Advances in anaerobic membrane bioreactor technology for municipal wastewater treatment: A 2020 updated review

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ABSTRACT

The application of anaerobic membrane bioreactors (AnMBR) for mainstream municipal sewage treatment is almost ready for full-scale implementation. However, some challenges still need to be addressed to make AnMBR technically and economically feasible. This article presents an updated review of five challenges that currently hinder the implementation of AnMBR technology for mainstream sewage treatment: (i) membrane fouling, (ii) process configuration, (iii) process temperature, (iv) sewage sulphate concentration, and (v) sewage low organics concentration. The gel layer appears to be the main responsible for membrane fouling and flux decline being molecules size and morphology critical properties for its formation. The review also discusses the advantages and disadvantages of five novel AnMBR configurations aiming to optimise fouling control. These include the integration of membrane technology with CSTR or upflow digesters, and the utilisation of scouring particles. Psychrophilic temperatures and high sulphate concentrations are two other limiting factors due to their impact on methane yields and membrane performance. Besides the methane dissolved in the effluent and the competition for organic matter between sulphate reducing bacteria and methanogens, the review examines the impact of temperature on microbial kinetics and community, and their combined effect on AnMBR performance. Finally, the review evaluates the possibility to pre-concentrate municipal sewage by forward osmosis. Sewage pre-concentration is an opportunity to reduce the volumetric flow rate and the dissolved methane losses. Overall, the resolution of these challenges requires a compromise solution considering membrane filtration, anaerobic digestion performance and economic feasibility.

HIGHLIGHTS

- Five main challenges were identified for mainstream AnMBR implementation.
- Membrane fouling appears influenced by molecules size and morphology.
- New AnMBR configurations are gaining attention to improve fouling control.
- Psychrophilic temperatures and influent sulphate are limiting factors for AnMBRs.
- Sewage pre-concentration could improve the applicability of AnMBR technology.

KEYWORDS: Anaerobic digestion; Anaerobic membrane bioreactor (AnMBR); Forward osmosis (FO); Fouling; Water Resource Recovery Facility (WRRF); Municipal sewage

Abbreviations

AeMBR, aerobic membrane bioreactor; AFBR, anaerobic fluidised bed reactor; AFMBR, anaerobic fluidised membrane bed reactor; AnMBR, anaerobic membrane bioreactor; AnDMBR, anaerobic dynamic membrane bioreactor; AnMF-OMBR, anaerobic osmotic membrane bioreactor coupled with microfiltration; AnOMBR: anaerobic osmotic membrane bioreactor; BSA, bovine serum albumin; CAS, conventional activated sludge; CFV, cross-flow velocity; COD, chemical oxygen demand; CSTR, completely stirred tank reactor; DAMO, denitrifying anaerobic methane oxidation; DLVO, Derjaguin-Landau-Verwey-Overbeek; EGSB, expanded granular sludge bed; EPSs, extracellular polymeric substances; FO, forward osmosis; GAC, granular activated carbon; Gl-AnMBR, Gas-lift anaerobic membrane bioreactor; HRT, hydraulic retention time; LMH, liters per square meter per hour; MF, microfiltration; MLSS, mixed liquor suspended solids; OLR, organic loading rate; PAC, powdered activated carbon; PET, Polyethylene terephthalate; PVDF, polyvinylidene fluoride; RO, reverse osmosis; SGD, specific gas demand; SMP, soluble microbial products; SRB, sulphate-reducing bacteria; SRT, solids retention time; TMP, transmembrane pressure; UASB, upflow anaerobic sludge blanket; UF, ultrafiltration; WW, wastewater; WWTP, wastewater treatment plant.

1. Introduction

Climate change and resource depletion are pushing a paradigm shift in municipal wastewater management from end-of-pipe treatment towards integrated resource recovery [1]. New schemes consider wastewater as a source of energy, nutrients and water rather than as a source of pollution [2,3].

Anaerobic digestion represents a more sustainable technology to manage the organics contained in wastewater than the conventional activated sludge (CAS) treatment [4,5]. The CAS process is an energy-intensive treatment, accounting for more than 50% of the total energy consumption of a typical wastewater treatment plant (WWTP) [6,7]. Paradoxically, this energy is spent into converting organic matter into CO_2 and poorly biodegradable microbial mass [8]. Alternatively, anaerobic digestion provides several advantages such as renewable methane energy production, lower biomass production and no aeration requirements [9,10]. Anaerobic digestion is an emerging technology for municipal wastewater treatment.

Upflow anaerobic sludge blanket (UASB) and expanded granular sludge bed reactors (EGSB) are the most important anaerobic technologies for wastewater treatment [11,12]. The competitive advantage of these technologies (also known as high-rate anaerobic reactors) is due to the retention of biomass in the reactor that allows decoupling the hydraulic retention time (HRT) from the solids retention time (SRT) [11,13]. The full-scale application of UASB technology as mainstream sewage treatment has been applied in warm climates such as Brazil, India and the Middle East [14]. However, in many applications, the performance of full-scale UASB plants treating municipal sewage is suboptimal with chemical oxygen demand (COD) removal efficiencies around 60% [14–16]. This has been attributed to poor operating and maintenance procedures as well as to improper design [15]. Consequently, the UASB process can generate effluents with a high biodegradable organic matter concentration which may require aerobic post-treatment [17].

Anaerobic membrane bioreactor (AnMBR) technology overcomes the limitations of UASB technology and further improves the competitiveness and applicability of anaerobic systems as mainstream process for municipal sewage treatment [18–20]. Both ultrafiltration (UF) and microfiltration (MF) membranes enable a complete decoupling of hydraulic retention time (HRT) from solids retention time (SRT), which allows high controllability of the biomass in the digester, while obtaining a high-quality effluent free of suspended solids and pathogens [21,22]. The higher quality effluent is a competitive distinctive advantage of AnMBR over UASB technology [20,23–25]. However, a certain level of post-treatment is required since mainstream AnMBR effluents do not comply with the nutrients (i.e. N, P, S) discharge limits [26,27]. It should be noted that the concentration of nutrients in AnMBR effluents is typically higher than in their influent as a result of organic matter degradation [22].

As for 2019, several pilot-scale and demonstration plants equipped with AnMBR technology have been satisfactorily operated to treat municipal sewage [28]. Many studies have demonstrated that mainstream AnMBR application could make a WWTP energy neutral or even positive due to the potential energy production in the form of methane-rich biogas [17,29–32]. However, some technological challenges need to be addressed to make AnMBR a technically and economically competitive alternative for municipal sewage treatment.

Most of the operational and technical challenges of AnMBR technology (e.g. membrane fouling, reactor configuration, operational conditions, dissolved methane) have been previously discussed in literature reviews devoted to AnMBR technology [9,21,22,27,33–

39]. However, the resolution of these challenges is complex and includes a broad range of considerations that require a compromise solution considering membrane filtration, anaerobic digestion performance and economic feasibility. In this literature review, the most important challenges associated with mainstream AnMBR technology are discussed to support its implementation now that AnMBR is getting closer for full-scale commercial application. Specifically, this review discusses the implications of membrane fouling, process configuration, temperature, influent sulphate concentration and sewage pre-concentration on AnMBR performance and economic feasibility to clarify and simplify the decision-making process.

Membrane fouling is widely recognised as the main barrier for a widespread application of AnMBR technology [35,40]. However, despite extensive research, the mechanisms underlying this phenomenon are yet to be unfolded. Membrane fouling occurrence has been linked to several factors such as operational conditions (e.g. HRT, SRT, temperature), biomass characteristics (e.g. type of foulants, size of foulants) and membrane characteristics (e.g. material, pore size, type) [37,41–43]. The operational challenges associated with membrane fouling have resulted in the development of a wide range of physical and chemical cleaning strategies (e.g. backwashing, relaxation cycles, chemical reagents) and fouling control methods [27,44–46]. The strategy used to control membrane fouling is particularly important since it represents the main operational cost of AnMBR [28,47]. To date, gas sparging is the most used method for fouling control for submerged membranes [48–51]. However, novel AnMBR configurations are gaining attention to partially or totally replace gas sparging and further optimise AnMBR treatment [35,39]. This review provides a holistic updated understanding of the causes and implications of membrane fouling (Section 2) as well as an in-depth analysis of the most promising AnMBR configurations (Section 3).

Temperature is one of the most important process variables in anaerobic digestion systems due to its impact on metabolic kinetics and equilibrium constants. The application of AnMBR technology in temperate and cold climates has been identified as possible but challenging [52,53]. In these climates, the low concentration of organics in municipal sewage makes psychrophilic conditions (< 20 ºC) the only energetically feasible option. However, the lower process kinetics under psychrophilic conditions imply higher retention times and, therefore, higher capital costs. Moreover, the higher amount of methane dissolved in the effluent at lower temperatures is especially worrisome considering that the global warming potential of methane is 34 times higher than CO_2. [54]. Consequently, developing strategies to maximise the recovery of dissolved methane is essential to increase methane yield and reduce greenhouse gas emissions of AnMBR technology (Section 4).

