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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**

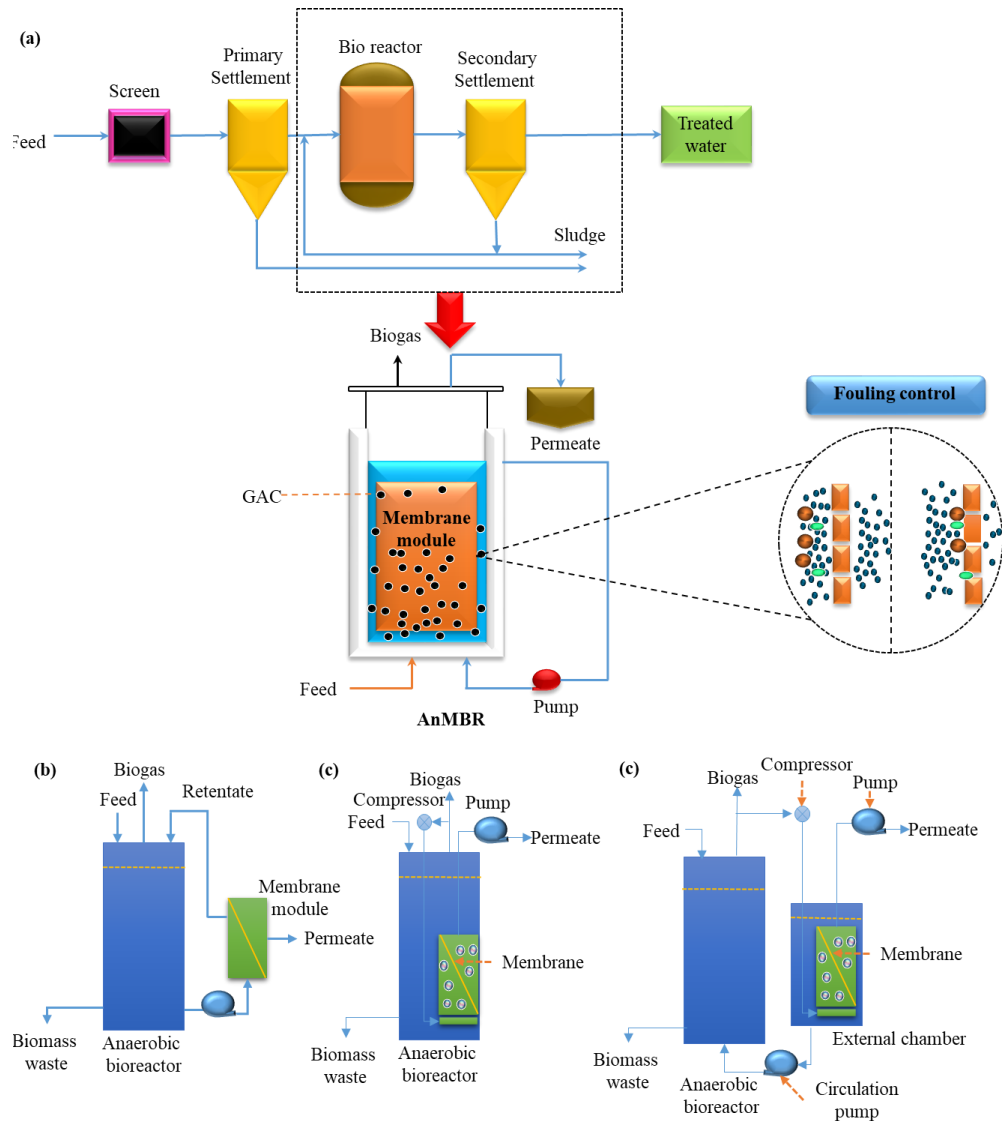
2 The scarcity of water resources and environmental consequences caused by the addition of
3 wastewaters into main water bodies and soil has triggered the need of treating wastewater. Wastewater
4 is considered as additional sources of water for domestic, agricultural and industrial use (McCarty et
5 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 (CH₄), carbon dioxide (CO₂), and nitrogen oxide (N₂O) 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 CH₄ 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 CH₄
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.

