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Fatima Anjum[1], Irfan M. Khan[1], Jeonghwan Kim[2], Muhammad Aslam[3], Gaetan Blandin[1,4], Marc Heran[1], Geoffroy Lesage[1].

[1]IEM, Univ Montpellier, CNRS, ENSCM, Montpellier, France

[2]Department of Environmental Engineering, Inha University, Incheon, Republic of Korea

[3]Department of Chemical Engineering, COMSATS University Islamabad (CUI), Lahore Campus, Defense Road, Off Raiwind Road, Lahore, Pakistan

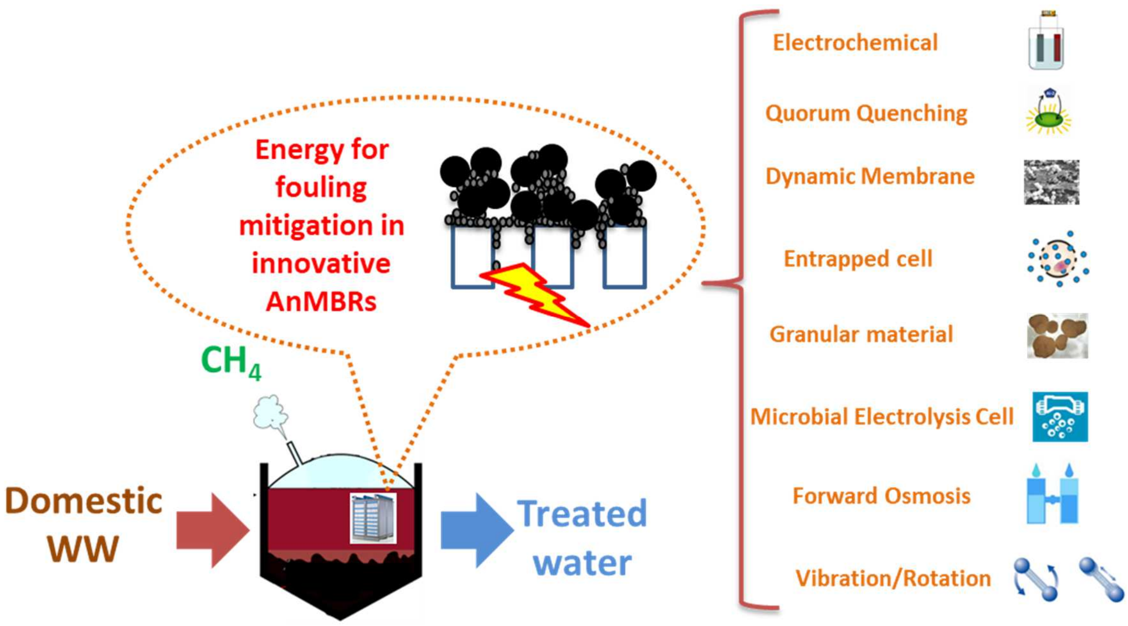
[4] Eurecat, Centre Tecnològic de Catalunya, Water, Air and Soil Unit, Manresa, Spain

*corresponding author: geoffroy.lesage@umontpellier.fr

Keywords

Anaerobic Membrane Bioreactor, Domestic wastewater treatment, Fouling, Hybrid processes, Biogas.

Graphical abstract



Abstract

Anaerobic membrane bioreactor has emerged as an innovative technology in treating domestic wastewater due to its excellent produced effluent quality and high potential of neutral or positive energy balance. One of the biggest challenges in positive energy objective is fouling mitigation which contributes towards 70% of the total energy requirement of MBR-based domestic wastewater treatment. Numerous studies were carried out to address this issue, utilizing various reactor design configurations and operating conditions for energy minimization as well as membrane performance enhancement. The latest research trend in this sector is the establishment of hybrid processes like Granular Anaerobic Membrane Bioreactors (G-AnMBR), Forward Osmosis Anaerobic Membrane Bioreactor (FO-AnMBR) and Microbial Electrolysis Cell - Anaerobic Membrane Bioreactor (MEC-AnMBR) for domestic wastewater treatment which not only provides efficiency in treatment but also improves fouling mitigation. Also, the application of techniques developed particularly for fouling mitigation like quorum quenching and sensing, cell entrapment and membrane module vibrations in AnMBRs were assessed. This paper reviews the latest trends in anaerobic membrane bioreactors research with regards to water quality produced, removal efficiencies and fouling mitigation.

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Table of Abbreviations

AeMBR: Aerobic membrane bioreactor
AHL: N-acryl homoserine lactone
AnDMBR: Anaerobic dynamic membrane bioreactor
AnEMR: Anaerobic electrochemical membrane bioreactor
AFMBR: Anaerobic fluidized membrane bioreactor
AnGS: Anaerobic granular sludge
AnMBR: Anaerobic membrane bioreactor
ATP: Adenosine triphosphate
BB: Building blocks
GSP: Biogas sparging
CFV: Crossflow velocity
COD: Chemical oxygen demand
CSTR: Continuous stirred tank reactor
DLVO: Derjaguin, Landau, Verwey, and Overbeek
DM: Dynamic membrane
DWW: Domestic wastewater
DWWT: Domestic wastewater treatment

EB: Empty beads
EG-AnMBR: Externally submerged granular AnMBR
IG-AnMBR: Internally submerged granular AnMBR
ES-AnMBR: Externally submerged membrane bioreactor
EPS: Extracellular polymeric substances
F/M ratio: Food to microorganism ratio
FO-AnMBR: Forward Osmosis Anaerobic Membrane Bioreactor
GAC: Granular activated carbon
G-AnMBR: Granular anaerobic membrane bioreactor
GHG: Greenhouse gases
GSIm: Biogas sparging intensity
GSP: Biogas sparging
HRT: Hydraulic residence time
LMH: Liter per square meter per hour
LMW: Low molecular weight
MF: Microfiltration
MEC: Microbial electrolysis cell
MEC-AnMBR: Microbial Electrolysis Cell - Anaerobic Membrane Bioreactor
MLSS: Mixed liquor suspended solids
MMV: Magnetically induced membrane vibration
MT: Membrane tank
MWh: Megawatt hour
P/C ratio: Protein to carbohydrate ratio
PSP: Particle sparging
PVDF: Polyvinylidene difluoride
QQ: Quorum quenching
QQB: Quorum quenching beads
RMEM: Rotating membrane
SCSA: Specific cathode surface area
SDG: Sustainable development goal
SG-AnMBR: Internally submerged granular AnMBR
SMP: Soluble microbial products
SRT: Solid retention time
TMP: Trans-membrane pressure
UASB: Up-flow anaerobic sludge blanket
UF: Ultrafiltration
VLR: Volumetric Loading Rate
VSEP: Vibratory shear enhancement process
WWTP: Wastewater Treatment Plants
WRRF: Water Resource Recovery Facilities

1. Introduction

In 2015, United Nations adopted the 2030 Agenda for Sustainable Development (SD) (UN 2019); SD Goal (SDG) 6 aims at ensuring availability and sustainable management of water and sanitation for all. To help address these issues, domestic wastewater (DWW) is thus now considered to be an alternative water (SDG 6), nutrients (SDG 2) and energy (SDG 7) source for various applications. Wastewater Treatment Plants (WWTPs) have even been rebranded as Water Resource Recovery Facilities (WRRFs) to enlighten the significant resource recovery potential that exists in DWW streams (Jain, 2018; Fulcher, 2014). The biological treatments such as aerobic and partial nitritation-anammox granular sludge are becoming increasingly popular in DWW treatment due to their high tolerance of the toxicity of feed, compactness, settling efficiencies and effectiveness in treating complex effluents with high organic load and low biodegradable compounds (Baeten et al., 2019; Taghipour, 2017; Taghipour and Ayati, 2017; Zhang et al., 2016). However, aerobic processes are highly energy-intensive and produce a large amount of sludge (Kim et al., 2011).

Recently, a change of paradigm was made from reliance on aerobic treatment towards anaerobic treatment which uses organic carbon from the DWW to produce energy in the form of methane gas and reduces the amount of sludge to be handled. However, anaerobic treatment alone may not be enough to meet stringent discharge limits and to hold anaerobic micro-organisms that have a very slow-growth rate. The introduction of membrane technology into anaerobic bioreactor is a promising solution to these issues.

Since more than twenty years, Anaerobic Membrane Bioreactors (AnMBRs) are in operation for the treatment of high strength WW i.e. industrial and agricultural WW (Amha et al., 2019; Bu et al., 2017; Thongmak et al., 2016). Kanai et al., (2010) have presented the successful demonstration of Kubota’s submerged anaerobic membrane biological reactor (a patented technology with 15 full-scale plants) at a spillage treatment plant in Japan. In addition to that its' applications include food waste and garbage treatment. The technology combines submerged membrane separation and anaerobic digestion in a single unit with a COD removal efficiency of up to 92% and an energy recovery of around 12 GJ/d. Christian et al., (2011) have reported the two years operation of a full-scale AnMBR treating food industry wastewater achieving 99% COD removal and resulting in up to 50% reduction in the total operating expenses of the wastewater treatment plant facility. For domestic wastewater treatment, no full-scale plant has been commissioned so far owing to the highly variable feed conditions and fouling issues. Robles et al., (2018) have underlined the need of further works on fouling behavior understanding, fouling mitigation and process control in AnMBRs for DWW treatment. New trends in circular economy calls for the utilization of AnMBR for domestic wastewater treatment since AnMBRs are able to:

- Retain anaerobic bacteria completely (Hydraulic and Solid Retention Time (HRT and SRT) are uncoupled in AnMBR) and work with higher loading capacity;
- Produce excellent permeate qualities (i.e. high removal of suspended solids, organic matter and microorganisms) thanks to microfiltration (MF) or ultrafiltration (UF) membranes (Lim et al., 2020);
- Keep the nutrients of the influent available for their recovery or direct reuse (e.g. struvite crystallization, microalgae cultivation, fertigation…) (Judd et al., 2015);
- Reduce Green House Gas (GHG) emissions by saving energy consumption: net energy production of 0.1 kWh/m^3 with AnMBR when aerobic treatment consumes approximately 0.25-0.6 kWh/m^3 of treated DWW (Pretel et al., 2016; McCarty et al., 2011).
- Reduce the amount of sludge to dispose of due to the lower growth yield of the anaerobic biomass (up to 90% reported by a study (Jeison et al., 2008));
- Reduce footprint due to higher compacity of AnMBR (15 kg$_{COD}$/m^3/d) compared to conventional activated sludge (CAS) systems (4 kg$_{COD}$/m^3/d).

Recent studies have shown the potential applicability of AnMBR for mainstream DWWT (Maaz et al., 2019; Galib et al., 2016; Pretel et al., 2016). Extensive work on AnMBR has been published at pilot scale to pave the path for its full-scale implementation (Shin and Bae, 2018). Nevertheless, membrane fouling remains one of the most challenging issues impeding the development of AnMBRs (Maaz et al., 2019; Saleem et al., 2016), especially with high free biomass concentration widely used in conventional AnMBRs.

Membrane fouling is an inevitable phenomenon and conventional AnMBRs mostly make use of biogas sparging (GSP) or crossflow velocity (CFV) to remove (or limit) fouling by promoting turbulences close to the membrane surface (Hu et al., 2007). Total energy requirements of AnMBR with biogas sparging were reported to be from 0.038 to 5.68 kWh/m³ (Martin et al., 2011) and crossflow velocity was reported to consume from 3 to 7.3 kWh/m³ (Aslam et al., 2017) depending upon CFV and mixed liquor suspended solids (MLSS) of the reactor. It has been reported that fouling mitigation could account for up to 70% of total energy consumption in AnMBRs (Shin and Bae, 2018). To design an energy-efficient fouling mitigation technique, many studies were carried out during the last decade understanding fouling behavior (Maaz et al., 2019; Charfi et al., 2017b; Chen et al., 2017a; Xiong et al., 2016; Ding et al., 2015; Chen et al., 2014; Charfi et al., 2012; Herrera-Robledo et al., 2011) and investigating various fouling control strategies (Robles et al., 2018; Shin et al., 2018; Krzeminski et al., 2017; Aslam et al., 2014). Fouling mechanisms in AeMBR (Aerobic Membrane Bioreactor) and AnMBR were compared and were found to be quite different. In AeMBR, adenosine triphosphate (ATP) concentration was 30-fold higher than in AnMBR which leads to higher microbial activity and results in a large fraction of extracellular polymeric substances (EPS) on the membrane surface. Thus, the main contributors to fouling in AeMBR arise from microbial activity during substrate biodegradation. In AnMBR, soluble microbial products (SMP) levels are up to 5 times higher and with higher molecular weights than in AeMBR (i.e; 2526 kDa and 180.1 kDa of carbohydrates detected in the SMP and 630-640 kDa and 0.9 kDa of protein MW detected in SMP for AnMBR and AeMBR respectively). This caused a higher rejection of proteins and carbohydrates in AnMBR which further deposit on the membrane surface thereby resulting in more severe organic fouling and biofouling in AnMBRs (Xiong et al., 2016). It has also been reported that inorganic fouling propensity is higher in AnMBR than AeMBR due to higher carbonate, bicarbonate, ammonia and phosphate concentrations in anaerobic reactors (Lin et al., 2013).

It has been established that the fouling in AnMBRs has remained one of the biggest hurdles in positive energy treatment of DWWT (Maaz et al., 2019). Thus, the focus of this review is, particularly on fouling. It aims to present the work that has been done so far in understanding and optimizing fouling mitigation in AnMBRs for domestic wastewater treatment, highlights the different potential technologies in this regard and provides researchers, working in this area, with information that can help in giving them directions for future research work.