The presence of sulphate in municipal sewage is another important constraint for the feasibility of AnMBR technology since sulphate-reducing bacteria (SRB) reduce sulphate to sulphide oxidising COD to CO_2 [55,56]. The lower amount of COD available for methanisation and the formation of sulphide could compromise the economic feasibility of the process [18,57]. Sulphide is a corrosive compound which has been reported to affect membrane performance by reducing its fluxes and durability. Concomitantly, the presence of hydrogen sulphide in the biogas increases the capital cost due to the need to use corrosive resistant equipment and piping as well as to implement measures for odour control. Sulphate-rich municipal sewage may require the implementation of a desulphurisation unit to reduce the biogas hydrogen sulphide concentration [58]. Therefore, the sulphate concentration of sewage is an important parameter for the design, profitability and operation of AnMBR (Section 5).

The high volumetric flow rate and the low concentration of organics in municipal sewage limit the applicability of AnMBR technology owing to the higher capital and operating expenditures, the higher amounts of dissolved methane lost through the effluent and the lower methane production per volume of wastewater treated. In this regard, municipal sewage pre-concentration by different membrane technologies has been considered to overcome these limitations, including direct membrane filtration, dynamic membrane filtration and forward osmosis (FO) [59]. In particular, FO stands as a promising technology to pre-concentrate municipal sewage with low energy inputs [60–63]. However, little attention has been given to this approach in previous AnMBR reviews. Municipal sewage pre-concentration opens new windows of opportunity for future AnMBR applications (Section 6).

This publication critically reviews five challenges that limit the applicability of AnMBR technology for municipal sewage treatment. These five challenges are: (i) membrane fouling, (ii) process configuration, (iii) process temperature, (iv) sewage sulphate concentration, and (v) sewage low organics concentration. For each challenge, novel knowledge and updated solutions proposed in the literature are critically analysed and future research needs are identified.

2. Membrane fouling mechanisms

Membrane fouling is the main cause for membrane flux decline over time. This is critical since membrane flux determines the membrane area required, which has a direct impact on both capital and operating expenditures. Additionally, membrane fouling leads to complex cleaning protocols that hinder the operability of the process due to more frequent backwash cycles and chemical cleanings. In the last years, significant advances on fouling control have been made including new AnMBR configurations [28,64], optimisation of operational conditions [29,65], gas sparging optimisation [48,49] and improvements on backwash and chemical cleaning protocols [44,66]. However, most research has focused on reducing fouling rather than on understanding the underlying fouling formation mechanisms. Studies focusing on the mechanisms leading to membrane fouling are inconclusive and, in some cases, contradictory. This situation can be attributed to several interconnected factors taking part in the complex network that comprises membrane fouling.

Extracellular polymeric substances

Extracellular polymeric substances (EPSs) have been considered the primary precursors of membrane fouling [42,67,68]. EPSs can be defined as organic macromolecules that are present outside and inside microbial aggregates mainly composed of proteins, polysaccharides, and humic acids [69–71]. EPSs can be classified into (i) tightly bound EPSs (TB-EPSs), (ii) loosely bound EPSs (LB-EPSs) and (iii) soluble EPSs (sEPS) [27,72]. The latter is commonly known as soluble microbial products (SMPs) [69,73]. The adhesion forces causing the attachment of these substances on the membrane surface is challenging and remains under discussion as recently reviewed by Zhen et al. [27]. The composition of EPSs appears determinant to understand the interaction between the EPS substances and the membrane surface and, consequently, to understand the occurrence, structure and composition of membrane fouling [27,67,74].

Membrane hydrophobicity has been reported as an important factor for membrane fouling since it affects the interaction with hydrophobic and hydrophilic EPSs compounds [65,75,76]. Lin et al. [77] attributed the higher impact of protein EPSs on membrane fouling to the higher hydrophobicity of proteins compared to polysaccharides. Lin et al. [77] concluded that the EPSs protein to polysaccharide ratio was a better indicator for fouling control than the total amount of EPSs. Similarly, Arabi and Nakhla [78] reported that an EPS protein to carbohydrate ratio of 8/1 and 2/1 in the influent exhibited the highest and the lowest fouling rates, respectively. The importance of EPS proteins on membrane fouling has been reported in several publications [79–88]. However, other publications concluded that polysaccharides are the main responsible for membrane fouling [89–93]. These discrepancies can be related to multiple factors including AnMBR operational conditions, macromolecules composition, microbiome composition, influent composition, membrane configuration and properties, and the extraction and analytical methods used for EPS analysis. Nonetheless, these studies did not elucidate the EPS attachment mechanisms leading to membrane fouling.

Teng et al. [94] recently published a systematic research study concluding that small size and disperse EPSs are thermodynamically favoured to adhere on the membrane and cause fouling. This was attributed to the smaller real separation between the SMP and the membrane. Teng et al [94] also reported that SMPs morphology plays a role on membrane fouling since it controls SMP attachment to the membrane surface. This is in agreement with previous publications that indicated that membrane roughness and surface characteristics are important factors on membrane fouling [95–97]. Teng et al. [94] study is highly relevant since it shows that molecules size and morphology, rather than the composition, may control membrane fouling. However, further research is required to elucidate the relative impact of the different factors on membrane fouling.

Particle size distribution

Particle size distribution has been reported as an important factor to predict and control fouling in aerobic and anaerobic MBRs [53,65,77,80,98–101]. Particularly important appears the role of particle size distribution on back-transport mechanisms which has been highlighted as a crucial factor for foulants deposition [53,102]. Zhou et al. [103,104] demonstrated the importance of sub-visible particles (0.45-10 μm) and their associated microbial community on membrane fouling. Specifically, Zhou et al. [104] reported that micro-particles (5-10 μm) and colloidal particles (0.45-1 μm) had different microbial communities and played different roles on membrane fouling. On the one hand, micro-particles were mainly formed by filamentous microorganisms and were associated with the cake layer resistance. On the other hand, colloidal particles were mainly formed by sulphate-reducing bacteria and were linked to the initial fouling formation. However, further research is needed to better comprehend the role of particle size distribution and microbial composition on membrane fouling.

Fouling structure and composition

Different theories have been proposed to provide a reliable explanation for the role of foulants on filtration resistance. According to the literature, different fouling layers can be distinguished depending on the filtration resistance [91,105]. However, this distinction has been different for aerobic and anaerobic MBRs. In AnMBRs, it is generally accepted that pore clogging is followed by cake layer formation on the membrane surface [36,77,106], which confronts the most recent findings in aerobic membrane bioreactors (AeMBRs) [93,94]. These AeMBR studies have differentiated two distinct layers on the membrane; a gel layer and a cake layer. The gel layer is formed by the precipitation and gelation of colloidal and soluble polymeric substances (including SMPs and EPSs) on the membrane, while the cake layer is formed by the adhesion and accumulation of sludge flocs on the gel layer [105,107].

The gel layer has been reported to contain negatively charged groups such as carboxyl, hydroxyl, and phosphoric. These groups have an important role in the gelling processes and filtration resistance [69,91]. Gelling properties have been mainly attributed to carboxyl groups in polysaccharides, especially in the presence of divalent or multivalent cations in the mixed liquor [67]. Cations complexation has been reported to be an important mechanism for the formation and consolidation of a three-dimensional gel matrix on the membrane surface [90,105]. In AeMBR, Teng et al. [94] reported that the specific filtration resistance of the gel layer is 700 times higher than the cake layer. However, it has also been reported that the gel layer pores are much larger than the cake layer pores [93].

Chen et al. [108] and Zhang et al. [109] reported the decisive role of osmotic pressure on filtration resistance in both AnMBRs and AeMBRs. These two studies laid the foundations for new theories regarding the role of the chemical potential to the gel layer filtration resistance. Hong et al. [91] reported that the negatively charged functional groups of the gel layer led to high concentrations of counter-ions surrounding the membrane surface. This results in an osmotic gradient between the permeate and the gel layer that can generate a filtration resistance much higher than the cake layer [91]. However, this theory cannot provide a reliable explanation for the high filtration resistance in zones where the gel layer is nearly electro-neutral. Chen et al. [93] suggested that Flory-Huggins theory could give a response to this phenomenon, since their experimental results showed that the filtration resistance depends on the gel thickness rather than on the pH and ionic strength. Accordingly, the chemical potential variation in the gel layer could be one of the main mechanisms affecting the filtration resistance of the gel layer [93,94,110].

Future research on membrane fouling

Future research efforts on membrane fouling in anaerobic systems should focus on broadening the understanding and applicability of the aforementioned theories. Most of the AnMBR publications have not differentiated foulant layers and only a few recent publications have identified the formation of a gel layer under anaerobic conditions. This is important since (i) membrane filtration resistance appears to be governed by gel layer formation rather than the cake layer formation and (ii) it can lead to contradictory and confusing information in the literature. Further research should evaluate the implications of gel layer formation in AnMBRs performance from which researchers can conceive and develop improved configurations and operational conditions for fouling control.