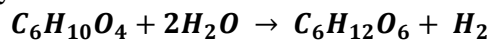
1 Membrane module is present outside reactor and permeate obtains under pressure provided by
2 recirculation pump in external cross flow configuration (Fig. 1b). In this approach, filtration cake on
3 membrane surface is disturbed by cross flow velocity (Skouteris et al., 2012). The drawback of this
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

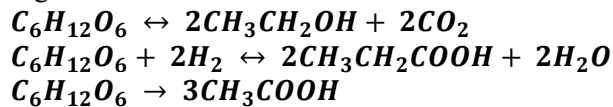
2 2.2.1. Hydrolysis

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):

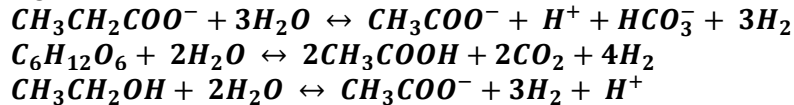
Hydrolysis



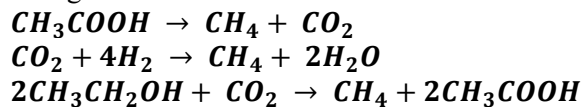
Acidogenesis



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.

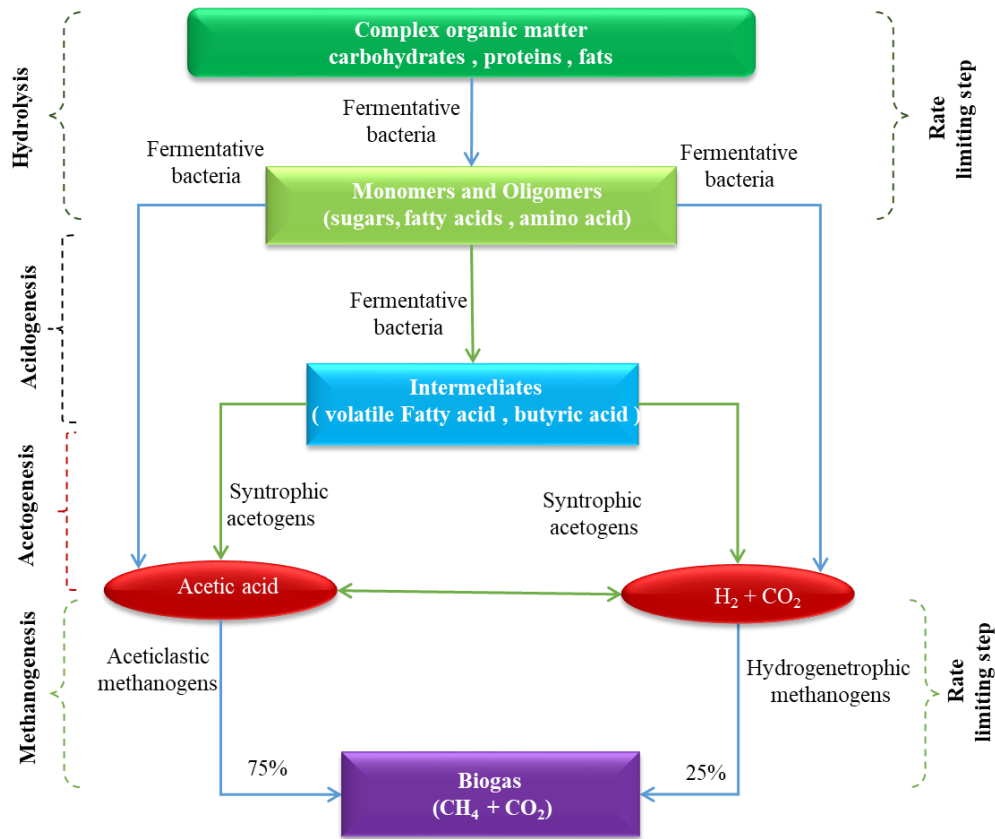
7 2.1.1 Acetogenesis

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 H₂ producing bacteria), and intake of H₂ and
12 CO₂ to produce acetate by homoacetogens (Ketheesan & Stuckey, 2015). Homoacetogens also
13 consume sugar and alcohols, generating acetate and H₂. 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.