The paper has been divided in to two major sections, the first will shed light on the current state of the art based on the return of experience and progresses of AnMBR at the pilot-scale for DWWT. The aim of this section is to inform the readers with the basics of fouling mechanism in the AnMBRs, the important definitions and concepts for the understanding of fouling behaviors, factors effecting and fouling control. It will also provide an overview of the latest research trends for fouling mitigation strategies in such systems. In the second section, emerging novel configurations like granular AnMBR (G-AnMBR), AnMBR combined with forward osmosis membrane (FO-AnMBR), entrapped cell-based AnMBR, dynamic AnMBR (AnDMBR) and microbial electrolysis cell AnMBR (MEC-AnMBR) will be discussed in term of removal efficiencies and fouling. These configurations are selected based on their contribution to fouling control. They were designed by researchers to provide solution to the issues of energy intensive fouling control and enhance membrane operation with both technical and economic optimization. Respecting the review's focus on fouling, other innovative techniques that are particularly developed to tackle fouling like quorum quenching and rotation/vibration application are also discussed. Some insight on methane production and potential energy generation are also provided.

2. Current status of operation and fouling in pilot-scale AnMBR

The majority of pilot-scale AnMBR systems were designed using commercially available membrane modules and were fed with real or synthetic domestic wastewaters to reflect seasonal and daily variations of the wastewater composition. Data acquired from various pilot-scale AnMBR experiments is crucial to upscale, design and develop full-scale AnMBR. This section reviews the latest outcomes of the pilot-scale AnMBR systems for DWW treatment to extract practical data for the commercial use of AnMBR based plants in the future.

2.1. Configuration models of pilot-scale AnMBRs for fouling control

This section deals with the evaluation of 13 pilot-scale AnMBR studies performed from 2011 to 2019 (Table 1). The two-stage reactor was used by 12 pilot-scale AnMBRs, featuring anaerobic reactor with an externally submerged membrane tank (MT). Only one AnMBR system with gas sparging (Config-9) having one stage integrated with an up-flow anaerobic sludge blanket (UASB) combined with a submerged type membrane in the upper portion of the UASB reactor system was reported. In the case of a 2-stage reactor configuration, the membrane is immersed in a separate tank so that intensive shear stress can be applied only on the membrane to control fouling. Moreover, in situ membrane chemical cleaning can be performed in the membrane tank without endangering the whole active biomass. The retentate from the membrane tank is circulated back into the anaerobic reactor to enhance the degradation of organic compounds. In config-12, instead of the continuous stirred tank reactor (CSTR) and separate membrane tank, anaerobic fluidized bed reactor (AFBR) and anaerobic fluidized membrane bioreactor (AFMBR) are used in 2-stages (Config-12; Table-1).

The bibliographic study of the 13 AnMBRs (Table-1) shows that three different scouring approaches were used: gas sparging (GSP) (Config-1 to Config-11), particle sparging (PSP) (Config-12) and rotating membrane (RMEM) (Config-13). Most of the membranes were hollow-fiber (HF) submerged membrane except the first one which was a flat sheet (FS) membrane module called GSP-FS (Config-1). AnMBR systems used ultrafiltration (UF) membranes except for GSP-HF_{MF} (Config-6) which featured a MF membrane.

Apart from gas sparging, particle sparging and membrane rotation, some additional mechanisms were also employed in different configurations. In Config-1, circulating sludge was used to create additional shear to control membrane fouling. Ventilation and backwashing mechanisms were used in Config-3 and Config-4. Backwashing was used as an anti-fouling mechanism in Config-6 and Config-9. Chemical cleaning was employed for Config-7 and Config-8. Section 2.3 contains a detailed discussion regarding those different anti-fouling strategies.

2.2. Fouling mechanisms in pilot-scale AnMBRs

Membrane fouling is indicated by many factors like increased transmembrane pressure (TMP) (at constant flux), reduced flux (at constant TMP), presence of extracellular polymeric substances (EPS) and soluble microbial products (SMP) in the bulk phase. A significant number of studies are dedicated to understanding EPS and SMP behavior and role in AnMBR fouling (Chen et al., 2017a; Xiong et al., 2016; Ding et al., 2015; Chen et al., 2014; Herrera-

Robledo et al., 2011). EPS and SMP play a significant role in membrane fouling due to surface blocking and pore accumulation. SMP which consists mainly of soluble proteins, polysaccharides and humic-like material is defined as an organic matter that can pass through a filter of 0.45 µm and is responsible for adsorption on the surface and within the membrane pores (Chen et al., 2017a). The composition of extracellular polymeric substances (EPS) depends on the process parameters and wastewater composition as well as its origin. However, in general, it consists of insoluble carbohydrates, proteins, lipids and nucleic acid in a highly hydrated gel matrix (Laspidou et al., 2002). EPS contributes mainly to the cake layer fouling due to its ability to flocculate sludge on the membrane surface (Chen et al., 2017a). A study (Liu et al., 2012) explained that the cake formation in such systems was due to higher soluble microbial products (SMP) and higher tightly bound to loosely bound extracellular polymeric substances (EPS) ratio. Pore constriction and blockage results mostly in irreversible fouling but correspond only to 0.9% of total resistance to filtration, hence minimization of cake formation is more strategic than pore fouling in overall fouling mitigation (Chen et al., 2014).

Ding et al. (2015) reported that EPS extracted from bulk sludge and cake sludge were responsible for the largest decline in membrane flux. Herrera-Robledo et al. (2011) found that 80% of the fouling was caused by EPS and SMP in PVDF membrane tubes. Shin et al. (2014) employed an intermittent membrane relaxation mechanism to control the fouling and drastic rise of TMP.

Fouling phenomenon is also influenced by the type of membrane modules in pilot-scale AnMBR. Flat membranes appear to be more sensitive to fouling than hollow fiber membranes. Indeed, it appears that scouring methods are less effective for FS than for HF and a higher fouling rate of 3.33 Pa/s was observed for Config-1, i.e., flat-sheet membrane module (Martinez-Sosa et al., 2011); where a much lower fouling rate of 0-2.5 Pa/s was observed for Config-13, i.e., a hollow fiber membrane module (Martinez-Sosa et al., 2011).

Different fouling rates are observed also due to the difference in TMP. TMP of 17.7 kPa and 10 kPa were observed for Config-1 and Config-10 respectively indicating higher fouling in Config-1. It is evident from TMP and fouling rate values that Config-1 is more affected by fouling than Config-10 due to the requirement of higher TMP to achieve the same permeate flux. A higher level of fouling rates needs higher energy to reduce it. Config-1 requires higher energy for fouling control (1.28 kWh/m^3) when compared to Config-10 (0.23 kWh/m^3).

2.3. Fouling mitigation strategies in pilot-scale AnMBRs

Three major strategies for fouling mitigation are described below: scouring methods, filtration cycles and chemical cleaning cycles.

2.3.1. Scouring methods

Three methods: biogas sparging (GSP), particle sparging (PSP) and rotating membrane (RMEM) can be used to provide shear stress to mitigate membrane fouling. GSP approach is the most common strategy to control fouling in AnMBR; this technique has been used in 11 out of 13 pilot-scale studies referenced in Table 2. The crossflow velocity (CFV) is considered as a key factor for fouling mitigation when sludge recirculation is applied (Skouteris et al., 2012). Almost all the pilot-scale AnMBR systems (except GS-H5) incorporates sludge recirculation in the membrane tank to allow turbulent flow regime close to the membrane surface thereby reducing the thickness of the laminar boundary layer with better mixing of sludge (Lin et al., 2013; Smith et al., 2012). The applied CFV and Gas Sparging Intensity (GSI_m) values were 9.7–94 m/h and 0.15–1.20 N.m^3/(m^2h), respectively. Config-7 used $FeCl_3$ (26 mg/L) for flux enhancement. Addition of coagulant results in the reduction of fouling potential by enhancing floc sizes of the colloidal foulants present in the bulk, i.e., improvement of membrane fouling mitigation (Judd, 2011; Holbrook et al., 2004).

AnMBR with PSP (Config-13) is known as AFMBR (i.e., anaerobic fluidized membrane bioreactor). In AFMBR, fluidizing granular activated carbon (GAC) acts as the support medium for microorganisms along with playing a role in the scouring of the membrane surface (Kim et al., 2011). AFMBR requires a lower amount of energy and shows lower fouling potential as compared to AnMBR with gas sparging (Aslam et al., 2016; Shoener et al. 2016; Aslam et al. 2014).

Rotating membrane sparging is the last fouling control approach employed in Config-13 of pilot-scale AnMBR. This system makes use of the turbulence produced by the rotating membrane module to mitigate the foulant deposition on the membrane surface (Jiang et al., 2012). This technique has recently emerged as an efficient method with higher fouling control and lower energy consumption (Ruigómez et al. 2017; Ruigómez et al. 2016;).

2.3.2. Filtration cycles in AnMBRs

All the above pilot-scale configurations except Config-13 were operated intermittently, i.e. filtration cycles were followed by relaxation cycles with optional cycles of backwashing. Most of the configurations used intermittent cycles of filtration with an integrated backwashing cycle except for the case of Config-12 and Config-7, respectively. Generally, the filtration time with respect to the total time of operation was larger with backwashing (around 79–96 %) than without backwashing (around 80–83 %).

2.3.3. Chemical cleaning cycles

A preventive approach to uphold the membrane permeability is intermittent maintenance chemical cleaning. Only 2 out of 13 experiments used chemical cleaning. In Config-7, maintenance cleaning was performed after every 7 days using a backwashing liquid consisting of 2000 mg/L citric acid at a flux of 32.2 LMH. Config-7 also underwent recovery cleaning by soaking the membrane sequentially (for 16 hours) in the solution of 2000 mg/L of NaOCl and 2000 mg/L of citric acid, respectively. In Config-8, recovery cleaning was performed 7 times (for 4–6 hours) during a period of 1350 days with 1000 mg/L solution of NaOCl at room temperature.

2.4. Removal of organics in pilot-scale AnMBRs

Operating conditions and performances of pilot-scale AnMBR used for DWWT are illustrated in Table 1 and Table 2. It was observed in all 13 studies that COD feed concentrations ranged from 198 mg/L to 1460 mg/L and HRT was in the range of 2.20–33 hours. For WW having higher concentrations of COD in the influent (i.e., COD> 500 mg/L), HRT was generally higher than 10 hours. The maximum HRT of 33 hours was observed for Config-13 while the minimum HRT of 2.2 hours was observed for Config-6. SRT for all AnMBR configurations ranged from 6 days to infinite value (no waste of biomass, except for sludge sampling). The maximum SRT of infinite days was observed for Config-8 and minimum SRT of 6.2 days was observed for Config-12. On the other hand, the maximum Organic Loading Rate (OLR) range of 3.77-4.97 $kg_{COD}/(m^3.d)$ was observed for Config-10 while Config-3 showed the minimum OLR range of 0.3-1.1 $kg_{COD}/(m^3.d)$.

The majority of the AnMBR systems were conducted under room temperature conditions (i.e., 17–35 °C) and Config-10 was operated over a broad range of temperatures (from 9 to 30 °C). The operating permeation flux was within 4.1–17 LMH; the highest flux of 17 LMH was achieved after the incorporation of 26 mg/L of $FeCl_3$ to Config-7 and Config-10. The concentrations of MLSS ranged from 600 to 32000 mg/L. It must be noticed that MLSS was higher in GSP systems (around 7000–32000 mg/L) than in PS systems (around 600–12000 mg/L).

All the configurations exhibited COD removal efficiencies greater than 85%. Config-12 shows a high COD removal efficiency (more than 90%) even if this process was conducted in psychrophilic conditions (9–11 °C). Introduction of the flux enhancer (e.g., $FeCl_3$) improved COD removal efficiency (79.9% to 93.7%) for Config-7 and Config-10 due to aggregation of colloidal organics followed by their rejection by the membrane.

2.5. Energy requirements in the pilot-scale AnMBRs for fouling mitigation

An increasing crossflow velocity allows reducing membrane fouling but requires more energy consumption. For example, increasing crossflow velocity from 1 to 2 m/s increased permeate flux by about 20 %, but also the energy requirement by about 60 % (Bourgeous et al., 2001). Besides, the high shear rate can provide a negative effect on microbial activities in AnMBR due to the disaggregation of microbial flocs. The data of energy consumption along with the corresponding transmembrane pressure for all the AnMBR configurations (11 GSP AnMBRs, one PSP AnMBR and one RMEM AnMBR) is given in Table 3. It is evident from Table 3 that the energy requirement for fouling control in AnMBRs is the most significant constituent of the system, as it constitutes more than 75% of the total energy intake (Smith et al., 2014; Lin et al., 2011). The energy requirement for fouling mitigation mainly relies on the scouring method (Pretel et al., 2014). The estimated energy demand in the GSP AnMBR system varies between 0.05 kWh/m^3 and 1.66 kWh/m^3 with an average energy of around 0.39 kWh/m^3. Among all the GSP AnMBRs, the highest energy demand was observed for the FS membrane (Config-1). The average energy demand for Config-1 was 1.66 kWh/m^3 having a peak GSI_m value of 1.2 N m^3/ (m^2.h) with a comparatively lower critical flux of 7.0 LMH (Krzeminski et al., 2012; Verrecht et al., 2008). The average energy requirement for GSP AnMBRs with HF membranes (Config-2 to Config-10) was 0.28 kWh/m^3. However, GSP

AnMBRs still requires roughly 65% less energy than AeMBRs (0.70–0.90 kWh/m^3) (Krzeminski et al., 2012).

The energy requirement for a GSP AnMBR depends on the flux conditions and GSI_m. HF AnMBRs require lower GSI_m than FS modules. Amongst hollow fiber AnMBRs, Config-7, Config-9 and Config-10 required the lowest energies due to lower GSI_m values of around 0.15-0.32 N.m^3/(m^2.h). For Config-7 and Config-10, lower GSI_m was achieved by the addition of coagulant (i.e., flux enhancer). On the contrary, lower GSI_m in Config-9 is due to lower solid concentration in the membrane containing tank coupled with recirculation of liquid between the membrane tank and the biological reactor.