3. Novel configurations for membrane fouling control

Many pilot and lab-scale AnMBR configurations have been trialled to treat municipal sewage. These AnMBR technologies can be classified in two groups (i) completely stirred tank reactor (CSTR) (Figure 1a and 1b) and (ii) UASB reactors (Figure 1c) [10,39]. In both systems, gas sparging is the most used strategy for membrane scouring and fouling control (Table 1 and 2). However, gas sparging consumes a large amount of energy (0.21 $\pm$ 0.13 kWh m^{-3}) and, therefore, its optimisation is important to minimise energy consumption and related operating costs [28,30,57]. This section discusses the most promising new configurations proposed to replace or reduce gas sparging requirements. It is worth mentioning that anaerobic dynamic membrane bioreactors (AnDMBR) are out of the scope of the present review since AnDMBRs were recently reviewed by Hu et al. [33].

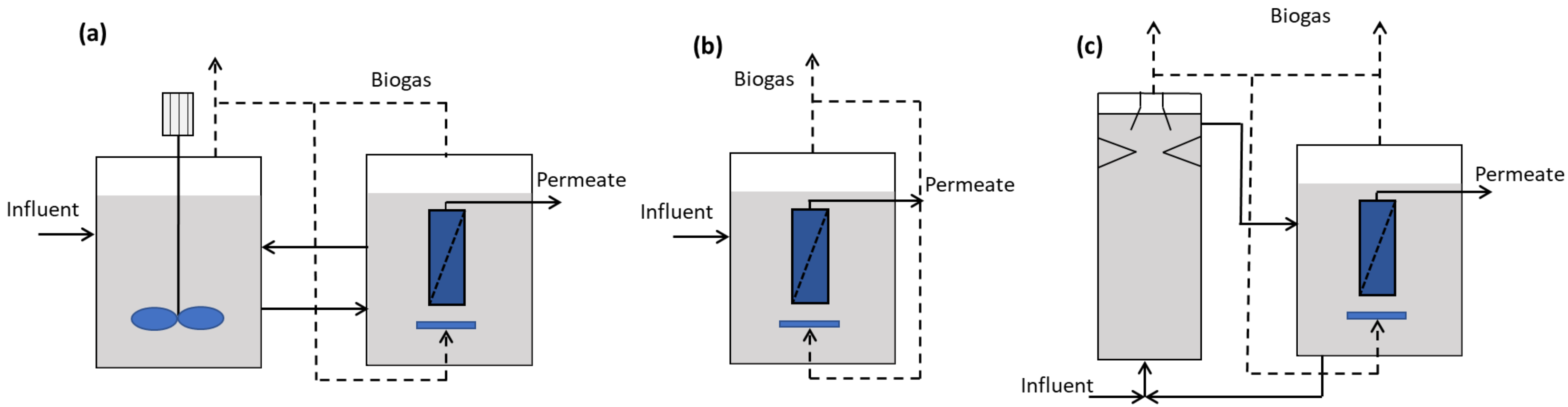

Figure 1. AnMBR configurations for municipal sewage treatment. (a) AnMBR-CSTR with separated membrane tank; (b) AnMBR-CSTR in a single tank; (c) AnMBR-UASB with separated membrane tank.

Table 1. Membrane characteristics and performance in AnMBR-CSTR configurations for municipal wastewater treatment.

Membrane configuration	Pore size (μm)	Material	Flux (L m^{-2} h^{-1})	Filtration area (m^2)	Fouling control	Chemical cleaning	Reference
Hollow fibre	0.05 UF	-	10[1]	30	Gas sparging (0.23 m^3 m^{-2} h^{-1})	No	[18]
Hollow fibre	0.05 UF	-	9-13.3[1]	30	Gas sparging (0.23-0.33 m^3 m^{-2} h^{-1})	No	[44]
Hollow fibre	0.04 UF	PVDF	17	5.4	Gas sparging + $FeCl_3$	Yes	[46]
Hollow fibre	0.05 UF	--	9-13.3	30	Gas sparging (0.23 m^3 m^{-2} h^{-1})	No	[48]
Flat sheet	0.2 MF	Polyethersulfone	5-7	0.0387	Gas sparging (7.24 m^3 m^{-2} h^{-1})	No	[52]
Hollow fibre	0.2 MF	-	6	5.4	Gas sparging	No	[56]
Flat sheet	0.45 MF	Polyethersulfone	5.3-7.9	0.118	Gas sparging	Yes	[65]
Hollow fibre	0.04	PVDF	11.7-12.3	0.9	Rotating membrane	Yes	[111]
Flat sheet	Dense	Cellulose triacetate	3.5-9.5	0.025	Gas sparging (4.8 m^3 m^{-2} h^{-1})	-	[112]
Flat sheet	Dense	Cellulose triacetate	3-10	0.025	Gas sparging (4.8 m^3 m^{-2} h^{-1})	-	[113]
Flat sheet	Dense	Cellulose triacetate	2-6	0.025	Gas sparging (2.4 m^3 m^{-2} h^{-1})	-	[114]
Flat sheet	0.2 MF	PVDF	<6	0.025	Gas sparging (2.4 m^3 m^{-2} h^{-1})	No	[114]
Flat sheet	0.038 UF	Polyether sulfone	7	3.5	Gas sparging	No	[115]
Hollow fibre	0.03 UF	PVDF	-	0.031	CFV (0.3 m/s)	No	[116]

[1]Flux normalized to 20 ºC (LMH)

Table 2. Membrane characteristics and performance in AnMBR-UASB configurations for municipal wastewater treatment.

Membrane configuration	Pore size (μm)	Material	Flux (L m^{-2} h^{-1})	Filtration area (m^2)	Fouling control	Chemical cleaning	Reference
-	0.08-0.3	Alumina	6	-	Gas sparging	No	[41]
Hollow fibre	0.04 UF	PVDF	9-15	0.93	Gas sparging (0.2-2 m^3 m^{-2} h^{-1})	Yes	[50]
Hollow fibre	0.03 UF	PVDF	4.1-7.5	39.5	Granular activated carbon	No	[64]
Hollow fibre	0.045 UF	PVDF	10-14	0.93	Gas sparging (0.4-1 m^3 m^{-2} h^{-1})	Yes	[117]
Hollow fibre	0.045 UF	PVDF	8-15	0.93	Gas sparging (0.4- 1 m^3 m^{-2} h^{-1})	No	[118]
Tubular	0.03 UF	PVDF	10-15	0.013	Gas lift mode (CFV of 0.3-1 m s^{-1})	Yes	[119]
Tubular	0.03 UF	PVDF	4.5	0.066	Gas lift mode (CFV of 0.12 m s^{-1})	Yes	[120]
Hollow fibre	0.1 UF	PVDF	10	0.091	Granular activated carbon	Yes	[121]
Hollow fibre	0.4 MF	-	11.3	0.19	Granular activated carbon	-	[122]

3.1 Novel configurations in AnMBR-CSTR systems

3.1.1 Rotating membranes

Rotative membrane modules have been considered as a possible alternative to gas sparging [123,124]. This configuration consists of coupling the membrane in a rotatory axis that generates shear stress (Figure 2a). Ruigómez et al. [125] compared, in lab-scale short-term assays (~1h), a conventional gas sparging system and a rotating system for membrane fouling control. The latter consisted of a hollow-fibre module rotating between 330 and 100 rpm to guarantee the generation of a scouring effect on the membrane surface. Ruigómez et al. [125] showed that the fouling rate (defined as dTMP/dt) decreased with the rotating velocity. However, when fouling rate reached values close to 0.01 kPa s^{-1} further improvements on fouling reduction could not be achieved by increasing the rotating velocity. Ruigómez et al. [125] attributed this phenomenon to the formation of a primary irreversible layer that could not be removed with physical methods. Regarding the gas-sparged AnMBR, Ruigómez et al. [125] reported that the membrane fouling rate decreased by increasing the specific gas demand (SGD) until the SGD reached 1.3 m^3 m^{-2} h^{-1}. Beyond this value, no fouling mitigation was achieved by increasing the SGD intensity. The authors attributed these results to the higher resistance that the membrane module offered to the gas passing through the fibres than passing through the membrane sides. Ruigómez et al. [125] concluded that membrane rotation can be more effective for fouling control than gas sparging since the power supplied by the rotating engine is more homogeneously distributed on the membrane. Ruigómez et al. [111] compared both configurations (i.e. conventional gas sparging system and a rotating system) at pilot-scale. The rotating membrane system exhibited better performance than the gas sparging confirming their lab-scale results. Specifically, the rotating system achieved critical fluxes around 20% higher than the gas sparging. In a subsequent study by the authors, a lab-scale AnMBR equipped with a rotating hollow-fibre module system allowed achieving a stable flux of 6.7 L m^{-2} h^{-1} (LMH) at 340 rpm with long term-assays (400h) [126].

Despite the improved fouling control of the membrane rotating system, the main concern of this technology is related to energy consumption. Shin and Bae [28] estimated that the energy consumption of the Ruigómez et al. [111] pilot-scale AnMBR was 0.30 kWh m^{-3}, which was higher than the reported in other pilot-scale AnMBRs using gas sparging for fouling control. Therefore, it is required to optimise the energy consumption of AnMBRs using rotating membranes as fouling control strategy by exploring different mixing modes and intensities. Finally, the impact of high rotating velocities on process performance and stability as well as on microbial community capacity is yet to be explored. Intensive mixing has been reported particularly counterproductive during shock loads or during start-up of the digestion process [127].