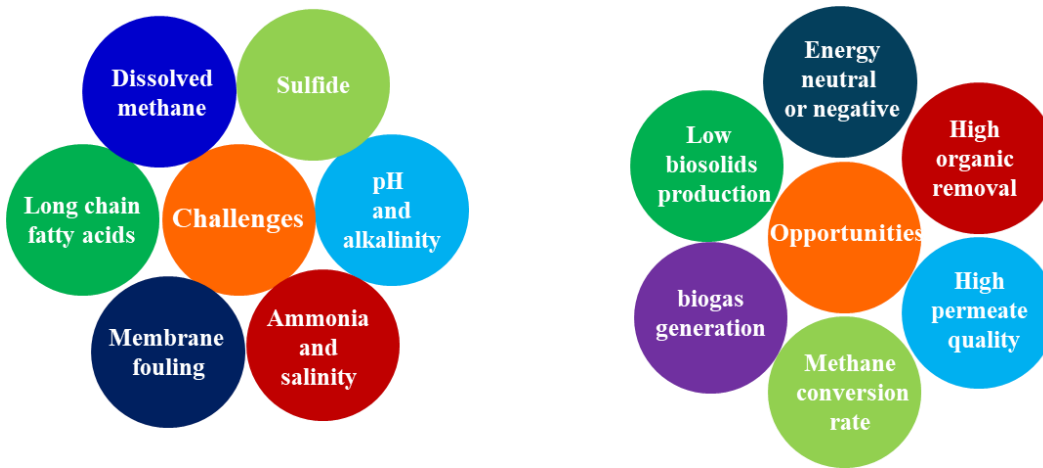


Fig. 3. Challenges and opportunities in AnMBRs

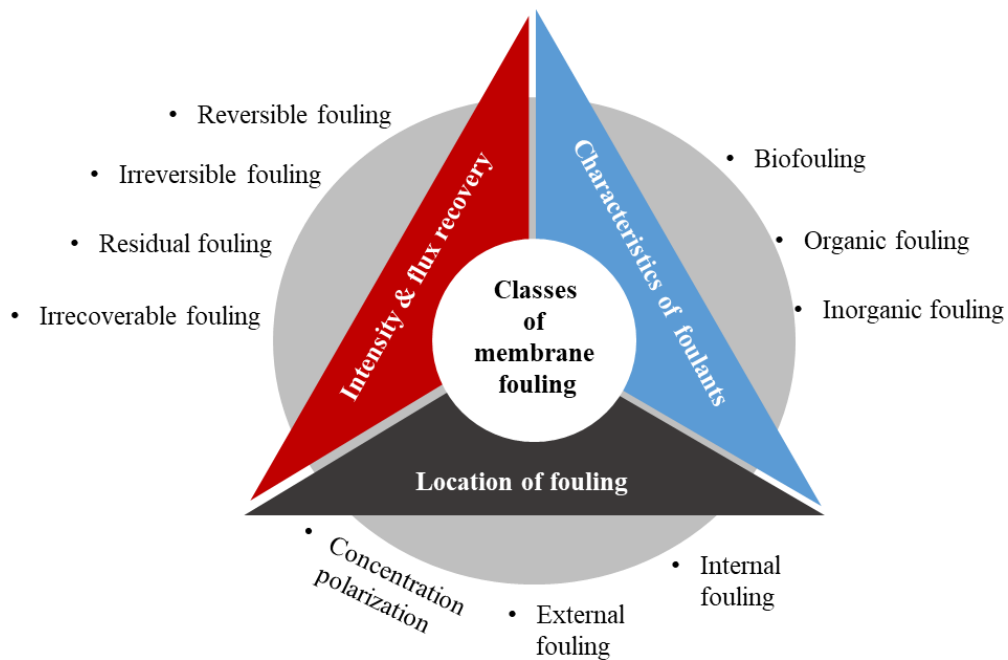
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External fouling occurs by the deposition of particles, colloids, and macromolecules with larger than pore sizes of membrane. It is divided into two types on the basis of layer formation. First one is cake layer which leads to agglomeration of grasp 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 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 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 H₂S 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. H₂S is
5 produced as result of reduction of sulphate which is toxic, corrosive, and malodourous gas (Park et al.,
6 2014). H₂S 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, SO₄²⁻-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/SO₄²⁻-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/SO₄²⁻-
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/SO₄²⁻-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 CaCO₃/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 HCO₃⁻, NH₃, Ac⁻, S⁻², OH⁻, HS⁻, and
14 CO₃⁻². The major controlling parameters in methanogenic process are carbonic acid related system,
15 mainly HCO₃⁻, 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⁻¹ (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**