The relationship between energy requirement and flux is illustrated by drawing a comparison of flux, TMP and energy demands for Config-3 and Config-4 operated under the same GSI_m. The increment of flux from 10 LMH (Config-3) to 13.3 LMH (Config-4) at the same GSI_m resulted in lower energy demand (0.164 kWh/m^3) for Config-4 as compared to Config-3 (0.198 kWh/m^3). Similarly, the specific energy demand of PSP AnMBR can also be reduced by increasing the flux. The energy demand in the PSP AnMBR (Config-10) for GAC fluidization was reported to be 0.102 kWh/m3. McCarty et al. (2011) assessed the decrease in energy requirement to be 0.070 kWh/m3 for PSP AnMBR. The estimated energy demand for a staged anaerobic fluidized AnMBR (Config-10) is 0.23 kWh/m3 with a flux of 10 LMH and membrane rotation velocity of 100 rpm.

3. Novel AnMBRs for fouling mitigation

This section has designed to demonstrate (1) novel AnMBRs configurations and their impact on fouling; (2) impact of specialized fouling mitigation techniques on total fouling control in AnMBR for DWWT. Figure 1 shows the schematics of different novel configurations (a, c, e and f) of AnMBRs and specialized fouling mitigation strategies (b, d, g and h).

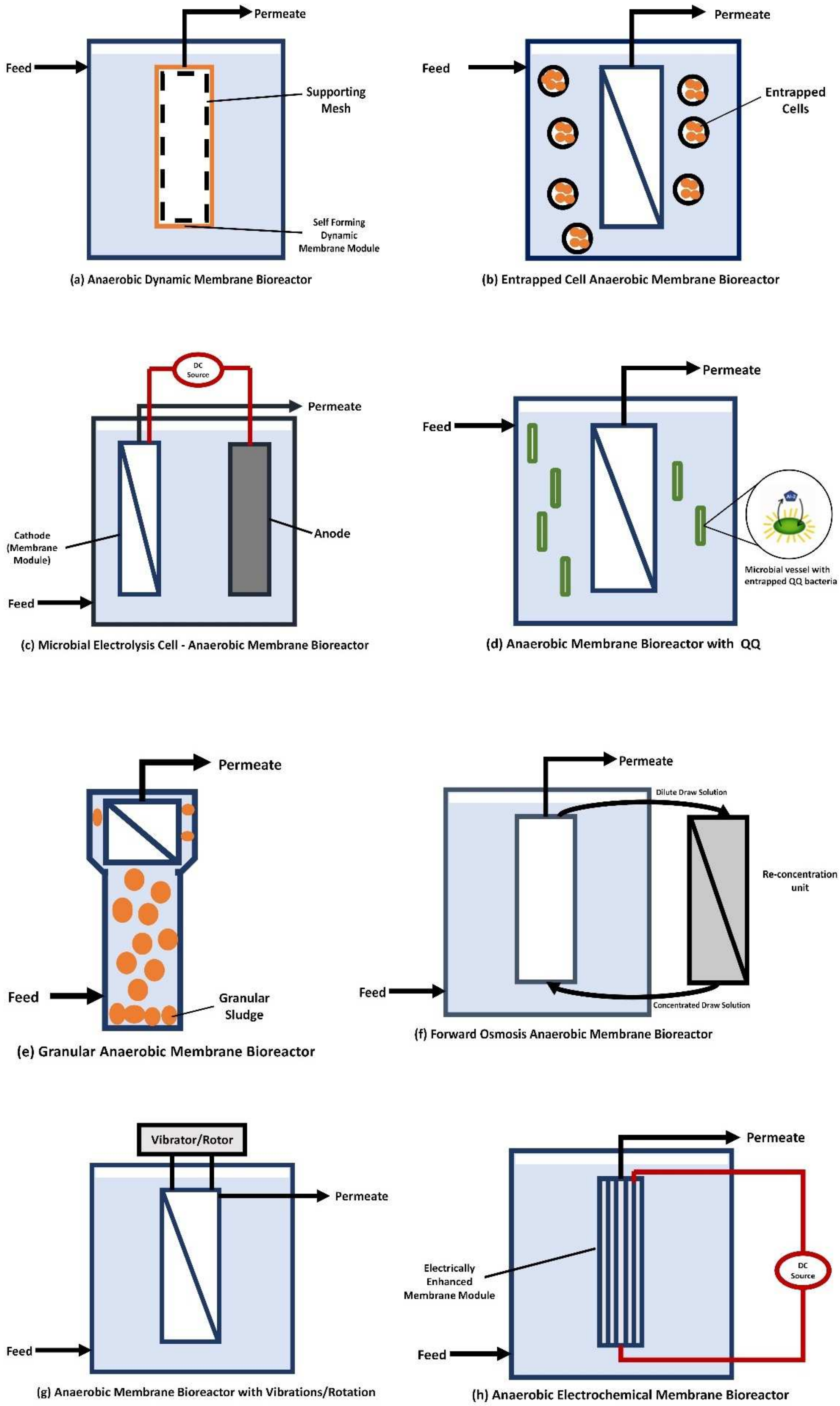

Figure 1 Schematics of different AnMBR schemes. (a) Dynamic AnMBR(b) Entrapped Cell AnMBR (c) MEC-AnMBR (d) AnMBR with QQ (e) G-AnMBR (f) FO-AnMBR (g) AnMBR with vibration/rotation (h) Electrochemical AnMBR

3.1. Granular-AnMBR

Coupling of membrane separation with anaerobic granular sludge (AnGS) reactors constitutes an interesting perspective. The most promising feature in G-AnMBR is the way to control membrane fouling. The structure of AnGS is not only justified for decantation but also to limit fouling phenomena. AnGS recirculation promotes mechanical scouring actions alongside the membrane surface and contributes to reduce membrane fouling. Fluidized AnGS comes into physical contact with the membrane surface, and their physical movement helps to reduce membrane fouling at a relatively low energy cost (Kim et al., 2011). Furthermore, when AnGS are used as fluidized media, the biological kinetics rate increases leading to a reduction of colloidal and dissolved organic matter (DOM), further decreasing irreversible fouling due to this DOM and increasing treatment stability and efficiency (Aslam et al., 2014). However, very little knowledge is available regarding the mechanisms of the mechanical cleaning process and fouling mitigation during DWWT by AnGS in G-AnMBR. Although G-AnMBR has a great potential to achieve energy-positive domestic wastewater treatment, the extent of fouling and energy consumption in the G-AnMBR can be strongly affected by the biological and physio-chemical properties of the AnGS. There are still several research questions to be resolved to optimize the selection of granular sludge. Besides, the energy requirement for fluidization in the G-AnMBR is closely related to the packing ratio of AnGS and their physical properties such as nature, size and shape. A better understanding of the links between AnGS characteristics, operating conditions (characteristic of the feed (DOM, mass loading rate), hydrodynamics, solid and liquid residence time) and fouling mitigation efficiencies (TMP, resistances), are thus needed (Aslam et al., 2017; Choo et al., 1998). The following section will discuss studies on G-AnMBR in detail and will also present a comparison with conventional AnMBR.

3.1.1. G-AnMBR v/s Conventional AnMBR

G-AnMBR combines the benefits of granulation and membrane separation and overcome drawbacks associated with conventional anaerobic sludge bioreactor like long startup time, high operating temperature, poor settling, poor nutrient removal and need for post-treatment (Chen et al., 2016). A study (Martin-Garcia et al., 2011) comparing the performance of G-AnMBR versus conventional flocculated AnMBR during 250 days showed almost similar COD removal in both the cases (84% in AnMBR and 86% in G-AnMBR). However, a great difference in MLSS and SMP bulk concentrations (598±150 mgCOD/L for AnMBR and 198±73 mgCOD/L for G-AnMBR) was found, indicating less fouling propensity in G-AnMBR. Protein to carbohydrate (P/C) ratio in SMP was found higher for G-AnMBR (2.8 in G-AnMBR v/s 2.3 for AnMBR), probably due to infinite SRT compared to AnMBR which resulted in accumulation of proteins in SMP and thus indicating low biodegradability of proteins in case of G-AnMBR.

Superficial liquid velocity (V_L) induced by gas velocity (V_G) (nitrogen in this case) was much higher in the case of G-AnMBR for similar V_G of 0.02 m/s, (i.e. had V_L of 0.26 and 0.09 m/s for G-AnMBR and AnMBR respectively). Thus, injection ratio ($IR = V_G/ (V_G+ V_L)$) was much lower for G-AnMBR (0.07 to 0.2) compared to AnMBR (0.26 to 0.71), indicating better scouring action in the case of G-AnMBR. Interestingly, for the same flux of 11 to 12 LMH, the fouling rate in G-AnMBR was only 1.67-3.33 Pa/s compared to a much higher fouling rate of 13.33 to 41.67 Pa/s in AnMBR. Also, using fluid pumping to generate crossflow velocity (CFV) for the same increase in CFV (0.4 to 2m/s), a greater increase of flux was observed in G-AnMBR (4 to 41 LMH) compared to AnMBR (4 to 19 LMH) indicating lesser CFV requirements for suppression of cake enhanced concentration polarization in case of G-AnMBR. Even in the case of an immersed HF module, gas (nitrogen) demand was 50% less in G-AnMBR as compared to AnMBR (Martin-Garcia et al., 2011). These low scouring requirements and less fouling make G-AnMBR a potential solution for fouling and energy-related issues in AnMBR for DWWT. Figure 2 summarizes the pros and cons of G-AnMBR.

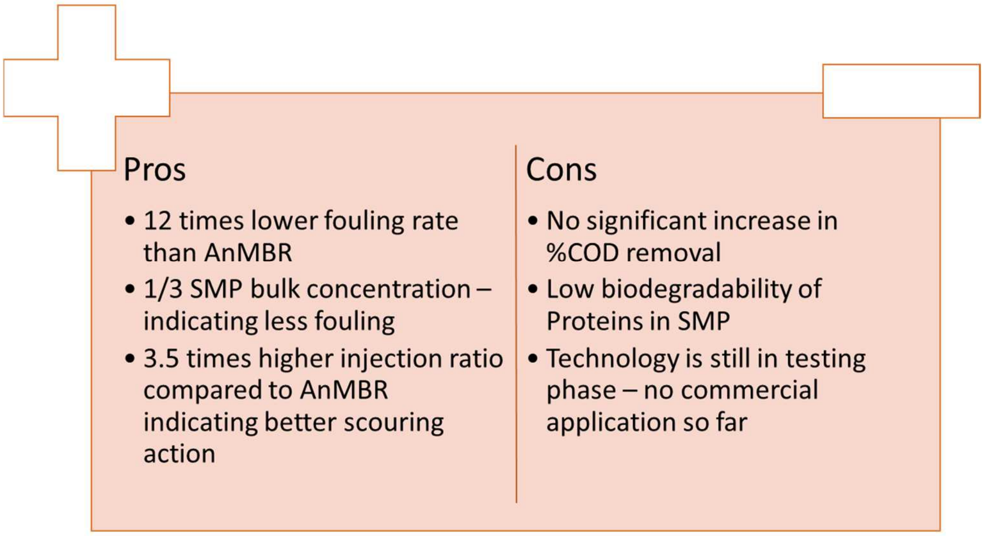

Figure 2 Pros and Cons of G-AnMBR compared to AnMBR

3.1.2. Effect of configuration on fouling in G-AnMBR

When internally submerged (SG-AnMBR) and external (EG-AnMBR) membrane configurations were compared, it was found that with approximately the same COD removal efficiencies SG- AnMBR was more prone to fouling than EG-AnMBR (Chen et al., 2017b). It was explained that the direct addition of membrane in the reactor results in almost complete colloidal flocs retention resulting in their accumulation and thus enhancement in the growth of bulking sludge which is dispersed and has poor immobilization. This phenomenon dominates over the growth of granular sludge and thus resulted in low biomass growth rate (0.02 gMLSS/d v/s 0.05 gMLSS/d), settling velocity (12.1-17.2 m/h v/s 14.1-28.5 m/h) and zeta-potential (-19.1 v/s -13.1) in SG-AnMBR compared to EG-AnMBR. Accumulation of fine and small flocs in SG-AnMBR also accounted for higher MLSS (180.2 ± 9.12 mg/L) which in turn was responsible for higher EPS and SMP production thus more fouling. Moreover, protein-based EPS in the cake layer was higher for SG-AnMBR (11.7 mg/g of cake layer) than EG-AnMBR (8.5 mg/g of cake layer), indicating a stickier cake layer resulting in more severe fouling and permeate flux drop. The same was observed for SMP (18.6 v/s 10.2 mg/g of cake layer) resulting in more adhesion of sludge on the membrane surface. This could cause irreversible fouling by forming a thin gel layer at the membrane surface (Chen et al., 2017b). Biopolymers were found to be major contributors in organic fouling with a noticeable difference of concentration in both configurations. Reported values in SG-AnMBR and EG-AnMBR were 14.6 and 6.8 mg/L, respectively. Their high concentration might result in the build-up of a hydrophilic layer by biopolymers attachment on the membrane surface. Building blocks (BB), low molecular weight (LMW) neutrals and acidic compounds can also enhance biopolymer production and attachment to the membrane surface thereby contributing to the increased fouling. Their concentrations were also higher for SG-AnMBR (6.3 mg/L BB and 9.7 mg/L LMW) compared to EG-AnMBR (4.6 mg/L BB and 5.9 mg/L LMW) (Chen et al., 2017b).