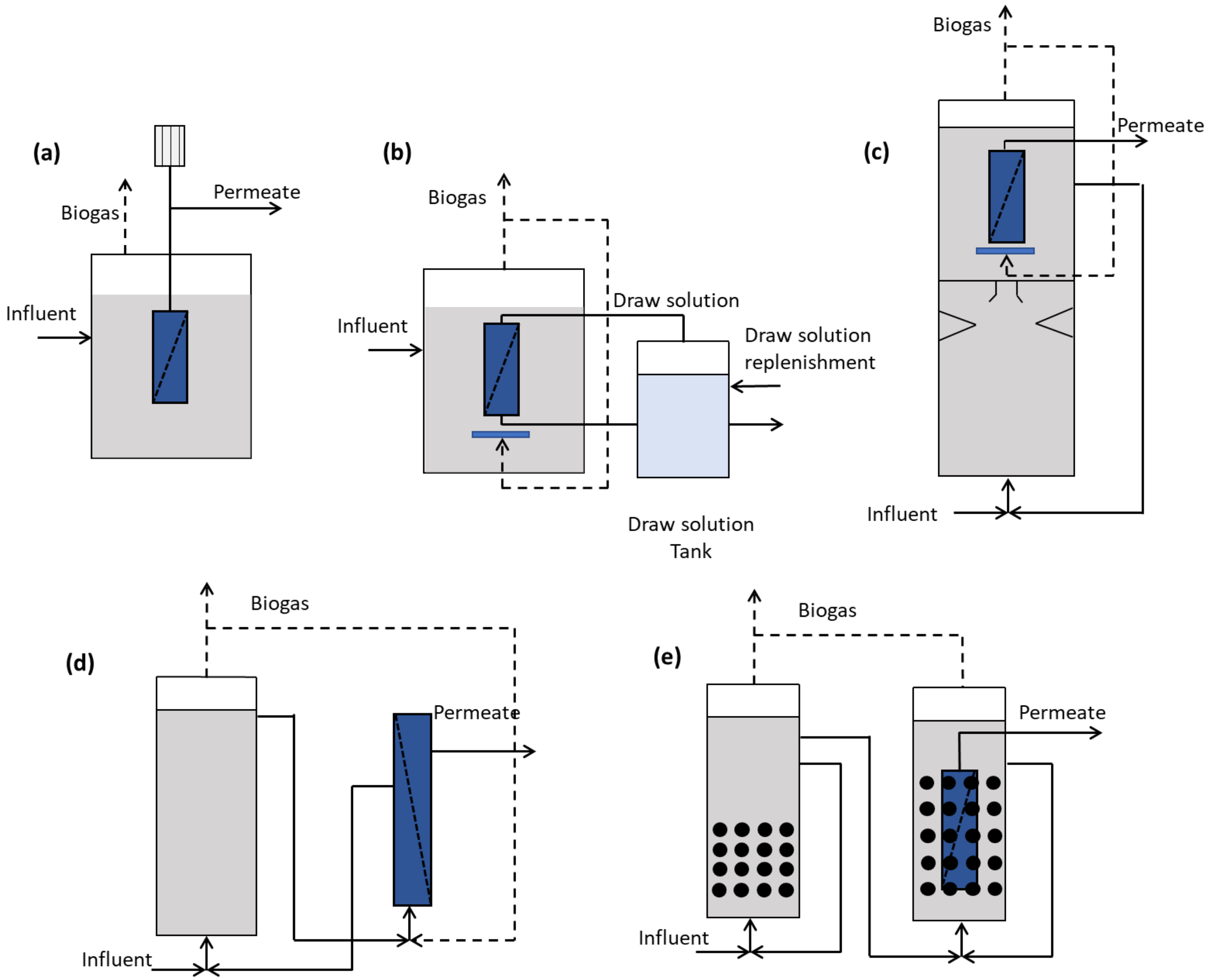

Figure 2. Novel AnMBR configurations for municipal sewage treatment. (a) Rotating membranes (adapted from Ruigómez et al. [125]) ; (b) Anaerobic osmotic membrane bioreactor (adapted from Chen et al. [112] and Gu et al. [113]); (c) membrane coupled at the top of the AnMBR-UASB (adapted from Gouveia et al. [118]) ; (d) Gas-lift AnMBR (adapted from Prieto et al. [119]); (e) two-stage anaerobic fluidized membrane bed bioreactor (adapted from Shin et al. [64] and Kim et al. [121]).

3.1.2 Anaerobic osmotic membrane bioreactors (AnOMBR)

The forward osmosis (FO) process is getting attention to reduce fouling in both AnMBRs and AeMBRs due to the lower fouling and the higher rejection of dissolved pollutants than UF and MF membranes [128]. FO is driven by an osmotic gradient generated by saline draw solutions that facilitates water permeation through a semipermeable dense membrane from the region of lower osmotic pressure (mixed liquor) to the region of higher osmotic pressure (draw solution) [129]. Accordingly, FO systems do not require hydraulic pressure to achieve water permeation. The installation of FO membranes in AnMBR systems is known as anaerobic membrane bioreactor coupled with FO (AnOMBR) (Figure 2b).

Chen et al. [112] treated synthetic municipal wastewater at 25 ºC using AnOMBR. The reactor exhibited more than 96% of organic carbon removal. However, the membrane flux decreased from 9.5 to 3.5 LMH within 22 days. This flux drop was attributed to the salinity build-up in the bioreactor which increased the conductivity from 1.0 to 20.5 mS cm^{-1}. Gu et al. [113] also reported that the flux gradually decreased due to the accumulation of ions in the AnOMBR reactor. The accumulation of ions in the mixed liquor reduces the driving force (i.e. osmotic gradient) and exacerbates membrane fouling [130,131]. The reasons leading to membrane fouling exacerbation under high salinity conditions remain under discussion. It has been hypothesised that this phenomenon could be explained using the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory. Briefly, the presence of counter-ions in the mixed liquor compresses the electric double layer around the floccules, reducing the electrostatic repulsion and increasing the attractive Van Der Waals forces [132–134]. According to this theory, the interaction between foulants and membrane increases with the ionic strength. However, other studies have stated that this theory partially fails to describe membrane behaviour under specific salinity conditions [97,135,136].

Miao et al. [97] tested the influence of the ionic strength on membrane permeation by adding different NaCl concentrations in a synthetic bovine serum albumin (BSA) solution. Miao et al. [97] reported that membrane fouling increased as the NaCl concentration increased from 0 to 0.06 g L^{-1}. However, membrane fouling was significantly reduced when the NaCl concentration ranged between 0.6 and 6 g L^{-1}. Miao et al. [97] linked membrane fouling behaviour to the hydration repulsion forces. At low ionic strength, hydration forces are weak and, therefore, the variation of the electrostatic force generated by the compression of the double layer increased foulants attachment on the membrane surface. On the other hand, at high ionic strength, the hydration repulsion forces originated by the highly hydrated sodium ions surrounding negative charges of the polyvinylidene fluoride (PVDF) membrane and BSA are more relevant. From Miao et al. [97] results, it can be concluded that membrane flux is not only affected by the accumulation of salts in the solution. Thus, other factors (e.g. biologic performance, operational conditions, membrane material) have to be also taken into account. However, the relative importance of these factors on membrane fouling is yet to be unfolded. In any case, salinity and ionic strength have a well-known impact on microbial community morphology, structure and capacity [137–139] which can affect floccules composition and particle size distribution [140–143]. Therefore, successful AnOMBR operation requires achieving a compromise solution between membrane performance and biological process.

Wang et al. [114] proposed a new AnMBR configuration combining MF membrane and FO membrane (AnMF-OMBR) to control the accumulation of salts in the mixed liquor. This configuration allowed to keep the conductivity of the mixed liquor between 2.5 and

4.0 mS cm^{-1}. However, biofouling and inorganic scaling, that could not be removed with physical methods, were observed on the FO membrane. In a subsequent study, Wang et al. [66] applied a chemical cleaning method to mitigate the long-term fouling on the FO membrane. The optimum chemical cleaning protocol consisted of applying a 0.5% H_2O_2 solution at 25ºC during 6 h. In a subsequent study, Wang et al. [144] proposed a new operational strategy consisting of a two-stage pattern: a first stage using a low driving force (i.e. low draw solution concentration) and a second stage using a high driving force (i.e. high draw solution concentration). Wang et al. [144] reported that this operational mode alleviated the flux drop and enhanced the filtration performance of the FO membrane.

Despite the recent advances, the implementation of AnOMBR is still challenging. The impact of salinity on the long-term feasibility of this technology requires research focused on both anaerobic digestion and membrane performance. The AnMF-OMBR system is a step forward to overcome salinity constraints. However, the necessity of using two membrane processes (i.e. FO and UF) in the bioreactor significantly hinders the technical and economic prospect of this approach (i.e. higher capital and operating costs, more complex physical and chemical cleaning procedures, and draw solution regeneration among others). Overall, the development of FO membranes able to achieve high water fluxes and low reverse solute fluxes is crucial to reduce capital and operating costs of AnOMBRs.