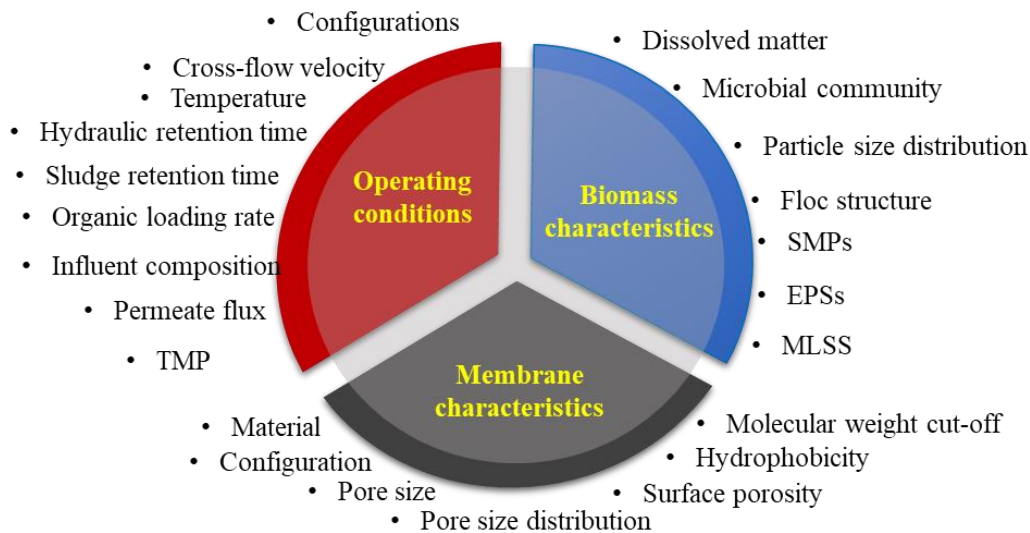
16 Fig. 5 presents key bottlenecks controlling membrane fouling in AnMBRs. Membrane fouling
17 can either be controlled by optimizing the operating conditions or surface modifications of membrane
18 or developing membranes less prone to fouling or controlling biomass properties.

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⁻³d⁻¹ (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⁻³d⁻¹ then biogas production rate also increased from 5 to 35 Ld⁻¹ 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 μm (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 μm and flocs size greater than 1 μm . 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 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 μm 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 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 LCH₄/gCOD_{Removed} (Gu et al., 2015). Chen et al. (2014) investigated methane recovery ability
12 of FO-AnMBR system and found that methane of 0.21 LCH₄/gCOD_{Removed} 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 mLCH₄/gCOD_{Removed}) than conventional AnMBR

1 (0.142±0.034 mLCH₄/g COD_{Removed}) 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⁻³.d⁻¹). 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.