3.1.3. Effect of gas sparging regime on performance and energy demand in G-AnMBR

To evaluate the effect of the GSP regime on performance and energy demand of G-AnMBR, (Wang et al., 2018a), a study was performed with pilot-scale SG-AnMBR treating DWW and operating it under three different GSP conditions that were: (1) continuous filtration and GSP, (2) continuous filtration with intermittent GSP and (3) intermittent filtration and GSP in pseudo dead-end filtration (filtration cycle of nine minutes without GSP followed by one minute of GSP and membrane relaxation). HRT was kept at 8 hours and the temperature of the influent was around 16.3±3.7 °C. 40 % bed expansion was achieved by maintaining an up-flow velocity of 0.8-0.9 m/h. During the 400 days trial, there was no withdrawal of biomass. Intermittent filtration and sparging in pseudo dead-end filtration was found to be the most suitable configuration among all three that had provided sustainable fouling rates with minimum energy requirements (0.133 kWh/m^3 with net SGD of 0.2 $m^3/(m^2.h)$). The authors explained that in the case of dead-end filtration, particulate materials, colloids and soluble materials deposited simultaneously compared to continuous GSP where induced shear stress close to the membrane surface caused particle size segregation resulting in preferential migration of colloids towards the membrane surface. This deposition resulted in a heterogenous cake layer. However, this is only possible when solid concentration is low enough to allow limited cake deposition in a specific filtration cycle since TMP and filtration time influence cake compressibility. At 13.5 LMH, SGD per unit permeate of 14.8 m^3/m^3 was similar to that in a plant scale AeMBR treating municipal waste (14-30 m^3/m^3) despite more complex fouling behaviors on anaerobic treatment.

3.1.4. Sponge assisted G-AnMBR

To minimize the fouling rate in SG-AnMBR, the effect of polyurethane sponge incorporation was studied (Chen et al., 2017c). Decreasing the fouling rate in SG-AnMBR will make it superior to external configuration since it has lower capital and operational costs. Sponges act as media to provide a high specific area for the growth of biomass by increasing the flocculation of sludge due to porosity and resistance against hydrolysis. Sponge assisted G-AnMBR contains about 84 % of total granular sludge and this is about two times higher than conventional granular bioreactor (about 42.5 %) due to the immobilization of fine particles on or inside the pores of the sponge. EPS plays an important role in granular formation by allowing cell integration and helping to keep sludge intact. The average fouling rate in sponge assisted G-AnMBR was about 0.006 Pa/s which is lower than that of conventional G-AnMBR (0.014 Pa/s). This difference in fouling rates can be explained by SMP and EPS concentrations in mixed liquor as well as the cake layer. In conventional G-AnMBR, granules and floc breakage increase SMP concentration (47.3±7.6 mg/L) and EPS (24.5±11 mg/L). In contrast, sponge assisted G-AnMBR exhibits relatively low SMP and EPS concentrations of

15.9±3.5 mg/L and 17±6.2 mg/L, respectively. Lower SMP and EPS concentrations are due to adsorption of sludge on the sponge and the majority of biodegradation occurs inside granules and sponge-attached biomass. Reduced free fine flocs and colloids in sponge assisted G-AnMBR resulted in a less dense fouling cake layer than that observed in conventional G-AnMBR. Overall fouling resistance is reduced since cake layer fouling is dominant in AnMBR. Moreover, cell lysis in conventional G-AnMBR results in higher protein EPS (12.1 mg/g of cake layer in conventional v/s 10.7 mg/g of cake layer in sponge assisted) and protein SMP (8.2 mg/g of cake layer in conventional v/s 5.6 mg/g of cake layer in sponge assisted) which forms stickier cake and thus contributes more to fouling filtration resistance. In addition to less fouling, sponge assisted G-AnMBR also provides the benefit of a 17 % higher methane yield. Another configuration that has employed sponges and that was designed for fouling control was sponge assisted AnMBR with rotary disk (Kim et al., 2014). It utilized polyurethane sponge to support microbial growth and the disk rotates parallel to the surface of 2 flat sheet submerged membrane for fouling control. 96% COD removal and 150±29 mL CH_4/g COD methane yield were obtained by performing experiments using a 50 % volume fraction of sponge, 6 hours HRT, 11 LMH flux and 70 rpm rotation speed of the disk. TMP rise rate of 0.15 Pa/s was observed compared to 5 Pa/s without rotation indicating effective fouling mitigation. This was attributed to high collision energy between sponge and membrane surface, reducing cake enhanced concentration polarization. Operational electricity consumption was ten times lower by combining rotation and sponge media than the energy produced by methane combustion. As a result, the operation of this hybrid AnMBR provided net positive energy of 0.04 kWh/m^3. This study demonstrated a potential configuration for the net positive energy operation of AnMBR for DWWT. However, a detailed study regarding fouling behaviors under different operational conditions and floating media is necessary before scale-up.

3.2. Entrapped cell AnMBR

Cell entrapment makes use of a polymer matrix to artificially entrap cells providing increases resistance to washout. Juntawang et al. (2017) used phosphorylated polyvinyl alcohol for cell entrapment in AnMBR and compared it with conventional suspended cell AnMBR. Both units showed the same COD removal efficiencies (approximately 84 % COD removal) which indicates that cell entrapment has no negative impact on treatment. Fouling resistance in entrapped cell based AnMBR was found to be 0.32×10^6 1/m for pore blockage and 1.06×10^6 1/m for the cake layer. These are less than the suspended one (2.54×10^6 1/m for pore blockage and 1.69×10^6 1/m for cake layer) indicating less fouling propensity. Cell entrapment results in lower EPS and SMP concentration as well as larger particle size. Conventional AnMBR has much higher pore blockage resistance which indicates more potential for irreversible fouling and thus would need more intensive chemical cleaning. However, bacterial communities in both the cases were similar except bacteroidetes colonies which were more abundant in suspended cell AnMBR than in entrapped cell AnMBR. The formation of these colonies is favored in the presence of proteinaceous EPS and thus high protein to carbohydrate ratio of EPS. Methane yields in both cases were almost similar i.e. 0.23 L/d for entrapped cell bases AnMBR and 0.28 L/d for the suspended one.

In 2019, Juntawang et al. (2019) have used the entrapment technique in FO-AnMBR. Polyvinyl acetate (PVA) based entrapped cells were used in side-stream configuration with thin-film composite (TFC) FO membrane to treat domestic wastewater having initial soluble COD of 542 mg/L. Two draw solutions namely NaCl (1.5M) and $(NH_4)_2SO_4$ (1M) were tested. In the case of entrapped cells, flux decline rates were 0.042 and 0.049 LMH/day for FO-AnMBRs using NaCl and $(NH_4)_2SO_4$ as draw solutions, respectively. Whereas, for suspended cell FO-AnMBR they were 0.057 and 0.074 LMH/day, respectively. Additionally, EPS was 35% and 13% less and SMP was 65% and 68% less in entrapped cell FO-AnMBR using NaCl and $(NH_4)_2SO_4$ as draw solutions, respectively. This could be explained by the fact that entrapment keeps the cell activity restricted, and thus control the formation of EPS and SMP. In addition, it might also keep EPS and SMP trapped in, suggesting that fouling potential should be higher with suspended cells than entrapped cells. Entrapped cell-based technologies could be a step forward in terms of fouling mitigation. As seen, they provide better fouling mitigation than conventional AnMBR. More research in this area is needed to conduct energy and cost comparisons so that the technique can be further developed.

3.3. Dynamic AnMBR

In the recent years, anaerobic dynamic membrane bioreactors (AnDMBR) have been investigated as a sustainable solution to wastewater treatment due to their low cost of the membrane, reasonable treatment efficiency (providing 60-90% COD removal and 90-100% turbidity and suspended solids removal) and less fouling (0.6×10^9 m^{-1}/h rate of increase of fouling resistance compared to 40×10^9 m^{-1}/h in AnMBRs) (Hu et al., 2018a). Dynamic membrane (DM) bioreactors make use of cheap materials like meshes or fiber cloth as support on which the cake layer is formed instead of expensive ultrafiltration or microfiltration membranes. This cake layer acts as an additional or secondary membrane (dynamic membrane layer) due to its capability of rejecting pollutants such as colloidal materials, microbial cells and organics. Although DM was reported firstly by workers of Oak Ridge National Laboratories in 1965, its application in AnMBR is relatively new and less developed. (Alepu et al., 2016). This section aims at reviewing the existing literature on AnDMBR and how they are useful in resolving the fouling issues for domestic wastewater treatment.

3.3.1. DM layer formation mechanism in AnDMBR

A study (Zhang et al., 2010) proposed a three-step formation mechanism that involves a separation layer formation stage, stage of stable growth and finally a fouling stage. This mechanism was confirmed by other studies as well (Hu et al., 2018b; Siddiqui et al., 2018; Alepu et al., 2016; Alibardi et al., 2016; Ersahin et al., 2016; Zhang et al., 2011). The time required for a stable DM layer formation depends on the operation mode of the reactor. In gravity-driven mode, which allows continuous extraction of effluent without relaxation, this time is about 40-100 minutes whereas in the pressure-driven mode it is about 10-25 days. Operating with intermittent permeate production leads to longer stabilization time both in pressure-driven mode (approximately 50 days) and gravity-driven mode (7 days) (Hu et al., 2018a) compared to the continuous operation.

3.3.2. Submerged versus external dynamic AnMBR

Membrane configuration plays a very significant role in defining performance efficiency as well as fouling propensity in AnDMBR. Submerged AnDMBR was found to be slightly better in terms of removal efficiency on COD and turbidity (99.5 and 99.7 %) than external configuration (Ersahin et al., 2017). The authors also found out that the time required to form a stable DM layer in submerged configuration was only 10 days while it was 20 days in external configuration at the same permeate flux of 2.2 LMH. The DM layer was less stable in the external dynamic membrane due to the high shear rate which affected the system performance. These results indicate that submerged configuration is more appropriate than the external configuration for applications that require smaller startup time and a more stable DM layer. It was also mentioned that increased shear stress in the external case disturbed the balance of the microbial ecosystem and reduced the methane formation thus providing a negative impact on the overall energy balance of the system. This was proved by the fact that the submerged configuration had an average methane flow of 2.4 L/days whereas the external one had 1.9 L/days. However, it was also seen that the submerged configuration has higher filtration resistance (10.2×10^{16} 1/m) and TMP (68.0 kPa) than the external one with 7.4×10^{16} 1/m resistance and 38.0 kPa TMP (Ersahin et al., 2017). Higher resistance in submerged dynamic AnMBR was probably caused by the formation of a thick DM layer. External recirculation has an additional scouring effect on the membrane. Differences in performance efficiencies were not too high. To understand which configuration is better for full-scale implementation, it would be important to establish complete energy and economical balances.

3.3.3. Effect of operating conditions in AnDMBR

Operating conditions have a significant effect on DM formation, performance and fouling in AnDMBR.

3.3.3.1. Effect of flux

Higher operating fluxes are required in AnDMBR operations to reduce HRT and thus capital cost and footprint. It was observed that higher flux led to a faster formation of the DM layer. However, the time required for stable operating was reduced from 10 to 5 days as flux increased from 50 to 150 LMH. Higher flux resulted in a faster adsorption rate of suspended solids on the membrane surface (Wang et al., 2018). However, as for other membrane processes, higher fluxes can result in more severe fouling; Other studies observed that higher initial flux causes the higher increase of TMP (Siddiqui et al., 2018; Quek et al., 2017; Saleem et al., 2016; Chu et al., 2014).

3.3.3.2. Effect of organic load and sludge recycling

Hu et al., (2018b) studied the effect of organic load and sludge recycling on the performance of a flat-sheet AnDMBR at 20 °C and a short HRT of 8 hours. In both cases, without sludge recycling (CFV=0.24 m/h) and with sludge recycling (CFV=0.83 m/h), when organic loading rate (OLR) was increased from 0.88 $kg_{COD}/(m^3.d)$ to 3.01 $kg_{COD}/(m^3.d)$ an increase in COD removal efficiency (approximately 23 % increase) as well as methane yield (approximately 5 times increase) were observed. Higher COD removal was attributed to higher retention of organic compounds by a thicker DM layer. However, when OLR was increased, by adding synthetic wastewater, the availability of organic matter to bacteria was increased thus allowing to produce more methane. With sludge recycling, CFV could be increased but it impacted the COD removal negatively. Similar results were observed by (Siddiqui et al., 2018) where an increase of CFV from 54 m/h to 90 m/h s increased turbidity in the permeate (from 10 to 17 NTU). Higher CFV has reduced the thickness of the DM layer, allowing particulate matter to pass through the membrane, which has led to lower permeate quality. The authors also observed that when the organic load was increased, the filtration properties of the membrane deteriorated. TMP did not exceed 15 kPa throughout the operational period when OLR was 0.88 $kg_{COD}/(m^3.d)$, but it rose quickly to 20kPa within 20 d of operation for OLR of1.5 $kg_{COD}/(m^3.d)$. For the OLR of 3.01 $kg_{COD}/(m^3.d)$, TMP was over 15 kPa in less than 20 d. This behavior could be explained by the fact that more biomass is accumulated in the DM layer at higher OLR.

The thickness of the DM layer with sludge recycling was less than without recycling (0.54-0.95 mm compared to 0.76-1.27mm without recycling) due to improved back transport by increased CFV. However, TMP increased faster (15-18 kPa with OLR of 0.88 $kg_{COD}/(m^3.d)$) because of sludge structure modification during recirculation. Sludge recycling promoted the formation of fines and colloids, which may have accelerated pore blockage. Moreover, higher amounts of EPS and SMP were observed with sludge recycling (27.5 mg/g MLSS total EPS and 2.67 mg/g MLSS total SMP) compared to without sludge recycling where 13.58 mg/g MLSS total EPS and 1.06 mg/g MLSS total SMP was observed. They were responsible for the higher fouling rate in the case of sludge recycling.