3.2 Novel configurations in AnMBR-UASB systems

3.2.1 Membrane coupled at the top of the AnMBR-UASB

Most common AnMBR-UASB configurations include the membrane module in an external tank [49,98,117,144,145] or at the top of the UASB reactor [77,146,147]. The former is the most reported system. However, if poorly managed, this configuration can lead to solids accumulation in the membrane tank and exacerbate membrane fouling. The accumulation of fine solids in the membrane tank occurs when the recirculation flow from the membrane tank to the UASB is not high enough or the system is not properly designed. This is of special concern considering the poor settling characteristics of the suspended biomass leaving the UASB and entering the membrane tank. According to Gouveia et al. [117], these particles tend to accumulate close to the membrane module due to their lower back-transport. The design of AnMBR systems able to provide high shear conditions on the membrane surface by combining gas sparging with other fouling mitigation alternatives is necessary to reduce the accumulation of fine solids and improve membrane fouling control in AnMBR-UASB systems. Consequently, optimising the recirculation flow between the UASB and the membrane module as well as providing enough turbulence in the membrane tank are important factors for fouling control in this configuration.

Gouveia et al. [118] proposed a new pilot-plant system in which the membrane module was submerged at the top of the reactor rather than submerged in an external membrane tank (Figure 2c). The novelty of this system was that the AnMBR-UASB consisted of two differentiated zones: (i) the biological zone, located at the bottom of the UASB, and (ii) the filtration zone, located at the top of the UASB. In the filtration zone, a high degree of mixing was achieved by (i) biogas sparging which was recirculated at the bottom of the membrane modules, (ii) the recirculation flow from the bottom of the membrane modules to the biological zone, and (iii) the use of two baffles placed between the filtration zone and the three-phase separator. Gouveia et al. [118] reported that the COD accumulation rate in the filtration zone was reduced from 239-702 mg COD L^{-1} d^{-1} to 90-119 mg COD L^{-1} d^{-1} when recirculation was turned on. These results suggested that proper mixing is needed to prevent solids accumulation and membrane fouling. Gouveia et al. [118] configuration was operated for three years with a permeate flux between 12 and 14 LMH, without requiring any physical or chemical cleaning.

Shin and Bae [28] estimated the energy consumption of the pilot-plant in Gouveia et al. [118] and reported that their configuration featured the lowest SGD (0.16 m^3 m^{-2} h^{-1}) and the lowest energy consumption (0.05 kW m^{-3}) among the eleven evaluated AnMBR pilot-plants. Peña et al. [24] recently used the same pilot-plant configuration to treat municipal sewage but operated without temperature control (10-28ºC). The temperature fluctuations led to a variable anaerobic digestion and membrane performance, which could be the factor behind the increased SGD requirements (0.66-0.74 m^3 m^{-2} h^{-1}) when compared to those of Gouveia et al. [118]. The impact of temperature and temperature fluctuations on SGD optimisation should be studied in more detail since it plays a notable role in the AnMBR operational costs.

3.2.2 Gas-lift AnMBR (Gl-AnMBR)

Submerged configurations are preferred over side-stream configurations for municipal sewage treatment due to their lower energy consumption [148,149]. The cross-flow velocity (CFV) required for fouling control is the main bottleneck of side-stream configurations [37]. Prieto et al. [119] proposed a hybrid gas-lift AnMBR (Gl-AnMBR), a side-stream system aiming to minimise the CFVs and the associated energy consumption. In Gl-AnMBR system, the CFV for fouling control is provided by both mixed liquor recirculation and biogas recirculation (Figure 2d). This strategy provides a two-phase (gas-liquid) flow through the membrane, where the rising bubbles improve the turbulence on the membrane surface. Consequently, lower CFVs are required which reduce energy consumption. Prieto et al. [119] configuration sustained stable membrane fluxes ranging from 10 to 15 LMH with a CFV of 0.5 m s^{-1}, and a gas to liquid ratio of 0.1.

Dolejs et al. [120] assessed the effects that temperature shocks would have on the performance of the Gl-AnMBR system (i.e. short-term shocks of 12 to 48 h from 35 to 15 ºC). The TMP slightly increased after the temperature shock owing to the higher water viscosity at lower temperatures. However, Dolejs et al. [120] concluded that both membrane flux and TMP remained stable during the 15 ºC shocks, which is important for the robustness of the Gl-AnMBR system.

The Gl-AnMBR system is of special interest since it addressees the optimisation of a side-stream configuration, which has been rarely considered for municipal sewage treatment. However, this configuration presents critical challenges for its full-scale implementation. Although Gl-AnMBR requires lower CFVs in comparison with classic side-stream systems, this configuration could have higher capital costs due to the installation of both gas and liquid recirculation systems. Additionally, the technical implications of combining gas and liquid turbulence for membrane fouling control still needs more research. Special attention should be given to the technical challenges associated with biogas recirculation and the associated implications in membrane operability (e.g. backwash, membrane chemical clean in place and out of place). Preferably, these studies should be carried out at pilot-scale since, to the best of our knowledge, the Gl-AnMBR configuration has not been tested at pilot-scale.

3.2.3 Anaerobic fluidised membrane bed bioreactor (AFMBR)

The use of granular activated carbon (GAC) as a fouling control method is gaining attention for AnMBRs [29,149]. In this configuration, GAC particles are fluidised and used for membrane scouring. Fluidisation is energy-intensive although its energy consumption can be lower than the required for gas sparging. Shin and Bae [28] reported GAC fluidisation as one of the most competitive approaches in terms of energy consumption for fouling control (0.102 kWh m^{-3}).

In its early stages, this process consisted of two separated reactors, an anaerobic fluidised bed reactor (AFBR) and an anaerobic fluidised membrane bed reactor (AFMBR) (Figure 2e). Both reactors were filled with GAC particles to (i) provide a carrier surface where the biomass was attached, (ii) scour the membrane, and (iii) adsorb soluble and colloidal matter surrounding membrane surface. Kim et al. [121] used the AFBR-AFMBR system for sewage treatment and biogas production. The short-term experiments showed that GAC fluidisation was an effective method for fouling reduction, while the long-term experiments showed that GAC addition was able to keep membrane fluxes at 10 LMH for 40 days with only a slight TMP increase (0.025 bar). Yoo et al. [150] operated an AFBR-AFMBR system for 192 days, and observed that the GAC scouring effect and relaxation periods were enough to prevent significant fouling and hence neither chemical nor physical cleaning were needed. The membrane reached fluxes up to 9 LMH during the first period of operation (160 days). However, when the membrane flux was increased up to 12 LMH an important TMP increase (0.2 bar) was observed.

As the technology evolved, some studies considered the possibility to use a single-stage system (i.e. AFMBR only) rather than a two-stage system (AFBR-AFMBR). Bae et al. [151] compared both systems under similar operating conditions and concluded that both systems exhibited similar COD removal efficiencies (93-96%) and TMPs (0.1 bar). Similarly, Wu et al. [152] reported COD removal efficiencies above 97% for both configurations. Therefore, according to Bae et al. [151] and Wu et al. [152] the first-stage AFBR could be avoided. However, the performance of the single-staged AFMBR needs to be tested at pilot-scale.

Alternative materials have been used for fouling control [149]. Hu & Stuckey [153] compared GAC and powdered activated carbon (PAC) and concluded that PAC could be a better material for fouling control than GAC. However, Yang et al. [154] reported that both materials were able to reduce cake layer formation, although GAC was slightly superior than PAC. The addition of polyethylene terephthalate (PET) or PVDF as scouring materials has also been tested [155–158]. Aslam et al. [156] reported that SGDs were reduced by 67% when gas sparging was combined with PET particles.

The durability of the membranes in constant contact with the scouring particles is important for the application of AFMBR technology. Shin et al. [159] operated an AFBR-AFMBR pilot-scale for two years and reported that the membrane was severely damaged due to the continuous contact with fluidised GAC. Shin et al. [159] observed that the middle and bottom of the membrane was significantly damaged due to the contact with more densely-packed particles. Larger particles are more damaging than smaller ones for the membrane but are better for fouling control. Accordingly, the selection of a suitable particle size is critical for membrane performance and integrity [160,161].

Ceramic membranes have recently gained attention due to their higher resistance to abrasion [162,163]. Additionally, these membranes characteristically achieve excellent membrane flux performance. Aslam et al. [163] used a single-stage AFMBR equipped with a ceramic membrane composed of aluminium oxide (Al_2O_3) and achieved high COD removals (~90%) and net fluxes of 17 LMH in long-term operation (395 days). In a subsequent study, Aslam et al. [164] reached higher membrane fluxes (~22 LMH) when using a ceramic membrane in an AFBR-AFMBR system. However, further studies are required to better understand the membrane performance differences between the AFMBR and the AFBR-AFMBR systems.

