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Review

Anaerobic membrane bioreactors for wastewater treatment: A review

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HIGHLIGHTS

- ▶ A critical review of the current situation of the AnMBR technology was made.
- ▶ Industrial scale AnMBRs are not reported but there are few cases at pilot scale.
- ▶ Excellent efficiencies in terms of COD and TSS removal are reported.
- ▶ The studies revised show that good fuel quality biogas can be produced.
- ▶ Membrane fouling is the key problem to solve before industrial implementation.

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ABSTRACT

This review provides an overview of the present situation, from 2006 to date, of the anaerobic membrane bioreactor technology with special emphasis on performance and bottlenecks in terms of its application at industrial scale. Most of the studies considered in this review were performed at bench scale; there is no description of real industrial applications in the literature and almost no pilot cases have been described. Anaerobic membrane bioreactors were fitted with flat sheet, hollow fibre or tubular membranes operating either in the microfiltration or in the ultrafiltration region, but the use of ceramic membranes has not been widely reported. Even though, under normal conditions, there should not be any difference in transmembrane pressure between hollow fibre and flat sheet membranes, hollow fibre membranes may lead to higher transmembrane pressures due to insufficient hydraulic shear on each of their fibres. Bioreactors were mainly tested under mesophilic or thermophilic conditions. The application of thermophilic conditions allowed treating higher organic loading rates. Chemical oxygen demand removal efficiencies up to 99%, total suspended solids removal efficiencies up to 100%, and complete removal of pathogens were reported. Therefore, treated waters may be directly discharged into water bodies or re-used for unrestricted crop irrigation if they meet the effluent discharge or irrigation standard of the area. The renewable energy produced within the plants (i.e. from methane production) was reported to cover the energy required for membrane filtration and the excess energy could be further used. Anaerobic membrane bioreactors are an attractive technology that needs further research efforts and industrialisation. However, membrane fouling, which still remains a major problem for all membrane bioreactors, seems much more severe under anaerobic conditions than aerobic ones.

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1. Introduction

Water scarcity is an increasingly severe global problem that may be mitigated by re-using wastewaters after suitable treatment methods [1,2]. Among the processes used for wastewater treatment (WWT), anaerobic processes have the advantage of reducing the organic matter of municipal and industrial wastewaters producing energy at the same time; their application to municipal wastewaters, however, appears to be more limited because methane (CH_4) production cannot cover heating requirements [3] being, for this reason, easily applied to countries with warmer climates [4–6].

The anaerobic degradation of complex organic matter to methane (CH_4) and carbon dioxide (CO_2), which involves the interaction of four different metabolic groups of bacteria, namely hydrolytic, acidogenic, acetogenic and methanogenic bacteria, [7] offers, in general, some significant advantages when compared to aerobic treatment. These are: less production of sludge, low nutritional requirements, ability to deal with high organic loads, low cost and finally biogas (CH_4) production [8–10]. However, due to their higher investment costs and their somehow complex operation, anaerobic processes are not always implemented. Furthermore, they are significantly influenced by a number of factors like the type and variability of wastewater, the type of organic contaminants in the influent, its pH, etc. [11].

Aerobic membrane bioreactor (MBR) technology was widely introduced for industrial application in the early 1990s. It is characterised by numerous advantages compared to conventional activated sludge (AS) processes: fast start-ups of the reactors [12,13], small footprint, high efficiency (high chemical oxygen demand (COD) and total suspended solids (TSS) removal; so, production of treated water of excellent quality), high organic loading rates without any biomass losses, control over solids retention times (SRTs) and hydraulic retention times (HRTs), maintenance of high mixed-liquor suspended solids (MLSS) concentrations, etc. [14–19]. Furthermore, compared to biofilters, which are the oldest and simplest biofiltration technology, membrane bioreactors (MBRs) are more efficient and they can deal with biomass accumulation and work at higher inlet concentrations [20]. The advantages of MBRs can be improved by working at anaerobic conditions [5,21]. Therefore, this technology is receiving remarkable interest by both researchers and industrialists [9]. The main disadvantage of both types of MBRs, aerobic and anaerobic, is membrane fouling, which leads to reduction in membrane permeate fluxes and, hence, increasing costs and preventing MBRs from an even faster commercialisation [14]. Also, even though membrane cost has significantly decreased during the last years, it still represents an important cost regarding full-scale application of anaerobic membrane bioreactors (AnMBRs) [22].