3.3.3.3. Effect of operating time

Operating time or filtration period can have a significant impact on the fouling rate in AnDMBR. It was observed that increasing filtration duration from 60 minutes to 720 minutes increased DM thickness from 2.29 mm to almost 3 mm. This results in irreversible fouling that could not be recovered by backwashing Moreover, the total amount of EPS increased from 28 mg/g VSS to 42 mg/g VSS and total SMP increased from 4 mg/g VSS to 6.5 mg/g VSS as the operation time increased from 60 to 720 minutes (Siddiqui et al., 2018).

3.3.3.4. Effect of mesh pore size

Mesh size from 10 µm to 200 µm was tested in AnDMBR studies. It can be observed (Table 4), that an increase in mesh pore size improves the flux due to reduced filtration resistance. However, it affects the removal efficiency negatively by allowing more passage of solids through mesh pores than UF/MF membranes with much smaller pore sizes. Thus, a trade-off exists between permeate flux and removal efficiency. However, the difference in COD removal efficiency is not significant whereas with larger pore size (46 µm pore size compared to 28 µm) filtration time can be increased up to four times indicating that smaller pore sizes are more prone to fouling (Quek et al., 2017). Moreover, meshes with larger pore sizes are cheaper than smaller pore size ones (Alibardi et al., 2016).

3.3.4. Fouling mechanism in AnDMBR

Fouling has a different meaning for AnDMBRs. Initial accumulation of material on the mesh is the desired DM formation and after DM formation, subsequent deposition that leads to increased resistance to filtration is regarded as fouling (Chu et al., 2014). In one study (Wang et al., 2018) the membrane fouling mechanism for self-forming AnDMBR at 25 °C with polyamide nylon mesh (150 µm pore size) supported by a hollow cylinder was described. The author experienced a three-stage fouling mechanism in AnDMBR operating at a flux of 100 LMH. At the first stage, a decrease in resistance was observed. This was due to the increase in membrane hydrophilicity. Hydrophilicity reduces the adsorption of pollutants on the membrane surface. In the second stage, the resistance was rather stable due to the low content of EPS in the system. Finally, in the third stage, resistance increased sharply indicating blockage of membrane pores and enhanced compactness of cake. Zhang et al., (2011) observed membrane foulants in AnDMBR and observed a two-layered structure with an inner layer tightly bonded to the mesh surface and an outer layer loosely bonded to the inner layer. SEM analysis showed that the cake layer developed during the first stage had larger pores and loosely packed sludge flocs. As the stages process, cake compactness has started to increase resulting in a very compact cake layer in the third stage. A similar observation was made by another study (Sun et al., 2018). It is assumed that solid accumulating varies in size, initial coarse particles get deposited but as the DM layer formed, the deposition of small particles leads to a denser cake (Figure 3). Moreover, EPS, polysaccharides, proteins and total cells (that are responsible for fouling in AnMBRs) increased in content as the stages proceed. It was also found (Ersahin et al., 2016) that the average amount of SMP, EPS and EPS P/C ratio of the DM layer was 21.5, 5.8 and 1.9 times (respectively) more than the bulk sludge indicating more resistance and stickier cake.

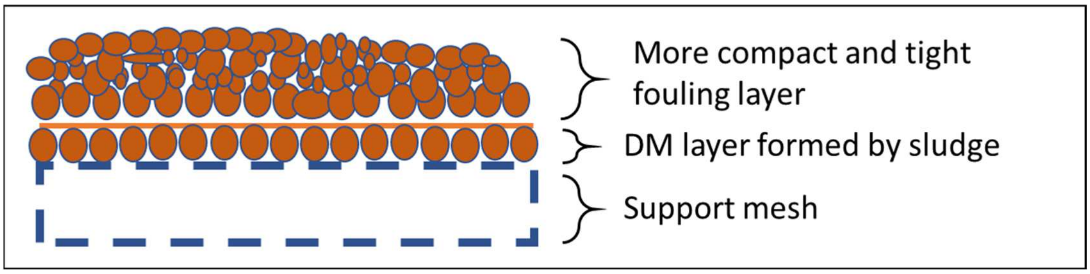

Figure 3 Foulant layer distribution in AnDMBR

3.3.5. Fouling mitigation in AnDMBR

AnDMBR has provided a cheaper alternative to expensive MF/UF membranes for AnMBRs. The buildup of cake is useful for AnDMBRs but beyond a certain level, it is regarded as fouling and must be controlled to have a long and stable operation. Fouling mitigation techniques like CFV, biogas sparging, back flushing and relaxation can serve the purpose. CFV could help in prolonging the AnDMBR operation however once the cake layer is developed application of large CFV, only has a short-term impact on fouling mitigation. It reduced the TMP value to half (from above 20 kPa to below10 kPa) when CFV increased from 1 to 5 m/h (Alibardi et al., 2014) but this effect did not last for long and a sharp increase in TMP was observed again after a short time. Moreover, high CFV could modify sludge resulting in a negative impact on filterability as well as biological activity (Hu et al., 2018b).

Some studies employed GSP as fouling control in AnDMBR (Li et al., 2017; Yang et al., 2017; Li et al., 2016). Application of biogas sparging (at a rate of 1 L/min) decreased foulants accumulation (38 days to reach maximum TMP with biogas sparging compared to 15 days without) on the mesh surface and offered a less rapid increase of TMP compared to the operation without biogas sparging. This indicates that biogas sparging can effectively scour the foulant layer in DM membranes and increase the operation time. A similar observation was made by Yang et al., (2017) that used N_2 sparging. However, it is important to control the sparging rate. Too much sparging can disturb the DM layer and affect the performance of AnDMBR. Compared to conventional AnMBR less sparging rate is required in AnDMBR as removal of foulants from mesh is easier than MF/UF (Li et al., 2016).

As AnDMBR makes use of the cake layer for filtration with comparatively cheap mesh supports which can be easily cleaned and the cake layer can be reformed, not much attention is given to in situ fouling control in AnDMBR. Hence there is a need to investigate in further detail the effect of in situ fouling control methods on performance as well as the lifetime of AnDMBRs. It is also needed to carry out economic and energy calculations on AnDMBR with and without in situ fouling control to establish comparisons with conventional AnMBRs and evaluate the feasibility of AnDMBRs for the long term and large-scale applications.

3.4. Forward Osmosis AnMBR (FO-AnMBR)

3.4.1. Configuration and working principle of FO-AnMBR

FO-AnMBR combines an AnMBR where the membrane is a FO membrane instead of UF/MF in the conventional AnMBR process. FO relies on an osmotic gradient as a driving force; and therefore, high osmotic pressure solution (seawater, seawater brine, fertilizer…)

also called draw solution is used to allow for permeation and extraction of water from WW (feed) through the FO membrane. As such, unlike conventional AnMBR, FO-AnMBR operates on the osmotic pressure difference between the feed and the draw solution with little or no external hydraulic pressure. FO membranes are dense, similar to the RO membrane but thinner to limit the internal concentration polarization phenomenon. Hybridization of FO-AnMBR systems can be classified into various types based on membrane configuration (side-stream or submerged), draw solution (DS) regeneration system (open without regeneration or close with regeneration) and objective of treatment (wastewater treatment, resource recovery, wastewater pre-concentration and/or reclamation).

FO can also be coupled with AnMBR as pre or post-treatment for concentration purposes; in that case, AnMBR features a UF/MF membrane and the FO process is placed upstream or downstream of AnMBR. Pre-concentration of domestic wastewater may allow for higher COD load in the AnMBR and therefore optimized operation (Bao et al., 2019; Ferrari et al., 2019; Wang et al., 2016; Zhang et al., 2014) and post-treatment allows the nutrient recovery (Ansari et al., 2017; Xue et al., 2016) or concentration of volatile fatty acids (Blandin et al., 2019).

Current limitations of FO were mainly low flux associated with the first generation of membranes (<10LMH) and salt accumulation due to feed salt accumulation and reverse salt transport from the draw solution. High cost associated with the draw solution (DS) regeneration processes (Wang et al., 2016a) such as pressure-driven system (Nanofiltration or Reverse osmosis) temperature-driven (membrane distillation) or electricity-driven (electrodialysis) is also a point of concern (Ansari et al., 2017; Wang et al., 2016a). Water flux in FO processes also reduces due to the phenomenon of concentration polarization (CP). It makes the feed solution more concentrated on one side of the active layer and dilutes the draw solution on the other side thus, decreasing the effective osmotic pressure driving force.

Based on membrane orientation i.e. active layer facing feed (AL-FS) and active layer facing draw side (AL-DS), CP has been classified into four categories i.e. concentrative and dilutive external concentration polarization (ECP) (ECP takes place on the membrane surface); concentrative and dilutive internal concentration polarization (ICP) (ICP occurs inside the support layer) (Shaffer et al., 2015). Figure 4 illustrates these phenomena.

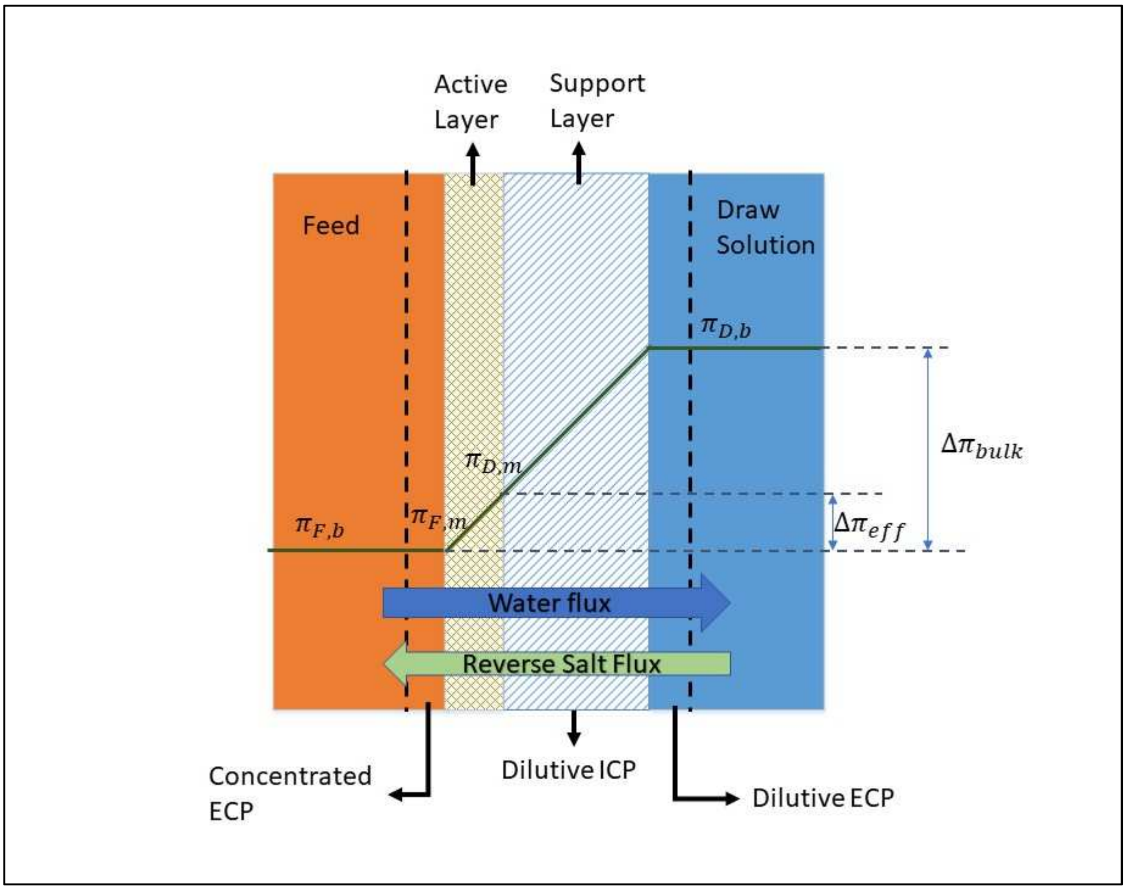

Figure 4 Illustration of internal and external concentration polarization across an asymmetric FO membrane module. Adapted from (Gulied et al., 2020).

However, recent development regarding new membranes and modules for osmotic membrane bioreactor allowing higher fluxes and very high rejection rate (including trace organic contaminants) as well as improved operation in submerged mode to limit clogging of challenging streams has opened doors to a new generation of FO-AnMBR (Blandin et al., 2018b; Blandin et al., 2016a). Salinity build-up is an inherent problem of the osmotic system and can be limited by the choice of appropriate draw solution. Finally, to limit draw recovery cost, synergies can be found with other processes such as fertigation, use of waste heat (MD) or combined desalination and water reuse.

3.4.2. Performance comparison of FO-AnMBR with conventional AnMBR

FO-AnMBR demonstrated higher removal efficiencies (96% organics, 100% total phosphate and about 62% ammonia nitrogen removal) than conventional AnMBR due to the use of dense FO membranes (Chen et al., 2014). FO-AnMBR has also shown a higher methane yield i.e. 0.25-0.3 L CH_4/g COD (treating dilute wastewater under mesophilic conditions) than conventional AnMBR (0.21 L CH_4/g COD at 25°C). It was reported that FO-AnMBR has lower fouling propensity than conventional MBRs due to the absence of hydraulic pressure and low flux. Due to this, foulants are not pushed on the surface of the membrane and hence result in less cake fouling.