The long-term effects of the scouring particles on membrane integrity is a primary barrier for the implementation of AFMBR. The use of alternative membrane materials (e.g. ceramic membranes) is a research direction which should be further explored to overcome this limitation. Evans et al. [29] recently compared two pilot-scales using gas sparging and AFBR-AFMBR for fouling control. The AFBR-AFMBR, which used GAC as scouring material, allowed to work at shorter HRTs than the gas-sparged AnMBR. This improvement was attributed to the higher resilience of biomass attached to the GAC particles. However, the gas sparging provided a more flexible operation due to the possibility to (i) adjust the gas sparging rate, (ii) avoid the damaging effect of GAC and (iii) keep the membrane permeability constant with higher concentrations of suspended solids and colloids in the mixed liquor. Evans et al. [29] concluded that a hybrid system combining a GAC-fluidised bioreactor and gas-sparged membranes would benefit from the capacities of both fouling control methods while improving the technical feasibility of GAC-fluidised AnMBR. However, the combination of these two energy-intensive alternatives (i.e. gas sparging and GAC fluidisation) could compromise the economic feasibility of the AnMBR system, despite their combination could improve membrane fouling control and biological performance.

4. Temperature

4.1 Temperature influence on anaerobic digestion performance

The diluted origin of municipal sewage makes unfeasible to heat the digester content and, therefore, AnMBRs are typically operated at ambient uncontrolled temperature conditions [165]. Psychrophilic conditions (< 20 ºC) have been used for AnMBR at lab-scale [52,53,62,65,114] and at pilot-scale [24,48,64,111,117,165] (Table 3 and 4).

Table 3. Biological performance in AnMBR-CSTR configurations for municipal wastewater treatment.

Reactor configuration	Scale	Type WW	T (ºC)	OLR (kg COD m^{-3} day^{-1})	COD removal (%)	MLSS (g TSS L^{-1})	Methane yield (m^3 CH_4 kg^{-1} COD_{IN})	HRT (h)	SRT (days)	Reference
Submerged	Pilot	Real	33	-	87	6-22	0.069	6-20	70	[18]
Submerged	Pilot	Real	17-33	0.3-1.1	85	10-30	-	6-26	30-70	[44]
Submerged	Pilot	Synthetic	23	-	-	11.3-21.3	-	8.5	40-100	[46]
Submerged	Pilot	Real	15-33	-	-	10-30	-	5-24	40-100	[48]
Submerged	Lab	Synthetic and real	15	0.44-0.66	92 (Synthetic) 69 (Real)	6-10.6[2]	-	16-24	300	[52]
Submerged	Pilot	Real	35	3	87	4.7-20.1	0.12	2.2	60	[56]
Submerged	Lab	Synthetic	25-30	1.10-1.65	95-99	5.5-10.4	0.124-0.25	8-12	30-∞	[65]
Submerged	Pilot	Real	19	1.1	91	21.3	0.012	33	270	[111]
Submerged	Lab	Synthetic	25	-	97	3.9-4.6[2]	0.21	15-40	90	[112]
Submerged	Lab	Synthetic	35	-	>95	-	0.25-0.3	15-40	90	[113]
Submerged	Lab	Synthetic	25	-	90-96[3]	-	0.25-0.28	35-60	80	[114]
Submerged	Pilot	Real	20-35	0.5-1.1	82-90	15-21	0.27-0.23	19.2	-	[115]
Side-stream	Lab	Synthetic	35	0.8-10	97-99	16	0.088-0.393	6-12	1000	[116]

[1] VS concentration (g L^{-1}); [2] VSS concentration (g L^{-1}); [3] total organic carbon removal (%).

Table 4. Biological performance in AnMBR-UASB configurations for municipal wastewater treatment.

Reactor configuration	Scale	Type WW	T (°C)	OLR (kg COD m^{-3} day^{-1})	COD removal (%)	MLSS (g TSS L^{-1})	Methane yield (m^3 CH^4 kg^{-1} COD_{IN})	HRT (h)	SRT (days)	Upflow velocity (m h^{-1})	References
Submerged	Lab	Real	25-30	-	86-89	12.8-12.9	0.1 ± 0.02	7.5	60	-	[41]
Submerged	Pilot	Real	16.3	-	83.0	0.384[1]	-	8	-	0.8-0.9	[50]
Submerged	Pilot	Real	9-30	-	81-94	-	-	4.6-6.8	6.2-36	27-75	[64]
Submerged	Pilot	Real	18	0.81-4.70	87	-	0.16-0.23	17-7	-	0.15-0.45	[117]
Submerged	Pilot	Real	18	0.6-3.18	75-90	-	0.26-0.14	9.8-20.3	-	0.12-0.34	[118]
Side-stream	Lab	Synthetic	37	0.42	93	22	-	72	60	-	[119]
Side-stream	Lab	Synthetic	15-35	0.62-0.88	94	-	0.19-0.07	30-36	-	-	[120]
Submerged	Lab	Synthetic	35	4.4-6.2	99	-	-	2.0-2.8	-	-	[121]
Submerged	Lab	Synthetic	15-35	1.21-1.44	51-74	-	0.14-0.19	6	-	-	[122]

[1] Concentration in the membrane tank.

Temperature fluctuations and temperatures below 10 ºC are two important challenges for AnMBR technology. Ferrari et al. [62] evaluated the influence of seasonal temperature variations and monitored COD removal efficiencies above 87% for temperatures between 23 and 34 ºC. However, when the temperature decreased to 15 and 17 ºC, the COD removal efficiency decreased to around 70%. Similarly, Peña et al. [24] operated an AnMBR without temperature control (10-28 ºC) and reported higher COD concentrations in the effluent when the temperature was 10 ºC. These results are in agreement with other publications studying AnMBR performance at psychrophilic conditions [106,120,122,165,166].

Temperature has an impact on the digesters microbial community and degradation rates [167]. Hydrolysis is typically considered the rate-limiting step in the anaerobic digestion of highly particulate waste and wastewater [168,169]. One advantage of AnMBR is that the membrane provides excellent retention of solids in the bioreactor giving more time for particles to be hydrolysed. Therefore, if the SRT is high enough, the decrease of the hydrolysis rate at lower temperatures may not be controlling the amount of methane recovered in AnMBR. Temperature changes may also affect the degradation rate of the other anaerobic digestion steps (i.e. acidogenesis, acetogenesis and methanogenesis) as well as the syntrophic relationships between microorganisms [170,171]. The slightly lower equilibrium constant and the higher H_2 solubility makes volatile fatty acids (VFA) degradation less favourable at psychrophilic temperatures [172]. If improperly managed, this can increase the VFA concentration and decrease the pH of the mixed liquor which, in turn, can partially, or totally, inhibit methanogenic activity. Besides the great adaptability of microorganisms to different environmental conditions, the lower degradation rate at lower temperatures can be compensated by increasing the amount of active biomass in the digester (higher SRT).

Acetoclastic methanogenesis and hydrogenotrophic methanogenesis are the two main pathways for methane generation [173]. Smith et al. [22] reported that hydrogenotrophic methanogenesis could be the predominant pathway in AnMBRs operated under psychrophilic conditions, which was attributed to the higher solubility of hydrogen at lower temperatures. However, in a subsequent publication, Smith et al. [52] reported that acetoclastic methanogens were more abundant than hydrogenotrophic methanogens in an AnMBR treating municipal sewage at 15 ºC. Acetoclastic methanogens were also reported as the dominant methanogens in other psychrophilic AnMBR studies [53,62,106]. Ozgun et al. [53] stated that the higher hydrogen solubility under psychrophilic conditions could have promoted acetate production through the homoacetogenic pathway. However, more studies are required to understand the impact of temperature on microbial community structure, degradation rates and degradation pathways. It is worth mentioning that the microbial community, and methanogens in particular, can be affected by several factors such as pH, loading rate and presence of inhibitors (e.g. Na^+, H_2S, NH_3, heavy metals, organics) among others.

COD removal efficiencies around 90% have been achieved in AnMBRs working at psychrophilic temperatures [52,53,57,115,174]. These results show the great adaptability of the microbial community to perform at low temperatures. However, these results are the combination of the microbial community capacity with other factors such as membrane configuration [52,53] and operational conditions (e.g. HRT, SRT and OLR) [21,22,35]. Ozgun et al. [53] and Lim et al. [57] attributed the high COD removals at psychrophilic conditions to the membrane separation process. The membrane retains particulate and colloidal COD in the digester providing a high-quality effluent. Smith et al. [52] reported that the biofilm on the membrane surface has a role in the removal of soluble organic matter under psychrophilic conditions. Indeed, several studies have reported significant differences between the bioreactor and the permeate soluble COD [52,115,174]. Smith et al. [174] observed that, under psychrophilic conditions, *Methanosaeta* (acetoclastic methanogenic) was the most abundant methanogen in the mixed liquor while *Methanospirillum* and *Methanoregula* (hydrogenotrophic methanogens) were the most abundant in the membrane biofilm. The principal coordinates analysis in Smith et al. [174] showed a distinct microbial community structure (including both archaea and bacteria) between the suspended biomass and the biofilm. Understanding the role, structure and development of the biofilm attached to the membrane surface are paramount for AnMBR technology.

