 0.22µm	35 - 37	30	2400	0.31	70 - 75%	82	(Amouamouha & Gholikandi, 2018)
Lab	Synthetic municipal wastewater	Submerged	6	FS PVDF 0.20µ m	25	8	670 ± 100	0.326	93	-	(Chen et al., 2017b; Chen et al., 2017c)
Lab	Synthetic food wastewater	Submerged	21.6	FS 0.4µm	35 ± 1	20-120 d	3000 - 27000	5 – 50 L/d	49-67%	94±8	(Casu et al., 2012)
Lab	Synthetic wastewater	Submerged	4	FS, CTA 0.025m <sup>2</sup>	25	15 - 40	460	-	65-78%	96%	(Chen et al., 2014)
lab	Synthetic municipal wastewater	External	4	HF, PVDF 0.22µm	20 ± 0.5	12	330 - 370	0.16±0.006	45.3%	91.9±1.5	(Chen et al., 2017a)
Lab	Pulping whitewater landfill leachate	Submerged	10	FS, PVDF 70kDa	37	-	2.78 - 3.35	-	68%	90	(Gao et al., 2010)
Lab	Synthetic wastewater	Submerged	7.4	FS	35.7	10 d	20,100	2.4 L/d	-	99.5	(Ersahin et al., 2017)
Lab	Synthetic municipal wastewater	submerged	6	FS, PVC 0.2µm	25±1	12	492±112	0.30	86.9	97.07	(Nie et al., 2017b)
Lab	Food waste	External	15	HF, PTFE 0.2µm	-	5-30 d	73610 ± 3100	-	58-61	80.4-92.9	(Cheng et al., 2018)
Pilot	Municipal wastewater	External submerged	310	HFM 0.045µm	6-30	10-13.4	892±271	0.235	67.1	-	(Gouveia et al., 2015)
Lab	Food wastewater	Submerged	5	HF 0.04µm	24 ± 2	24	1463	2.8 ± 0.2 L/d	38.4	87.50	(Galib et al., 2016)
Pilot	Domestic wastewater	External	50	UF 100kDa	37	15 - 60	685	30 L/d	70%	55 - 90	(Saddoud et al., 2007)
Lab	Synthetic municipal solid waste	Submerged	3	FS, PE 0.4µm	35	0.37 - 5.7	4000 - 26000	-	1-71%	93 - 94.5	(Trzcinski & Stuckey, 2009)
Lab	Domestic wastewater	Submerged	3.6	FS, Ceramic 0.2µ m	25 - 30	7.5	330.4 ± 89.8	0.3±0.001	85.70%	-	(Yue et al., 2015)
Lab	Sewage	Submerged	6	FS, PE 0.2µm	20-25	6-48	492±112	2.11-2.85 L/day	32-77	80-97	(Nie et al., 2017a)
Lab	Synthetic wastewater	External	0.7+0.06	FS	20-24	0.25-5.7	900	0-0.2	-	>89	(Alibardi et al., 2016)

FS: Flat Sheet, HF: Hollow fiber, CTA: Cellulose triacetate, Lab: Laboratory scale, PE: Polyethylene, PES: Polyethylene sulfone, PET: Polyethylene terephthalate, PTFE: Polytetrafluoroethylene, PVC: Polyvinylchloride, PVDF: Polyvinylidene fluoride, UF: Ultrafiltration

## 1 **7 Future perspectives**

2 Over last decade, many researchers have focused on applying new and novel technologies to  
3 mitigate membrane fouling in AnMBRs. Cell entrapment (CE), nano-materials (NMs), biological  
4 concepts, fluidized media and electrically based strategies are promising to reduce membrane fouling.  
5 The practicality of these technologies on the larger scale needs to be assessed to identify further  
6 research requirements. Current practices to prevent cake layer formation over the membrane surface  
7 involve biogas sparging with carriers addition, liquid recirculation with particle sparging while pore  
8 blockage is avoided by chemical cleaning. The results showed a great performance without affecting  
9 the biological activity in AnMBRs. Furthermore, membrane fouling can be reduced by using biomass  
10 entrapment technology that has low SMP and EPS in mixed liquor and large particle size of sludge  
11 (Juntawang et al., 2017). Application of antifouling membranes could be another way to mitigate  
12 membrane fouling. In-situ cleaning of membrane surface by the pretreatment of feed with suitable  
13 coagulants/adsorbents and particle sparging is also recommended. However, sustainability in  
14 membrane fouling mitigation can be achieved by integrating several approaches to get an optimal  
15 control of foulants.