A study by Chen et al. (2014) found out that the COD removal efficiency of FO-AnMBR was higher than the conventional AnMBR. Average COD removal efficiency was 96.7% (with cellulose acetate membrane) whereas in conventional AnMBR it varies from 28 to 90%. Thanks to the dense FO membrane, FO-AnMBR operation also resulted in almost complete phosphate removal and 60% removal of ammonia nitrogen which is favorable compared to conventional AnMBR with almost no removal of ammonia nitrogen. In addition to higher rejection, the longer residence time in the FO-AnMBR can lead to advanced biodegradation of organics resulting in lower COD in the supernatant and permeate. For example, supernatant COD was observed to be reduced from 210 mg/L to 180 mg/L as the operating time proceed (Chen et al., 2014). Similar behavior was observed by (Hou et al., 2017) where initially COD of the bulk increases and then after reaching a maximum value it dropped down with an increase in bio-degradation rate and a decrease in flux.

Table 5 summarized the COD removal efficiency and methane yield of different FO-AnMBR and conventional AnMBR units. Methane yield obtained was in the range of 0.21 to 0.3 L CH_4 / g COD (Gu et al., 2015; Chen et al., 2014); similar to that of AnMBR (i.e. 0.23 to 0.27 L CH_4 / g COD at 20-35 °C in Martinez-Sosa et al., (2011)).

3.4.3. Fouling and salinity build-up in FO-AnMBR

Flux decline (from 9.5 to 3.5 LMH) in continuous FO-AnMBR operation was observed (Chen et al., 2014) with an increase in conductivity from 1 to 20.5 mS/cm. This decline was attributed to two phenomena: membrane fouling and salinity increase. Membrane fouling occurred because of the attachment of foulants to the membrane surface thereby increasing hydraulic resistance and reducing flux whereas retention of feed solutes (by the dense FO membrane) and the reverse salt flux coming from the draw solution are the cause of the latter. A similar study on osmotic MBR highlighted that salinity increase may alter bacteria efficiency leading to deflocculating and enhanced fouling (Blandin et al., 2018a). Moreover, the cake layer disposition on the membrane hinders the back diffusion of the salt, resulting in the buildup of salt near the membrane surface. It could result in cake-enhanced osmotic pressure and lead to a reduction in the net driving force.

The presence of both protein and polysaccharides contributes to biofouling; A review (Wang et al., 2016a) commented that both EPS and SMP played a significant role. However, it was observed that the biofouling in the FO membrane was mostly external and nearly fully recoverable. When the membrane was rinsed and the supernatant was discharged to bring conductivity back to 1 mS/cm after one cycle of operation (22 days), 94% of initial flux was recovered (Chen et al., 2014).

3.4.4. Fouling mitigation in FO-AnMBR

Fouling mitigation can be done by modification of membrane properties, improved configuration and optimization of driving force, operating parameters and control strategies (Wang et al., 2016a). The effect of the driving force by using different concentrations of draw solution (NaCl) on membrane flux and fouling was studied (Wang et al., 2019). With increased driving force, initial flux increases but beyond a certain critical concentration the flux decline rate increased drastically (0.57 LMH/day to 1.24 LMH/day when concentration increased from 1 to 2 M). Moreover, an increase in the driving force resulted in more drastic fouling (fouling layer thickness increases from 35.20 μm to 96.5 μm with a concentration increase of 0.25 M to 2 M). Major constituents of the bio-foulant layer were found to be β-D-glucopyranose, proteins and bacterial cells whose bio-volume also increased with DS concentration. It is assumed that higher initial water flux increased the foulant deposition rate and raised the membrane resistance thereby, aggravated the fouling resulting in severe flux drop. As for MBR, evidence of critical flux has been mentioned for the osmotic system when using a new generation of FO membrane (Blandin et al., 2018a; Blandin et al., 2016b) and therefore similar behavior probably occurs in FO-ANMBR. Operating below critical flux would lead to a more stable and sustainable operation.

(Hu et al., 2017) compared the impact of NaCl and $MgCl_2$ as draw solutions on fouling behavior. Flux decline rate using $MgCl_2$ (0.3 LMH/day) was more severe compared to NaCl (0.16 LMH/day) indicating more fouling propensity. Moreover, the fouling layer in the case of $MgCl_2$ was 10 μm thicker than that of using NaCl as a draw solution. Enhanced fouling with $MgCl_2$ draw solution could be caused by the bridging ability between magnesium (divalent) ion and EPS. It was also observed that the fouling potential of the TFC polyamide membrane was higher compared to CTA membranes since fouling was reversible up to 94%

of flux recovery for the CTA membrane (section 4.3) but it was not the case with TFC probably as a result of higher flux.

3.4.5. Salinity build-up mitigation in FO-AnMBR

The salinity build-up in wastewater is a major issue to be resolved in FO-AnMBR which is inherent to the FO process (dense membrane). The accumulation of salts reduces permeate flux due to the loss of the osmotic gradient and can negatively impact the biological process.

One way to mitigate salt accumulation is to control reverse salt flux from the draw solution to the FO membrane. Using divalent salts (more rejected by the FO membrane) this could be envisioned. The study showed that (Tang et al., 2014) salt accumulation was greater when sodium chloride was used as the draw solution (salinity = 35 mS/cm) than other salts such as sodium sulfate (salinity = 11 mS/cm) under the same operating conditions in FO-AnMBR. Chloride ion has a smaller size than sulfate, thus exhibiting higher reverse salt flux. Moreover, the negatively charged FO membrane would offer more repulsion to sulfate ion than chloride due to differences in their charge and size. However, use of sodium sulfate lowered the methane production (only 2.6 % of methane in biogas compared to 12.9% as NaCl was used as the draw solution). The presence of sulfate can increase the production of sulfur-reducing bacteria hindering methanogens thereby affecting methane production. Thus, the use of a sulfate-based draw solution needs to be considered very carefully if significant methane generation is required.

Playing with HRT and SRT (wasting sludge to discharge accumulated salts) is an alternative to mitigate salt accumulation but should be controlled carefully and reduced SRT may negatively impact the process economics (Wang et al., 2016a). Another way to mitigate salinity build-up during the operating cycle is to couple a MF membrane with FO-AnMBR (Wang et al., 2017). Cellulose triacetate (CTA) FO membrane and polyvinylidene fluoride

(PVDF) MF membrane was submerged together into an anaerobic bioreactor with 0.5M NaCl of draw solution at 25 °C and at a biogas sparging rate of 2 L/min. It was found that the conductivity of the mixed liquor remained stable and low (2.5 – 4 mS/cm) compared to the previous study using FO-AnMBR, the MF membrane acting as the salt leak. The performances of both MF and FO membranes were in accordance with conventional AnMBR and FO-AnMBR yielding 90% and 96% of TOC removal efficiency, respectively. Also, methane yield ranged from 0.25 to 0.28 L CH_4/g COD. However, the fouling on the FO membrane was severe compared to conventional FO membranes because the permeate flux could not be recovered after chemical cleaning at the end of the operation (101 days).

3.5. Microbial Electrolysis Cell-AnMBR

Applying an electrical field in an anaerobic membrane process by coupling MEC and AnMBR is an innovative way to reduce membrane fouling (Ding et al., 2018). A PVDF hollow fiber membrane was submerged into a MEC where titanium mesh and carbon were used as cathode and anode respectively at external circuit resistance of 10 Ω and 36 hours of HRT. Voltage ranging from 0 to 1.2 V was employed to study the removal efficiency and fouling behavior of the system. COD removal with the voltage applied was about 20 % higher than that without it. However, COD removal was not constant with time but decreased slowly indicating a negative effect of high voltage on degradation kinetics of organic contaminants. This degradation was due to increased plasmatorrhexis, reduced metabolic activity and decrease in microorganism growth rate. Hence, there is a need to optimize voltage for maximum efficiency.

The rapid decline in resistance from 400 Ω at 0.4 V to 66.67 Ω at 0.6 V indicates the potential for fouling mitigation. Moreover, with an increase in applied voltage, cycle length, defined as the time of operation before cleaning is required, increased (60 h at 0.4 V to 98 h at 1 V) showing a reduction in cleaning requirement and hence lower fouling. The increasing voltage from 0 V to 1 V in MEC-AnMBR increases the zeta-potential from 22.3 mV to 30.9 mV. Higher zeta-potential of sludge resulted in less agglomeration of it on the membrane surface due to higher negative charge of the sludge, thereby reducing cake formation due to increasing electrostatic repulsion between the sludge and the membrane. In addition, as per DLVO theory, electrostatic repulsion will reduce the stability and compactness of the cake layer thus improving permeability. A decrease in protein to carbohydrate ratio of EPS (P/C ratio) was observed due to a reduction in proteins in EPS (positively charged amino acid groups) resulted from lower microorganism activity and reduced substrate adsorption on sludge. This will also result in increase of negative charge of the cake as less amino groups will be present to neutralize the negative charge of carboxyl groups and phosphoric acid groups and thus making it less sticky. A decrease in sludge viscosity from 4.3 mPa.s to 3.3 mPa.s at 0 V and 1V, respectively was observed and the sludge adsorption capacity was reduced. All these factors result in a decreasing fouling rate.

Another paper (Zhang et al., 2017) studied a system made by the integration of Microbial Electrolysis Cell with FO-AnMBR. The MEC coupled FO-AnMBR configuration aimed at providing better methane production, reducing fouling and mitigating concentration polarization. The author used stainless steel mesh as a cathode which was placed in contact with the membrane (cellulose triacetate based) and carbon brush as the anode. The applied voltage was 0.5V. It was found that the conductivity increase rate in FO-AnMBR was 0.11 mS/(cm.d) whereas in MEC coupled FO-AnMBR was 0.08 mS/(cm.d) indicating less reverse solute flux. The reverse flux of acetate ion was lower due to the repulsion created by the negatively charged cathode and to maintain charge balance of draw solution reverse flux of magnesium ion was also reduced. Application of electricity and the resultant reduction in reverse solute flux decrease the salinity accumulation which indeed offers benefits like better methane yield (11.07% more methane in biogas with MEC coupling compared to conventional FO-AnMBR), less fouling (8.93% less loosely bound EPS and 19.3% less SMP in case of MEC coupled FO-AnMBR compared to FO-AnMBR) and improved membrane flux (operating cycle prolonged to 1.3 times the simple FO-AnMBR). In addition to this MEC coupled FO-AnMBR provided 9.48% more COD removal efficiency compared to FO-AnMBR operated under the same conditions due to degradation of the organic compound at carbon anode in addition to anaerobic degradation. Moreover, the author explained that protein-based EPS and SMP in the fouling layer which cause severe membrane fouling was less in MEC coupled FO-AnMBR. This could be the result of electrostatic repulsion offered by cathode to negatively charged protein.

A similar configuration but with additional recovery mode was introduced by one study (Hou et al., 2017). FO-AnMBR coupled with the electrolysis system was operated at two modes, initially as MEC where two carbon anodes sandwiching a Ni-based cathode were used for biogas production. Later it was converted into recovery mode by placing the ionic exchange membrane between the electrodes. This configuration provided benefits that include diffusion of $NH4^+$ through the FO membrane thus improving effluent quality, phosphorous ion recovery resulting in reduced scaling, recovery of sulfate ion thus reducing methane loss due to sulfate reduction and desalination of the bulk solution resulting in less osmotic pressure loss.

During the operation, salinity build-up in MRC mode was less (0.7 to 12.7 mS/cm in 11 days) compared to MEC mode (0.7 to 17 mS/cm in 11 days). This was due to ion exchange during the recovery mode which was confirmed by an increase in conductivity of recovery solution from 1,1 to 11.25 mS/cm. Corresponding to salinity build up the flux decline was observed from 8.7 to 4 LMH. However, SEM analysis of the membrane indicated that bio-fouling accounts for this decline rather than scaling, therefore, indicating the positive impact of MEC coupling on scaling control in FO-AnMBR. The author also observed 97.3% flux recovery after membrane rinsing indicating that the reversible fouling dominates. During MRC mode, 41 to 65% of PO_4-P was recovered from the bulk at the end of each stage, which not only helped in alleviating scaling but also added to nutrient recovery.

Moreover, the system provided the current generation with average coulombic efficiency of 40% and provided a methane yield of 0.15 L CH_4/g COD. It can be concluded that the coupling of MEC and FO-AnMBR systems could be advantageous not only in terms of scaling mitigation and salinity control but also for nutrient and energy recovery. However, further research is needed to develop technology at a larger scale and wider operating conditions.

3.6. Anaerobic Electrochemical Membrane Bioreactor (AnEMBR)

AnEMBR combines MEC with AnMBR is such a way that the membrane act as one of the electrodes and serves in both hydrogen generation and effluent filtration. This novel technique uses a polymer-based hollow fiber membrane hybridized with an electrically conductive material such as Ni (Sapireddy et al., 2019; Katuri et al., 2014), graphene (Werner et al., 2016) or carbon nanotubes (Yang et al., 2019; Yang et al., 2018). The application of the voltage in this configuration has not only improved hydrogen generation but also improves the fouling reduction.

Yang et al., (2018) studied the fouling mechanism and performance of AnEMBR that uses carbon nanotubes HF membrane as cathode and titanium mesh as the anode. It was found that the application of voltage has a benefit in membrane fouling reduction. TMP in the case of AnEMBR (applied voltage = -1.2V) was 35 kPa after 30 days of operation which was much lower compared to the control reactor which observed 60 kPa (conventional AnMBR). It was explained that these results were observed because negative potential repelled the negatively charged pollutants from the membrane reducing the potential of cake formation. The electric field also aided in the destruction of high molecular weight molecules and prevented the formation of complex cross-links between organics and divalent metal ions thus alleviating fouling. Moreover, AnEMBR showed better performance, 98% COD removal compared to 95% COD removal in conventional AnMBR. Better performance can be explained by the fact that the creation of electro-active biofilm on cathode surface improved organic degradation and electric potential might have accelerated the biological degradation of organic matter.