4.2 Temperature influence on membrane performance

Temperature affects fluid and sludge properties [170]. The membrane permeability decreases as the temperature decreases due to the higher viscosity of water. Foulants properties also change with temperature. Watanabe et al. [106] and Martin-Garcia et al. [165] reported that membrane fouling was exacerbated at lower temperatures due to changes in SMP characteristics. Both studies associated the fouling rate increase to the higher protein to carbohydrate ratio at lower temperatures. Robles et al. [175] also reported an increase of membrane fouling when the temperature of an AnMBR pilot-plant was changed from mesophilic to psychrophilic conditions. However, these authors observed a lower SMPs protein to carbohydrate ratio at psychrophilic conditions. Nonetheless, as discussed in Section 2, the SMP protein and carbohydrate content and ratio do not seem to be a reliable indicator to predict fouling behaviour. Instead, particle size distribution appears to be more suitable for fouling evaluation and comparison. In this regard, Robles et al. [175] observed a smaller floc size at lower temperatures attributed to the lower biomass activity under psychrophilic conditions. Ozgun et al. [53] also observed that the average particle size decreased and the SMPs production increased when the temperature was decreased from 25 to 15 ºC. In both studies, the total filtration resistance significantly increased at lower temperatures. Peña et al. [24] evaluated membrane performance under annual temperature variations in a pilot-scale study. The filtration flux remained between 10 and 11 LMH at temperatures around 24 ºC. However, a gradual decrease in the flux (2-3.5 LMH) was reported at lower temperatures (~15 ºC). Future research should aim to improve membrane flux performance at low temperatures.

4.3 Dissolved methane

Methane solubility increases as the temperature decreases [176]. The methane solubility at 20 ºC is around 30% higher than at 35 ºC, hence, the methane concentration leaving the permeate is higher at psychrophilic than at mesophilic conditions [115]. The methane dissolved in the permeate has a double negative connotation: (i) it decreases the methane yield of the AnMBR and, therefore, the profitability of the technology and (ii) it is an important source of greenhouse gas emissions [177]. Smith et al. [52] found that 40-50% of the methane generated in a psychrophilic AnMBR remained dissolved in the permeate at 15 ºC. The authors hypothesised that the biofilm activity on membrane surface increased the concentration of methane in the permeate above oversaturation levels. Similarly, Lim et al. [57] found that 47% of the methane remained dissolved in the effluent between 15 and 20 ºC. This is critical since fugitive methane emissions significantly compromise the environmental feasibility of AnMBRs. Accordingly, developing technologies and operational strategies to minimise or recover the methane dissolved in the effluent is crucial for the success of AnMBR technology [23,31,178,179].

Technologies dealing with the methane dissolved in the AnMBR effluents include degassing membranes, aeration, and air stripping [180–182]. Degassing membranes appear as the most suitable technology due to (i) the capacity to recover the methane instead of oxidising it to CO_2 and (ii) the relatively high recovery yields achieved

[9,177,181,183–186]. Seco et al. [187] recovered 67% of the dissolved methane in the effluent of a pilot-plant AnMBR by using a hollow-fibre degassing membrane. In another pilot-scale study, Lim et al. [57] reported methane recovery efficiencies of 70 ± 5%.

Several studies analysed the economic impact of recovering the methane dissolved in the effluent of a psychrophilic AnMBR using degassing membranes. Crone et al. [181] estimated that the AnMBR technology could be operated without energy input if the dissolved methane was efficiently recovered. Pretel et al. [188] calculated that integrating a degassing membrane would allow operating the AnMBR with a very low energy input (0.04 kWh m^{-3}) and life-cycle cost (0.135 € m^{-3}). Evans et al. [29] reported that the energy requirements of a degassing membrane system were nearly negligible (0.01 kWh m^{-3}) when compared with the environmental and energy benefits. Similarly, Lim et al. [57] reported that a membrane contactor was able to recover up to 0.052 kWh m^{-3} from the methane dissolved in the effluent with an energy consumption of 0.008 kWh m^{-3}. Sanchis-Perucho et al. [186] estimated that the payback period for degassing membranes was around 10.5 years. Accordingly, the recovery of the methane is not only necessary, but also encouraging. Nonetheless, Lim et al. [57] noted that the methane remaining in the effluent was equivalent to 0.11 kg CO_2 m^{-3} and stated that further research is needed to reach higher methane recovery efficiencies. Another alternative is to combine gas contactors with other technologies to minimise methane emissions. The utilisation of denitrifying anaerobic methane oxidation (DAMO) process is an attractive biological process for the simultaneous removal of methane and nitrogen from AnMBR effluent [189,190]. However, this technology is still under development.

5. Sulphate

The presence of sulphate in municipal sewage significantly affects the anaerobic digestion and the filtration processes [191,192]. Sulphate reducing bacteria (SRB) use organic compounds and hydrogen as electron donors to convert sulphate into sulphide. In the presence of sulphate, SRB compete with methanogens for the same substrates decreasing the substrate availability for methanogenesis. Moreover, the production of sulphide from SRB can inhibit methanogenic activity, which could further decrease methane conversion [193,194]. The presence of hydrogen sulphide in biogas also requires the utilisation of corrosive resistant instrumentation and equipment [195], whereas the dissolved hydrogen sulphide lowers the durability of the membrane [9]. Therefore, the concentration of sulphate in sewage has a direct impact on the economic feasibility of AnMBR [47].

Shin and Bae [28] reported that AnMBR pilot-plants treating sewage with high sulphate concentrations (>99 mg SO_4^{2-}-S L^{-1}) obtained poor methane yields (0.08-0.15 L CH_4 g COD^{-1}) when compared to the average methane yield of those treating sewage with low sulphate concentrations (0.22 L CH_4 g COD^{-1}). Giménez et al. [18], who studied the influence of the COD/SO_4^{2-}-S ratio on anaerobic digestion performance, reported a sharp decrease of the methane production as the influent sulphate concentration increased. The methane production nearly ceased when COD/SO_4^{2-}-S was below the stoichiometric ratio for sulphate reduction of 2.01 mg COD mg^{-1} SO_4^{2-}-S (0.67 mg COD mg^{-1} SO_4^{2-}) [18]. The latter results showed that SRB outcompete methanogens and nearly all the sulphate is converted to sulphide if enough biodegradable COD is available. Furthermore, the presence of dissolved sulphide in the permeate can affect the overall treatment efficiency since sulphide contributes to the effluent COD [196]. The removal of sulphide from AnMBR effluents has been recently addressed by using coagulation-flocculation [57] and membrane distillation [197]. However, alternative methods such as electrochemical systems are also gaining attention due to their capacity to recover sulphide as sulphur or other oxidised sulphur species [198].

The corrosive nature of sulphide also affects membrane permeability and durability. Sulphide has been reported to damage the internal material when transported through the membrane cell, making it more susceptible to membrane fouling [9,199]. In this regard, Song et al. [191] observed that membrane fouling increased as the influent sulphate concentration increased. Specifically, the TMP sharply increased from 0.5 to 0.85 bar after the addition of more than 33 mg SO_4^{2-}-S L^{-1}. Song et al. [191] attributed these results to the larger release of EPSs under high sulphide concentrations. However, there is a little understanding of the impact of sulphide concentration on the microbial community activity, particle size distribution, EPS composition and membrane performance and durability.

Some publications have evaluated the impact of sulphate concentration in the economic and energetic prospects of the AnMBR process [28,200,201]. Ferrer et al. [200] estimated that the treatment of low-sulphate (57 mg COD mg^{-1} SO_4^{2-}-S) municipal sewage is more favourable than the treatment of sulphate-rich (5.7 mg COD mg^{-1} SO_4^{2-}-S) municipal sewage (0.070 and 0.097 € m^{-3}, respectively). Ferrer et al. [200] also stated that methane recovery from AnMBR effluents is more economically attractive when treating low-sulphate municipal sewage due to the low methane production at high sulphate concentrations.

These results clearly illustrate that sulphate concentration in sewage has a significant impact on AnMBR performance and profitability and, therefore, it has a key role in the decision-making process. Some studies have reported that COD/SO_4^{2-}-S ratios at or above 30 (10 mg COD mg^{-1} SO_4^{2-}) could be adequate to sustain a good anaerobic digestion performance and high methane yields [9,191]. However, further studies including technical, economic and energetic challenges connected to sulphate concentration are necessary. Particularly useful would be to determine a COD/SO_4^{2-}-S ratio threshold above which the AnMBR is recommendable for the treatment of municipal sewage. However, further understanding of the implication of sulphide concentration on anaerobic digestion and membrane performance is needed before carrying out such techno-economic study.

6. Forward osmosis pre-concentration (FO+AnMBR)

The application of anaerobic digestion to low-strength municipal sewage presents some challenges, including large AnMBR facilities (e.g. membrane area, digester volume and footprint), higher amounts of dissolved methane lost with the effluent, and low methane productivities per m^3 of wastewater treated. Municipal sewage pre-concentration by FO technology represents an opportunity to tackle these challenges since it allows to pre-concentrate municipal sewage with low energy inputs [60,62,202]. The FO process is spontaneously driven by an osmotic gradient between municipal sewage and a saline draw solution, which allows producing permeate without requiring hydraulic pressure [203,204].