The main objective of this review is to provide an overview of the actual situation, mainly from 2006 onwards, of the AnMBR technology focussing on the performance and main bottlenecks of its application at industrial scale.

2. AnMBR performance

The first commercial AnMBR was constructed long ago, in the early 1980s, by Dorr-Oliver for treating high-strength whey processing wastewater, a development known as membrane anaerobic reactor system [5]. Since then, AnMBRs have been studied for treating municipal and industrial wastewaters of all different contaminant loads, i.e. low- [14,23], medium- [24] and high-strength [25] wastewaters (Tables 1–3).

2.1. Comparison of AnMBRs with other WWT technologies

Anaerobic micro-organisms are known to grow and reproduce more slowly than the aerobic ones [21]. Due to their low growth rate, biomass retention is critical for high-rate anaerobic treatment of wastewaters. Granule- and biofilm-based technologies represent the traditional way of achieving the necessary biomass retention to enable the operation of bioreactors at high biomass concentrations, hence, at high organic loading rates [22]. However, under specific conditions, such as high salinities or thermophilic temperatures, biofilm and granule formations do not proceed well and are negatively affected. AnMBRs, can be used to achieve the required sludge retention [22] in non-conventional conditions. They can successfully operate at longer SRTs [26], implying not only the potential to retain all micro-organisms, but also the capacity to provide them with the chance of becoming fully grown, improving the anaerobic treatment significantly [27].

Cornelissen et al. predicted in 2001 that AnMBRs would be a very promising technology with an important future [28]. At present, however, they still appear to be under development, compared to aerobic membrane bioreactors (MBRs), which are now widely used in full-scale WWT systems [29]. Their limited development and use [30] is attributed to the fact that anaerobic digestion is a complex process [11]. In the past, anaerobic digestion was, in general, avoided due to its major drawback, its slow-growing bacteria for which doubling time can vary broadly from 12 h to 1 week [31]. AnMBRs did manage to solve this problem due to the complete retention of the micro-oragnisms within their tanks; however, membrane fouling, the major drawback of all MBRs, appeared to be more intense in AnMBRs than in aerobic ones [30]. Finally, wastewater toxicity has also been considered one of the main reasons for a non-generalised use of anaerobic digestion, as these processes are not capable of tolerating it due to the fact that methanogenic micro-organisms can be easily inhibited by toxins [32]. Aerobic systems were developed more easily and quicker than the anaerobic ones because they are more flexible and capable of

Table 1
Summary of AnMBR performance for municipal wastewaters.

Case study	Type of wastewater	Scale	Working volume (L)	MLSS[A] ($g\ L^{-1}$)	OLR[A] ($kg\ m^{-3}\ d^{-1}$)	HRT[A] (h)	SRT[A] (d)	Temperature (°C)	Influent COD[A] ($mg\ L^{-1}$)	Effluent COD ($mg\ L^{-1}$)	Maximum COD removal (%)
[3]	Real municipal	L[a]	12.9	–[b]	2.36	2.6	–	15–20	162.3–603.2	48–107	–
[45]	Primary effluent from a full-scale WWT plant	L	10	7.3 (Max)	0.02–2.11	12–48	18–233	32	23–118 (soluble COD)	24–38	76
[65]	Raw and UASB effluents	P[a]	849	–	–	6	–	–	287–563	25–41	90
[50]	Organic waste mixture	L	0.5–0.6	–	–	2–20 d	–	35	–	44,599	99
[42]	Real municipal	L	5–15	1.05–2.4	–	–	–	33–37	480	30–50	~98
[63]	Final effluent containing nitrates	L	5.6	1.32–1.97 (Ave)	–	3	20	25–28	48–76 (soluble organic carbon)	13–21	72
[40]	Real municipal	L	50	–	0.8–1.2	–	–	37	419–900	–	76
[51]	Municipal waste mixture	L	3	8.3–21	–	4.4	300	34–36	–	15–20	–
[49]	WWT plant secondary effluent	L	2.4	–	1.1–3.7 (kg_{VSS}[A] $m^{-3}\ d^{-1}$)	3–8 d	–	33–37	–	–	–