Another study (Yang et al., 2019), compared the AnEMBR with and without voltage application with a conventional AnMBR and found out similar results for COD removal and TMP i.e. in the case of electro-assisted AnEMBR (-1.2 V applied voltage). COD removal was higher than 95% and TMP was 35 kPa, whereas for AnEMBR and AnMBR COD removal was less than 95% and TMP was 50 kPa and 60 kPa, respectively. Thus, confirming that the application of voltage can not only alleviate fouling but can also improve COD removal efficiency based on the above-mentioned reasons.

It further investigated the EPS content of fouled membrane to study fouling and found that protein EPS (almost 180 mg/L compared to >180 mg/L) and carbohydrates EPS (almost 150 mg/L compared to >150 mg/L in AnEMBR without voltage application and >250 mg/L in AnMBR) were lower in electro-assisted AnEMBR. Lower EPS prevents the formation of the gel layer, that results in severe fouling, since sticky protein EPS is limited in quantity. Lower protein EPS means lower positively charged amino acids and thus better repulsion by negative electric field thus reduction in sludge deposition. It was also observed that the application of voltage increases the CH_4 generation (an increase of more than 40 mL/g VSS d in methane yield compared to the other two reactors) by enhancing the growth of microorganism over time thus improving volatile fatty acid's degradation and improving activity of methane-producing microbes. Higher methane recovery had a very positive effect on the overall energy balance of the system and maximum surplus energy of 51.46 kJ/day was calculated in the system.

Other studies (Werner et al., 2016; Katuri et al., 2014) also found that increase in applied voltage in AnEMBR systems help in fouling mitigation. At higher voltage, hydrogen production is increased (Figure 5) which acts as an additional scouring agent and reduces fouling. Increased hydrogen production was because at higher voltage COD removal was faster (due to high current density) hence cycle time was slower, so hydrogen was dominant because the rate of hydrogen production was higher than the rate of hydrogen consumption by methanogenic bacteria.

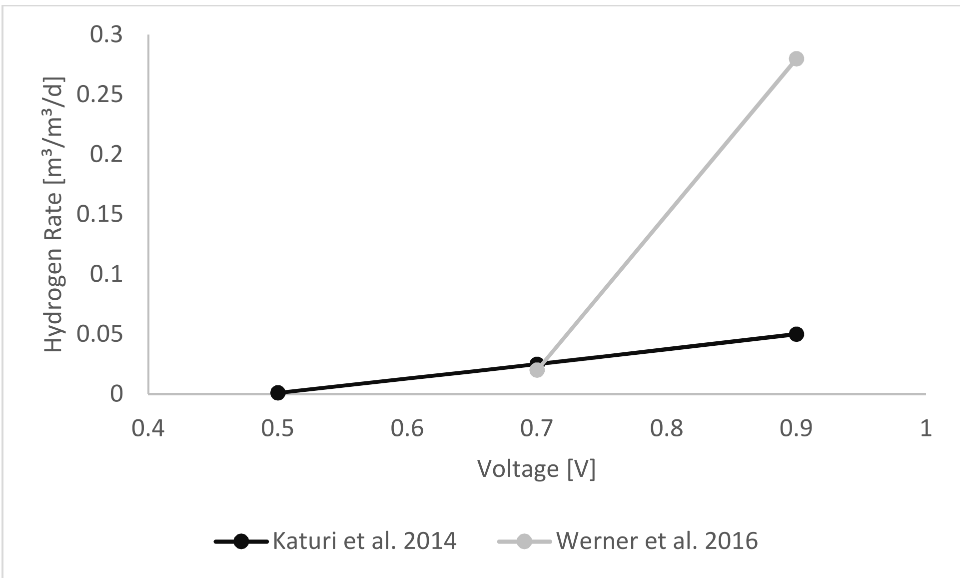

Figure 5 Hydrogen Rate at Different Applied Voltage

One research (Sapireddy et al., 2019) demonstrates the effect of cathode surface area by operating three AnEMBR, for acetate removal, with Ni-based Hollow fiber membrane as the cathode, of specific cathode surface area (SCSA) of 2 m^2/m^3, 4 m^2/m^3 and 8 m^2/m^3 respectively. The applied voltage was 0.7 V and the flux of 16 LMH. Acetate removal was almost similar in the case of all three AnEMBR and range between 77 to 94% throughout the operation. Coulombic efficiency is higher at the beginning of operation when hydrogen is dominant (cathode recovery of hydrogen being 77.00 ± 3.05% and that of methane being 0.82

± 0.26% in case of SCSA of 8 m^2/m^3) in gas and decreases as methane generation (cathode recovery of hydrogen being 24 ± 2.29 % and that of methane being 16.9 ± 1.02 % in case of SCSA of 8 m^2/m^3) increases. Hydrogen being an electron donor account for this behavior.

In terms of biofouling AnEMBR with SCSA of 8 m^2/m^3 showed the best performance with the lowest TMP. Higher hydrogen flux and increased bubble frequency with more proportion of small-sized bubbles are responsible for this behavior. Hydrogen was found to be more efficient in TMP control because of its faster evolution (less time required for molecular diffusion), higher gas velocity due to less density (0.0898 kg/m^3) than methane (0.656 kg/m^3) and higher gas volume since one mole of acetate produces 4 moles of hydrogen and one mole of methane. Higher bubble frequency produced more scouring effect due to increased bubble velocity and thus result in lower TMP. Moreover, smaller the size of the bubble better will be the scouring since small bubbles have higher slip velocity (velocity of gas relative to liquid) and shear force.

3.7. Vibration application in AnMBR

Vibratory shear enhancement process (VSEP) was first commercially introduced in flat sheet membranes by Armando and Culkin (New Logic International Inc, 1992). Shear forces generated by vibrations result in increased turbulence which enhances the removal of cake as the forces that were induced by shear dominated the physicochemical interactions between membrane and foulants and reduce cake layer fouling. Vibrations have a limited effect on the small foulants that can adsorb inside pores and cause pore blocking. However, since in AnMBR treating domestic wastewater cake layer fouling phenomenon dominates (Chen et al., 2014) use of vibrations can serve as a good mitigation technique. There are two ways of vibrations generation; shaker generated oscillations in liquid with a stationary membrane and/or vibrating or rotating membrane on a rotor where the liquid is held stationary (Kola et al., 2014). One study (Kola et al., 2014) demonstrated the effectiveness of transverse membrane rotation in HF AnMBR with increasing MLSS. At increased MLSS, conventional methods of fouling mitigation like gas sparging are not effective because increased viscosity greatly reduces the velocity of rising bubbles and thus reduces shear force. It was observed that at a vibration frequency of 6.7 Hz, a constant flux of 26 LMH could be maintained with increase in MLSS from 0.005 g/L to 5 g/L which in the absence of vibration decreased from 16 LMH (0.005 g/L MLSS) to 1 LMH (5 g/L MLSS). This indicated that vibrations produce enough shear to counter fouling even with increasing MLSS. Increasing the frequency of vibrations from 6.7 Hz to 20 Hz effects the hydrodynamic conditions in the reactor resulting in more turbulence (Reynolds number increases from 365 to 1095 respectively) which creates the higher shear rate i.e. (1372 1/s to 30108 1/s) thus reducing fouling and increasing critical flux from 26 LMH to 51 LMH. Additionally, when compared with gas sparging and cross velocity methods at a constant flux of 30 LMH and MLSS of 26 mg/L, the time required to reach a TMP of 60 kPa in case of vibration application was 3 times more. Similar results were observed by others (Ruigómez et al., 2016a) when they compared rotation with gas sparging and found that in case of rotation better cake redispersion was observed which improved the cycle duration by 10 times compared to gas sparging.

Ruigómez et al., (2016a) observed that the fouling resistance drop from 0.16 kPa/s to 0.01 kPa/s with the membrane rotation of 0 to 260 rpm respectively at a permeate flux of 14 LMH. They also found that the turbulence promotor efficiency (defined as the decrease in fouling resistance compared to initial value with mitigation technique employed) for membrane rotating at 260 rpm was 96% compared to gas sparging with specific gas demand of 3.8 $m^3/(m^2.h)$ which has the turbulence promotor efficiency of 44.4% at a flux of 8 LMH in both the cases. This difference was observed because, in the case of membrane rotation, vibration energy is more uniformly distributed compared to sparging thus provides better shear. Another study (Mertens et al., 2019) also found improved fouling mitigation (10 times less fouling resistance at a flux of 20 LMH) by increasing shear rate from 0 s^{-1} to 605 s^{-1} in their magnetically induced membrane vibration (MMV) AnMBR system. They also found out that with a system of 4 modules, the contribution of the MMV system to total energy was only 12% of total consumption.

It was found that by increasing vibration from 0 Hz to 13.3 Hz, the critical flux increase by 3.7 times. However, the critical flux at 20 Hz was only 24 % of that at 13.3 Hz (Kola et al., 2014), which is because, at higher vibrations, shear forces dominate Brownian, inertial and drag forces and thus causing particles to be moved away from the membrane surface. As a result of this, pore-blocking (on which vibrations have an insignificant effect) dominates. It is therefore important to select the vibration frequency carefully to optimize fouling mitigation with minimum energy demand.

The effectiveness of vibrations application can be further enhanced by coupling it with periodic relaxation or backwash. For the operation of 25 days (flux = 30 LMH) when AnMBR was run at a vibrational frequency of 4.2 Hz coupled with relaxation or backwashing after 30 minutes intervals for 30 seconds (Kola et al., 2014) time required to reach a TMP of 60 kPa was 100 hours for relaxation and 60 hours for backwash compared to 28 hours (approximately) with vibrations alone. The authors explained that in case of relaxation, the cake layer is loosened during relaxation and then application of vibration removed foulant thus improving the performance. In the case of backwash, when the cake layer was removed or restructured during washing (which is not the case in relaxation thus the cake layer serves as a protective layer) pore-blocking phenomenon may increase since the effectiveness of backwashing was not as high as periodic relaxation. On contrary to this, one study (Mertens et al., 2019) found backwashing to be 10 times more effective than relaxation in a 10-day operating cycle at a flux of 20 LMH and shear rate of 484 1/s where backwashing (or relaxation) time was 2 minutes after 8.5 minutes. Similarly, another study (Ruigómez et al., 2016a) conducted experiments with a filtration cycle of 12.7 minutes and cleaning duration of 30 s at a rotational speed of 180 rpm and permeate flux of 8 LMH and found out that the internal fouling resistance is less in case of backwashing (11% of total) compared to that in case of relaxation (15% of total). They also coupled both backwashing and relaxation and found that optimal results were obtained at a backwashing ratio of 0.67 and a specific TMP of 20 kPa. Under these conditions, reversible fouling resistance was 41% of the total and internal fouling resistance was 12% of the total. Moreover, the highest value of dispersed TSS (solid concentration of ~6.5 gSS/m^2) was obtained under these operating conditions.

From this discussion, we can conclude that the application of vibration in AnMBR operation can serve as an effective fouling mitigation technique especially if coupled with other cleaning techniques like backwashing and relaxation. However, the best-coupled combination is case dependent as it is controlled by many factors like cycle duration, permeate and backwashing flux, days of operation and cleaning time. Therefore, it is important to develop the best combination, experimentally, for employed operating conditions. To further understand the sustainability of this technique, studies must be conducted developing understanding of fouling behaviors and establishing energy balances to see the energy efficiency of vibrational systems compared to other available mitigation techniques.

3.8. Biological mitigation – Quorum Quenching

Quorum sensing (QS) is a communication technique between the cells that is used by microorganisms to control their activities like attachment or detachment, EPS formation, biofilm production and mobility. The communication is done by a generation of signal molecules that are called autoinducers e.g. N-acryl homoserine lactone (AHL). Deactivation of autoinducers results in disruption of communication between the cells and this process of deactivation is called Quorum Quenching (QQ). QQ is carried out by using QQ enzymes or bacteria that can be isolated from the sludge (Aslam et al., 2018). QQ enzymes are divided into two main types i.e. AHL-Lactonase and AHL-acylase. The former having a cleavage effect on the ester bond of the lactose ring and later breaks the amide bond. Additionally, the oxidoreductase enzyme also functions as a QQ enzyme by converting AHL to 3-hydroxy AHL thus preventing signal molecule to form bio-film (Kim et al., 2014a). Enzymatic QQ has some disadvantages such as high cost, instability and need for purification. These are overcome by the employment of QQ bacterial (Id et al., 2018). Kim et al. (2014a) have isolated 225 AHL degradation bacteria from waste sludge.

Many studies in the literature have proved the effectiveness of quorum quenching in aerobic MBRs where quorum quenching bacteria has been employed in carriers like QQ beads, QQ cylinders, QQ sheets or QQ vessels (Ergön-can et al., 2017; Hyun et al., 2017; Won et al., 2016; Weerasekara et al., 2014). However, not much work has been done so far in AnMBR. The first study that has tested the effectiveness of QQ in AnMBR was published in 2019 (Liu et al., 2019) followed by two more in 2020 (Xu et al., 2020a, 2020b).

Liu et al., (2019) tested four different bacterial species by entrapping them in alginate beads and then selected microbacterium sp. (a facultative anaerobe) for long term study due to its higher COD removal efficiency and higher methane production. QQ bacteria were tested in a sequence of three phases i.e. without beads (control), with empty beads (EB) and with QQ beads (QQB) respectively (Liu et al., 2019). Membrane flux was kept at 8 LMH and HRT of 18.5 hours was used. COD removal efficiency during all three phases was close (97.98% for control, 98.84% for EB and 98.75% for QQB) however there is a significant difference in time required to reach TMP jump which is an indicative phenomenon for biofilm and bio-cake structural change and represent fouling in AnMBR (Zhang et al., 2006). The time requires to reach TMP in three phases was 6, 8 and 45 days, respectively. Slight improvement in the EB phase was probably due to the scouring effect. These results show the potential of

QQB in fouling control in AnMBR. The effectiveness of the QQB phase was described by the analysis of AHLs concentration in the fouling layer that showed that the AHLs concentration (responsible for biofouling) in the QQB phase was lower than the control phase.