Configurations to integrate FO and AnMBR technologies

The configuration used to integrate FO sewage pre-concentration and AnMBR technologies is highly dependent on the draw solution availability. In coastal areas, seawater availability makes open-loop schemes particularly advantageous if the seawater can be discharged to the environment after its utilisation. However, some pollutants and nutrients can diffuse from sewage to seawater through dense FO membranes. Particularly worrying is the diffusion of ammonium nitrogen, since low ammonium rejections (< 80%) have been reported for FO membranes [205–207]. Accordingly, FO membranes future development should aim to reduce the diffusion of these compounds to prevent potential environmental impacts in coastal areas after seawater discharge. In contrast, closed-loop schemes require the re-concentration of the draw solution while producing reclaimed water. In both schemes, pre-treatment of raw wastewater is needed to prevent potential fouling in FO membranes [208].

Two main draw solution management alternatives have been conceived for open-loop schemes: (i) the draw solution is discharged after the FO step (Figure 3a) and (ii) the draw solution is discharged after reverse osmosis (RO) stage for reclaimed water production (Figure 3b). The latter could be more attractive since it allows to combine sewage treatment and high-quality reclaimed water production (dual barrier) in the same facility. However, this alternative incurs extra operating costs due to the high energy required to operate the RO system. The energy requirements to produce reclaimed water from diluted seawater has been estimated to be in the range of 1.6-2.0 kWh per m^3 of produced water [60]. Therefore, the implementation of RO systems for reclaimed water production should be particularly considered in coastal areas with water scarcity. Detailed information regarding the possibilities of implementing a hybrid FO-RO system is already available in several publications [203,204,209–211].

Closed-loop schemes are required when natural draw solutions are not available (Figure 3c). In closed-loop schemes, the synthetic draw solution is regenerated after the FO stage (e.g. by RO) to re-establish the draw solution osmotic pressure. Although NaCl is the most used solute for synthetic draw solutions [212,213], this solute can present high reverse solute fluxes (i.e. solute flux from the draw solution to the sewage). Consequently, alternative solutes (e.g. organics, Mg^{2+}, Ca^{2+}) are being evaluated as potential draw solutes. Diminishing the reverse solute flux is crucial to reduce draw solution replenishment cost [60], prevent the inhibition of the AnMBR microbial community [61,214,215], and facilitate the reuse of the digestate as fertiliser or soil conditioner [216,217].

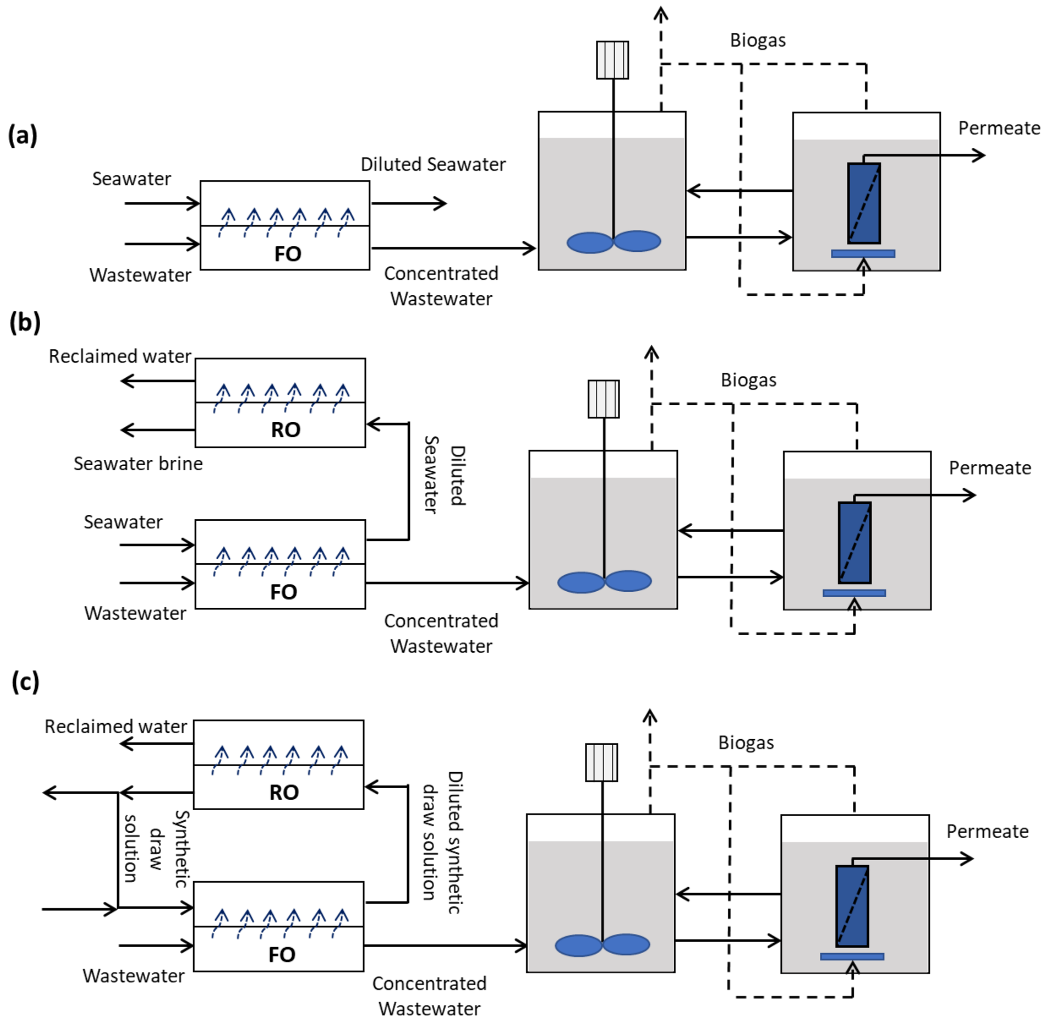

Figure 3. Configurations to integrate FO and AnMBR technologies for municipal sewage treatment (a) Open-loop FO+AnMBR; (b) Open-loop FO-RO+AnMBR; (c) Closed-loop FO-RO+AnMBR (adapted and expanded from Vinardell et al. [60]).

FO+AnMBR sewage pre-concentration and energy production

Pilot and lab-scale studies have pre-concentrated municipal sewage prior to AnMBR with FO water recoveries ranging between 50 and 90% leading to concentration factors of 2 and 10, respectively [61,63,206,218]. Ansari et al. [218] evaluated FO membrane and anaerobic digestion performance with ten different solutes as well as their impact on anaerobic digestion performance. Ansari et al. [218] reported that NaCl provides higher water fluxes (4.1 LMH) than other inorganic and organic solutes (e.g. NaAc and $MgSO_4$ had water fluxes <3.5 LMH). However, reverse solute fluxes were higher for NaCl (~3 g m^{-2} h^{-1}) than for other solutes such as NaAc (<1 g m^{-2} h^{-1}).

Solute selection must consider both water fluxes and reverse solute fluxes since high reverse solute fluxes highly increase the salinity of the AnMBR influent. Ansari et al. [218] reported that Na^+ inhibition is not significant for Na^+ concentrations below 3 g L^{-1}, which is in agreement with previously reported values [194,219,220]. In a subsequent study, Ansari et al. [61] also reported that NaAc (organic solute) led to higher methane yields than NaCl (inorganic solute). However, the study did not elucidate if these results are a consequence of (i) microbial inhibition as a result of the higher inhibition when using NaCl as solute, or (ii) the higher organic matter in the influent caused by the reverse solute flux of acetate when using NaAc as solute. Further studies are required to holistically evaluate the suitability of the different solutes, including FO performance, AnMBR performance, digestate management, and economic feasibility among others.

FO pre-concentration is an opportunity to make AnMBR energy self-sufficient. Wei et al. [116] conducted an energy balance for the FO+AnMBR system and showed that the biogas produced by the AnMBR could be sufficient to heat the influent to mesophilic conditions (~35 ºC). However, this alternative fails to transform the energy contained in the biogas to electricity and, therefore, it makes the process energetically negative instead of energetically neutral or positive. Accordingly, other alternatives (e.g. co-generation and biogas upgrading) appear more suitable attaining the emergence of the circular biobased economy.

7. Conclusions

Anaerobic membrane bioreactor (AnMBR) is a promising technology for mainstream municipal sewage treatment due to its capacity to produce high-quality effluents and renewable methane energy. However, there are still some technical challenges that need to be addressed to make AnMBR technically and economically competitive. Membrane fouling is a primary barrier for the applicability of AnMBRs. In AnMBRs, fouling has been generally attributed to pore clogging and cake layer formation. However, recent research has shown that the gel layer could be the main responsible for membrane fouling. Further research is needed to understand the relative importance of factors controlling the formation of the gel layer, from which new and improved mitigation strategies could be developed. Novel AnMBR configurations and operational conditions have also been researched to improve fouling control in CSTR and UASB reactors, bringing new opportunities for fouling control beyond gas sparging. Temperature affects metabolic kinetics, microbial community, membrane performance, particle size distribution and, most importantly, the amount of methane dissolved in the effluent. In this regard, FO pre-concentration could improve the process applicability by decreasing the AnMBR volumetric flow rate and reducing methane losses through the effluent. However, FO technology is still under development. Overall, the success of the AnMBR technology relies on improving membrane performance without hindering the biological process nor the economic feasibility of the process.

Declaration of competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. The authors also declare that this manuscript reflects only the authors' view and that the Executive Agency for SME/EU Commission are not responsible for any use that may be made of the information it contains.

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