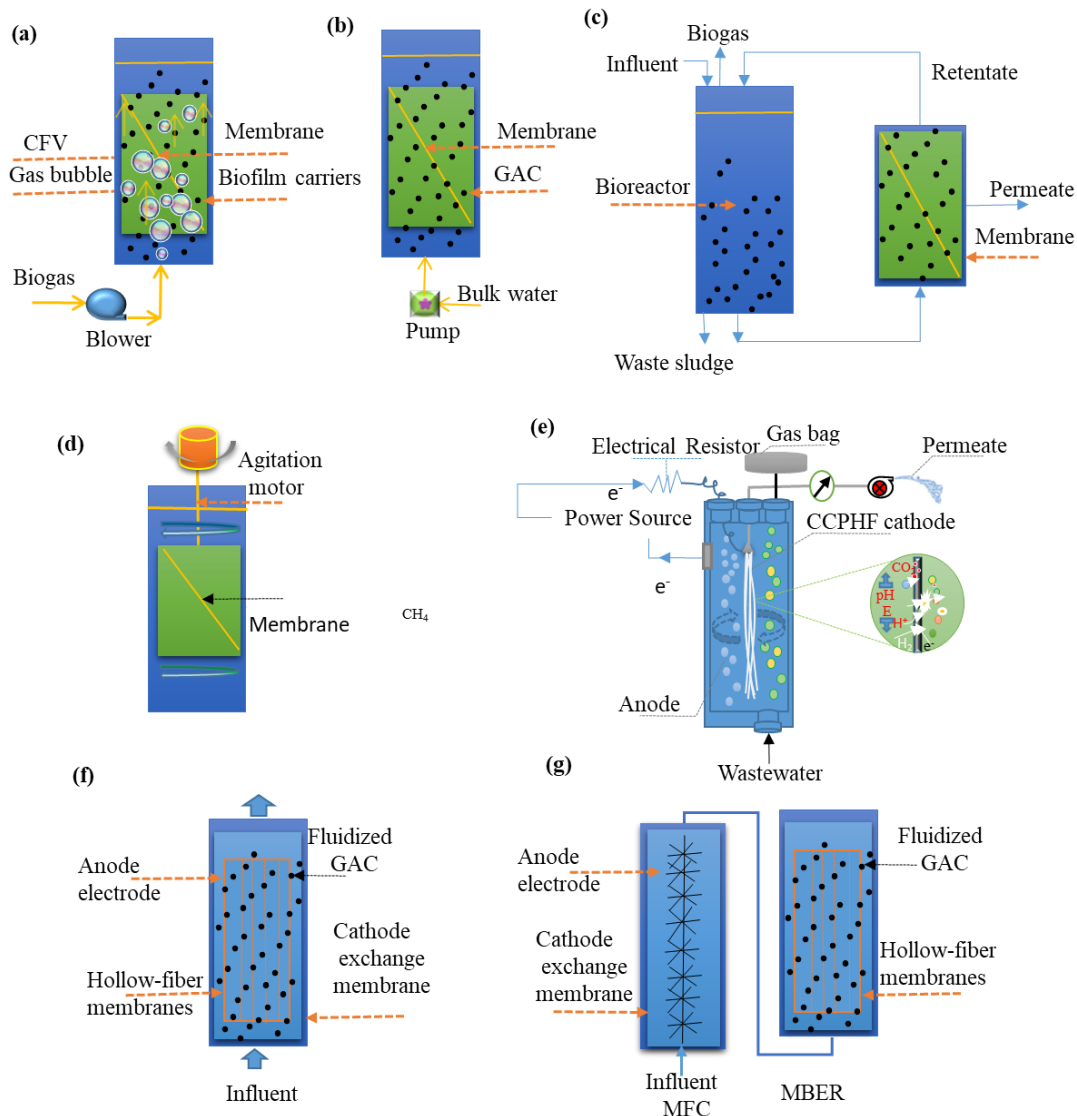
17 *5.7 Anaerobic membrane distillation bioreactors (AnMDBRs)*

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⁻³. While in side stream AnMBRs, energy demand was observed from 0.23 to 16.52
9 kWhm⁻³ 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⁻¹, energy obtained from biogas
11 also increases ranges from 0.62 to 34.8 kWhm⁻³ (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_{Removed} 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/m³ 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 LCH₄/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 LCH₄/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 LCH₄/g
 2 COD_{Removed} (Ersahin et al., 2016). Anammox-AnMBR produced methane of 0.223 LCH₄/gCOD_{Removed}
 3 (Chen et al., 2016). AnOMBR was found to have methane production potential ranges from 0.25 to 0.3
 4 L CH₄/g COD_{Removed} (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⁻³ and
 8 0.14 kWhm⁻³ for particle sparging bioreactor (Lei et al., 2018; Shin & Bae, 2018). Energy production
 9 in MBER was recorded about 0.011 kWhm⁻³. The coupled MFC plus MBER system recovered total
 10 energy of 0.047 kWhm⁻³ (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⁻³ (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⁻³. 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 ³)	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 LCH₄/gCOD
5 when treating low strength wastewater (Hu & Stuckey, 2007). Lowest methane yield also reported as
6 0.12 and 0.08-0.09 LCH₄ g⁻¹ 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 SO₄²⁻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 (SGD_m). Robles et al. (2012)
14 showed that as MLSS concentration increased from 23 to 28 g/L, 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 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 ₄ /gCOD _{Removed})	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 ²	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 CH₄ is strongly associated with the overall treatment cost of wastewater using
18 AnMBR. One way to reduce dissolved CH₄ 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⁻³ 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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24

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