[A] COD: Chemical Oxygen Demand, HRT: Hydraulic Retention Time, MLSS: Mixed-liquor Suspended Solids, OLR: Organic Loading Rate, SRT: Solids Retention Time, UASB: Upflow Anaerobic Sludge Blanket, VSS: Volatile Suspended Solids, WWT: Waste Water Treatment.
[a] L: Laboratory, P: Pilot.
[b] Value not reported.

Table 2
Summary of AnMBR performance for synthetic wastewaters.

Case study	Type of wastewater	Scale	Working volume (L)	MLSS[A] ($g\ L^{-1}$)	OLR[A] ($kg_{COD}\ m^{-3}\ d^{-1}$)	HRT[A] (h)	SRT[A] (d)	Temperature (°C)	Influent COD[A] ($mg\ L^{-1}$)	Effluent COD ($mg\ L^{-1}$)	Maximum COD removal (%)
[15]	Sucrose-based	L[a]	3	11.45–16.12 (VSS[A])	6–16	6–40	~250	34–36	4000	31–484	98
[24]	Sucrose-based	L	3	1.68–9.69 (VSS)	4–4.8	15–80	~150	34–36	4000	160–240	96
[14]	Meat extract/Peptone-based	L	3	2.5–3.9 (VSS)	–[b]	6	150	34–36	430–470	7–29 (Soluble COD)	96
[29]	Synthetic sewage	L	10	–	~5	24	50	30	500	20	>96
[44]	Synthetic simulating municipal	L	4	6–14	1	12	–	14–26	500	~40 to ~200	95
[52]	Synthetic simulating municipal	L	5	5–11.24	1.1–1.65	8–12	30–Infinite	25–30	550	–	97
[36]	Glucose-based	L	3	3.5–5.5	–	3–48	–	35	150–920	21.76–50.38	95
[78]	Synthetic simulating municipal	L	3	4.3–5.02	–	3–24	-	35	460	27.1–47.9	95
[23]	Low-strength	L	5 (Total)	4.3–5.72	1.1	12	30–60	25–30	550	5	99
[79]	Volatile fatty acid mixtures	L	3.7	37–43	–	–	–	30–55	–	–	–
[57]	Volatile fatty acid mixtures	L	3.7	35–40	10–70	–	–	30	5000–10,000	–	–
[62]	Volatile fatty acid mixtures	L	3.7	35–40	10–40	–	–	55	5000–10,000	–	–
[55]	Volatile fatty acid mixtures	L	3.8	13–35	<15	–	–	30–55	10,000	–	–
[74]	Volatile fatty acid mixtures	L	2	41 (Final)	10–15	–	–	55	10,000–17,000		
[56]	Synthetic simulating alcohol distillery wastewater	L	4.5	1.3–1.9	4	6.5 d	–	54–56	4200–5800	–	>84
[6]	Sodium acetate/Sodium propionate-based	L	2	–	4.1–6.2	1.8–3	–	35	513	3–11	99
[84]	Synthetic containing formic acid	L	10.9	1.03–1.81	–	8	–	31–35	–	–	–
[47]	Synthetic simulating municipal	L	50	~0.5 to ~4	1	–	–	37	800–1200	–	–
[30]	Whey/Sucrose-based	L	11	5.5–20.4	1.5–13	–	30–40	34–36	–	–	–
[76]	Synthetic of COD of 800 $mg\ L^{-1}$	L	25 (Total)	4–10	0.46–5.76	10.4	Infinite	–	800–2500	–	85
[2]	Synthetic sewage	L	3	–	2	20	250	34–36	445–485		98.8 (dissolved organic carbon)
[43]	Synthetic with nitrates	L	4.8	2.23	–	2 d	35	–	87–191	–	–
[60]	Molasse-based	L	9	1.6–10 (VSS)	5–12.2	–	–	27–33	700–24,200	81	–
[64]		L	45	5.9–19.8 (VSS)	–	8	–	10–15	87–154.8	51.2–63.4	95

[A] COD: Chemical Oxygen Demand, HRT: Hydraulic Retention Time, MLSS: Mixed-liquor Suspended Solids, OLR: Organic Loading Rate, SRT: Solids Retention Time, VSS: Volatile Suspended Solids.
[a] L: Laboratory.
[b] Value not reported.