16 Dissolved methane recovery in AnMBRs is another issue that offset their economics. The  
17 recovery of dissolved CH<sub>4</sub> is strongly associated with the overall treatment cost of wastewater using  
18 AnMBR. One way to reduce dissolved CH<sub>4</sub> is to operate AnMBR at high temperatures. However, the  
19 optimum temperature for methanogens is 37 °C. Above this temperature the microbial activity will be  
20 disturbed. The operational cost (and/or biogas production) of AnMBR could be reduced by operating  
21 the bioreactor at low ambient temperature. However, the methane solubility increases at lower  
22 temperature, which in turn increases the cost of recovery. Furthermore, low ambient temperature does  
23 not favor maximum microbial activity. Hence, it is strongly desired to develop a methane recovery  
24 method that enables economical methane recovery at minimum cost. Crone et al. (2016) employed  
25 vacuum membrane contactor which only requires 0.009 kWhm<sup>-3</sup> of energy. Dissolved methane content

1 can also be controlled by the combination of gas stripping and post-processing (Anammox) technology  
2 in which nutrient removal occurs simultaneously. During this process, denitrification of methane  
3 oxidizing bacteria occurs through the residual dissolved methane. This does not only reduce dissolved  
4 methane in effluent but is also useful in removal of nitrate and nitrogen that is generated in the  
5 Anammox process. To overcome the disadvantages of mechanical and chemical cleaning, biological  
6 methods are introduced. These methods improve the effluent quality as well as biogas production.

7 Many researchers are trying in the development of multifunctional conductive, catalytic,  
8 porous, hollow-fiber cathodes (CCPHF) that is more efficient in recovery of resources from  
9 wastewater. However, there is challenging environment to revolutionize polymeric hollow fiber  
10 membrane into CCPHF cathodes by using ALD (atomic layer deposition) or others traditional coating  
11 methods at large scale due to low chemical, thermal, and mechanical stability of polymeric hollow  
12 fiber membranes. Fabrication of CCPHF with ceramic hollow fiber membrane could be an alternative  
13 route by using different surface modification techniques because these membranes have high chemical,  
14 thermal, and mechanical stability. During fabrication it is important to notice that surface of ceramic  
15 based CCPHF cathodes should be biocompatible for MES applications or for anaerobic  
16 electrochemical membrane bioreactor (AnEMBR) applications. In contrast, Ni, Co, and Fe could be  
17 more efficient, abundant, and non-expensive metal catalysts that can be employed as thin film coating  
18 on ceramic hollow fiber membranes. The efficiency of CCPHF can be improved by using easy  
19 synthesis, cost effective 3D porous carbon nanotube (CNT) for AnBEMR/MES systems. Further  
20 improvement in the efficiency of CCPHF could be obtained by coupling CNTs with Fe, CO, Ni, and  
21 their composite.

22 AnMBRs outperform in several applications to treat domestic and industrial wastewaters under  
23 extreme operating conditions. An optimal solution to address issues of membrane fouling, inefficient  
24 dissolved methane recovery, poor nutrients removal, and removal of undesired compounds can be  
25 realized by using novel AnMBR configurations. Biological approaches, such as quorum quenching,

1 need to be explored for effective and energy efficient fouling mitigation in AnMBRs. Furthermore, the  
2 following important constraints must be addressed for further improvements to comply the reliability  
3 and commercial status of AnMBR technology:

- 4 • Operational optimization for minimizing energy consumption
- 5 • Energy positivity and/or energy neutrality to achieve significant market volumes by  
6 improving energy production potential
- 7 • Improvements in the membrane configurations using sole design optimization aiming to the  
8 efficient control of AnMBR performance.
- 9 • To explore the new cost-effective mineral based composite membranes with variety of  
10 improved intrinsic characteristics to withstand particle sparging and chemical cleaning  
11 protocols.

## 12 **8 Conclusions**

13 AnMBR performance is greatly influenced by temperature, pH, HRT and others parameters. It  
14 is promising due to energy saving, high COD removal, methane production, and low sludge yield.  
15 However, there are also some challenging aspects in the form of fouling and its association with  
16 energy expenditures, dissolved methane recovery, sulfide, pH and alkalinity, ammonia and long chain  
17 fatty acids. Different new configurations have been developed to make this strategy more energy-  
18 efficient and can enhance to commercial level. Control of membrane fouling and dissolved methane  
19 recovery play an important role to make AnMBRs technology an energy efficient system and require  
20 further study.

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