Authors (Liu et al., 2019) also observed a significant reduction in EPS content (responsible for cake fouling (Chen et al., 2017a)) i.e. 15.18% reduction in the bulk phase and 75.32% reduction in the foulant layer. The protein content of EPS which is responsible for sticky cake fouling (Chen et al., 2017a) was also reduced significantly compared to the control phase (80.98% reduction). Although the QQ was found very effective in combating fouling, the AHLs degradation by single QQ species declined after 45 days. Xu et al., (2020a) proposed the use of a facultative QQ consortium and study its effect of AHLs degradation and fouling control in AnMBR.

Xu et al., (2020a) studied three facultative QQ consortium consisting majorly of Proteobacteria (abundance > 90%), Firmicutes and Actnobacteria phylum isolated from activated sludge obtained from a local domestic wastewater treatment plant. All three consortiums were successful in diminishing multiple AHLs (acyl chains ranging from C4 to C10) that are responsible for biofouling in AnMBRs. The degradation rate in all cases was above 80% whereas without QQ the maximum rate obtained was 28%. It was also found that bead entrapped FQQ shows higher alleviation in EPS production (both carbohydrates and proteins) compared to free one signifying the importance of immobilization in protection against the harsh environment. FQQ with 6 carbon chain (FQQ-C6) showed the best performance in protein (72.3%) and carbohydrate (66.53%) reduction indicating its potential to mitigate fouling in AnMBR. When bead entrapped FQQ-C6 was used in AnMBR with a ceramic membrane of pore size 0.1 µm, 30 ºC temperature, gas sparging rate of 0.7 L/min, 30 days SRT and 17 hours HRT, a total COD removal 91% and methane yield was 0.07

$L/gCOD_{removed}$ indicating no negative effect of performance efficiency. Moreover, the presence of QQ increased the average operating period of filtration cycles by 75%, indicating efficient fouling mitigation. All these results showed the potential of QQ for fouling mitigation in AnMBR however, extensive research is required to understand the potential of long-term operation and scale-up.

4. Future perspective

AnMBR technologies allow positive energy treatment of domestic wastewater with optimization of operating conditions and fouling control strategies. Innovative mitigation techniques and process configurations are trying to resolve the issues of fouling that account for the biggest chunk of energy demand. Recent literature on lab-scale studies has suggested interesting and innovative solutions. Hybrid Processes like G-AnMBR, cell entrapment AnMBR, FO-AnMBR, MEC-AnMBR and dynamic AnMBR have shown lower fouling rate and higher % COD removal and methane yield than conventional AnMBR (Table 6). However, these are in very early stages of development and for scale-up, there is a need for an extensive study of the fouling mechanism, change in this mechanism with changing conditions and energy balance of the whole process. It is also important that the results of lab-scale studies should be validated at a larger scale and long-term studies should be conducted to broaden the range of practical application.

The application of QQ in AnMBR showed promising results (Table 6) both in terms of energy-saving and production, fouling control and % COD removal. It will be interesting to apply QQ in other configurations e.g. FO-AnMBR, AnDMBR, etc. and evaluate technical and economic feasibility. Another fascinating approach can be the coupling of the electric field or vibrations with QQ and study their effect on fouling control. However, these technologies require extensive research to understand and express their full potential.

Moreover, methane loss, especially under psychrophilic conditions (10-30 °C) is an area that requires more research covering the maximization of methane yield and minimization of methane leaks in the dissolved phase. Integration of methane recovery units (like membrane degassing units), improvement of membrane materials and reduction of operating energy should be given attention.

Other research areas that can positively contribute towards fouling mitigation in AnMBRs and help in making them water resource recovery facilities may include,

- Improved membrane materials with anti-fouling capabilities;
- Coupling of different hybrid processes to combine benefits and minimize disadvantages, for example, the coupling of MEC with FO-AnMBR or Vibrations with different hybrid AnMBRs;
- Post-treatment step integration with AnMBR for nutrient recovery. They may include adsorption, ion exchange, microalgae cultivation and forward or reverse osmosis (Shahid et al., 2020);
- Studies involving complete plant energy assessment to analyze the effect of the secondary operation, e.g. DS re-concentration in FO-AnMBR, methane recovery unit for psychrophilic AnMBRs, entrapment procedures and sludge modification techniques, on net power generation of the process;
- Life cycle assessment and cost-benefit studies of processes to evaluate the environmental impacts and implications.

5. Conclusion

Membrane fouling is a serious concern in the treatment of domestic wastewater treatment using AnMBR. At the pilot-scale, biogas sparging is the most employed fouling mitigation strategy but it accounts for a huge percentage of total energy requirements. Membrane material, membrane module, reactor configuration, operating conditions and sludge properties contribute towards fouling behavior as well as net energy requirements of AnMBR. Some hybrid processes have been proved to successfully reduce fouling problems of AnMBR and could be more suitable for efficient resource recovery. Additionally, applications of the electric field, voltage and vibrations have contributed positively towards fouling control. However, increasing their value beyond a certain limit will not only reduce performance efficiency but also unnecessarily increase the energy demand. Biological techniques like quorum quenching are found to be equally successful in fouling mitigation in AnMBR as they were in AeMBR and have good potential in this area especially if coupled with other techniques of fouling mitigation.

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Table 1. Description of different AnMBR configurations for domestic wastewater treatment (GSP: Gas Sparging, PSP: Particle Sparging, FS: Flat sheet, HF: Hollow Fiber, UF: Ultrafiltration, MF: Microfiltration, RMEM: Rotating membrane).

N°	**Configuration**	**Membrane Module**	**Material**	**COD (% removal)**	**Organic Loading Rate ($kg_{COD}/(m^3.d)$)**	**References**
Config-1	GSP-FS	**FS UF**	(PES)	>90	0.6 - 1.1	(Martinez-Sosa et al., 2011)
Config-2	GSP-HF1	HF UF	PURON (Polyester)	87	1.03-2.11	(Giménez et al., 2014)
Config-3	GSP-HF2	HF UF	PURON (Polyester)	85	0.3-1.1	(Robles et al., 2012)
Config-4	GSP-HF3	HF UF	PURON (Polyester)	85	1.25-2.24	(Robles et al., 2013)
Config-5	GSP-HF4	HF UF	PURON (Polyester)	>90	0.25-1.24	(Giménez et al., 2014)
Config-6	GSP-HF5	**HF MF**	RF-1 (PVDF)	87	3	(Mei et al., 2017)
Config-7	GSP-HF6	HF UF	ZeeWeed 500 (PVDF)	93.7	1.1	(Dong et al., 2015; Shin & Bae, 2018)
Config-8	GSP-HF7	HF UF	ZW-10 Zenon (PVDF)	87	1.2 - 1.44	(Gouveia et al., 2015a)
Config-9	GSP-HF8	HF UF	ZW-10 Zenon (PVDF)	90	1.6 - 2.0	(Gouveia et al., 2015b)
Config-10	GSP-HF9	HF UF	GE ZeeWeed 500 (PVDF)	93	3.77-4.97	(Dong et al., 2016)
Config-11	GSP-HF10	HF UF	W-10 Zenon, GE (PVDF)	>90	0.8-1.8	(Evans et al., 2019)
Config-12	**PSP-HF**	HF UF	Cheil Industries (PVDF)	>90	NA	(Shin et al., 2014)
Config-13	**RMEM-HF**	HF UF	GE Water & Process Tech (PVDF)	91	NA	(Ruigómez et al., 2016b)

Table 2. Description of different parameters in various AnMBR configurations (IG-AnMBR: Internally submerged granular AnMBR; ES-AnMBR: Externally submerged membrane bioreactor)

Configuration	Membrane Configuration	SS or VSS (g/L)	Days of Operation	GSI_m ($Nm^3/(m^2.h)$)	SRT (d)	HRT (h)	References
Config-1	ES-AnMBR	15-21 gSS/L	100	1.22	NA	19.2	(Martinez-Sosa et al., 2011)
Config-2	ES-AnMBR	22 gSS/L	70	0.23	70	6-20	(Giménez et al., 2011)
Config-3	ES-AnMBR	10-30 gSS/L	730	0.17-0.5	30-70	6-36	(Robles et al., 2012)
Config-4	ES-AnMBR	7-32 gSS/L	365	0.23	70	24.5-5.5	(Robles et al., 2013)
Config-5	ES-AnMBR	10-25 gSS/L	172	0.23	28.6-41.1	12.1-28.4	(Giménez et al., 2014)
Config-6	ES-AnMBR	4.7-20.1 gSS/L	340	0.31	NA	2.2	(Mei et al., 2017)
Config-7	ES-AnMBR	4 gSS/L	90	0.15	70	8.5	(Dong et al., 2015)
Config-8	ES-AnMBR	6 gVSS/L	1095	0.81–1.22	Infinite	7	(Gouveia et al., 2015a)
Config-9	ES-AnMBR	0.9-16.1 gVSS/L	1095	0.16–0.32	NA	12.8-14.2	(Gouveia et al., 2015b)
Config-10	ES-AnMBR	5-15 gSS/L	536	NA	40-70	8.5	(Dong et al., 2016)
Config-11	IG-AnMBR	4.30-7.54 gVSS/L	300	NA	60 ± 27	11 ± 3	(Evans et al., 2019)
Config-12	IG-AnMBR	0.600-1.2 gSS/L	485	NA	6.2-36	4.6-6.8	(Shin et al., 2014)
Config-13	ES-AnMBR	21.3 gSS/L	270	NA	270	33	(Ruigómez et al., 2016b)

Table 3. Water Treatment energy consumptions of different AnMBR configurations having different COD removal, critical flux and transmembrane pressure

Configuration	Fouling Control Energy (kWh/m3)	Critical Flux (LMH)	Total Energy (kWh/m^3)	TMP (kPa)	References
Config-1	1.28	7	1.66	17.7	(Martinez-Sosa et al., 2011)
Config-2	0.2	10	0.26	8.0	(Giménez et al., 2014)
Config-3	0.2	12-16	0.26	40.0	(Robles et al., 2012)
Config-4	0.2	10-13.3	0.26	<10.0	(Robles et al., 2013)
Config-5	0.19	7-11	0.24	<10.0	(Giménez et al., 2014)
Config-6	0.5	6	0.64	6.0	(Mei et al., 2017)
Config-7	0.08	17	0.1	8.8	(Dong et al., 2015)
Config-8	0.19-0.5	10-14	0.25-0.65	5.0-55.0	(Gouveia et al., 2015a)
Config-9	0.04-0.1	12-14	0.05-0.13	40.0-55.0	(Gouveia et al., 2015b)
Config-10	0.09	25-27	0.11	1.5-30	(Dong et al., 2016)
Config-11	0.09-0.27	7.6-7.9	0.1-0.3	NA	(Evans et al., 2019)
Config-12	0.1	4.1-7.5	0.13	10.0-27.0	(Shin et al., 2014)
Config-13	0.23	10	0.3	1.0-2.5	(Ruigómez et al., 2016b)

Table 4. Effect of Pore Size on Flux and % COD Removal in AnDMBR

Configuration	Module	Support Material	The average Pore size of the support (µm)	Operating Flux (LMH)	% COD Removal	References
Internally Submerged	Flat Sheet	Polypropylene	10	2.2	99.5	(Ersahin et al., 2017)
Side Stream	Tubular	Nylon	61	31.25	-	(Siddiqui et al., 2018)
Internally Submerged	Flat Sheet	Nylon	75	22.5	75-90	(Hu et al., 2018b)
Internally Submerged	Hollow cylinder	Polyamide nylon	150	100	80	(Wang et al., 2018)

Table 5. Performance of Different FO-AnMBRs and AnMBRs treating domestic wastewater (FS: Flat sheet, HF: Hollow Fiber).

Configuration	Module	Temperature (°C)	Feed COD (mg/L)	Methane Yield (L CH_4/g COD)	COD % Removal (%)	References
Internally Submerged FO-AnMBR	FS	25	460	0.21	96.7	(Chen et al., 2014)
Internally Submerged FO-AnMBR	FS	35	460	0.25-0.3	95	(Gu et al., 2015)
Internally Submerged FO-AnMBR + MEC + MRC	FS	25	270±10	0.15	>93	(Hou et al., 2017)
Externally Submerged AnMBR	HF	33	445	0.07*	87	(Giménez et al., 2011)
Externally Submerged AnMBR	FS	35-20	630	0.27	>90	(Martinez-Sosa et al., 2011)

Table 6. Summary of Different Novel Configurations in AnMBR

Configuration	% COD Removal (%)	Fouling Rate (Pa/s)	Methane Yield (L CH_4/g COD)	Membrane Flux (LMH)	Organic Loading Rate ($kg_{COD}/(m^3.d)$)	Reference
AnMBR	90	3.33	0.27	7	0.6 - 1.1	(Martinez-Sosa et al., 2011)
G-AnMBR	92	0.005	0.16	7	-	(Chen et al., 2017b)
Entrapped Cell AnMBR	85	0.057	0.08	10.63	0.57	(Juntawang et al., 2017)
AnEMBR	98	0.014	-	23.22*	0.5	(Yang et al., 2018)
MEC-AnMBR	71	-	-	1.8**	5	(Ding et al., 2018)
FO-AnMBR	96.7	0.21	-	6.5**	0.46	(Chen et al., 2014)
Dynamic AnMBR	90	-	0.21	7	0.37	(Li et al., 2016)
AnMBR with Vibrations	-	0.11	-	26***	-	(Kola et al., 2014)
AnMBR with QQ	98.8	0.024	0.34	8	-	(Liu et al., 2019)

*Average flux provided per unit TMP

**Average value

***Critical flux at a frequency of 6.7 Hz

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