Table 3
Summary of AnMBR performance for wastewaters other than municipal and synthetic.

Case study	Type of wastewater	Scale	Working volume (L)	MLSS [A] ($g\,L^{-1}$)	OLR[A] ($kg_{COD}\,m^{-3}\,d^{-1}$)	HRT[A] (h)	SRT[A] (d)	Temperature (°C)	Influent COD[A] ($mg\,L^{-1}$)	Effluent COD ($mg\,L^{-1}$)	Maximum COD removal (%)
[59]	Landfill leachate	L[a]	29 (Total)	–[b]	0.7–4.9	24–168	–	35	5000	417	95
[11]	Thermo-chemical whitewater	L	10 (Total)	4.9–10.7	2.0–2.8	–	~280	36–38	2782–3350	300	90
[75]	Thermo-chemical whitewater	L	10	8.3–9	1.66–1.94	–	–	36–38	1823–3504	217.5–421.1	87
[37]	Kraft evaporator condensate	L	10 (Total)	3.5–8.5	1–7	–	200–260	37–56	2400–2600	50–200	95
[54]	Kraft evaporator condensate	L	10 (Total)	–	2.3–13.3	–	–	36–56	9500–10,500	74–276	99
[85]	Kraft evaporator condensate	L	10	3.7–5.7	–	–	–	36–38	5500–10,000	63–192	–
[46]	Thermo-chemical whitewater	L	10	6.7–11.3	2.6–4.8	–	280	36–38	2782–3460	280–425	90
[67]	Swine manure	L	6	–	1–3 (kg_{VSS}[A] $m^{-3}\,d^{-1}$)	–	–	36–38	–	200–250	>96
[10]	Cheese whey-based	L	20	–	3–19.78	1–4 d	–	35–39	–	–	~98.5
[83]	Slaughter house wastewater	L	50	10.1	1.59–16.32	30–80	–	37	15,880	–	>99
[72]	Brewery wastewater	L	4.5	12–25 (VSS)	12	–	–	30	2300	190	99
[58]	Landfill leachate	L	3	7.2–10.8 (VSS)	8–11.8	1.1–19 d	30–300	10–35	–	–	>95 (soluble COD)
[25]	Fischer Tropsch acid water	L	23	30	25 (Max)	31.5	175	37	19,101	612	–
[41]	Dairy manure-based	P[a]	200	–	2.4 ($kg_{VSS}\,m^{-3}\,d^{-1}$)	9 d	28	–	–	–	92
[38]	Kraft evaporator condensate	L	3.5	2.1–24	1–24	–	–	36–38	5600–10,000	50–200	99
[31]	Landfill leachate	L	50	<3 (VSS)	1–6.27	7 d	–	37	15,000–41,000	960–4100	>92
[48]	Swine manure	L	5	–	1–2 ($kg_{VSS}\,m^{-3}\,d^{-1}$)	6	118–211	–	–	–	>95

[A] COD: Chemical Oxygen Demand, HRT: Hydraulic Retention Time, MLSS: Mixed-liquor Suspended Solids, OLR: Organic Loading Rate, SRT: Solids Retention Time, VSS: Volatile Suspended Solids.
[a] L: Laboratory, P: Pilot.
[b] Value not reported.

accommodating the net bacterial growth rate at shorter SRTs, and can be operated efficiently at lower temperatures [33,34].

2.2. Energy recovery

AnMBRs can also play a key role in energy recovery due to their capacity to produce CH_4 from the utilisation of a large fraction of organics in wastewaters [35]. AnMBRs can convert up to 98% of the influent COD into biogas [25]; moreover, due to the low growth yield of anaerobic micro-organisms, very small sludge production is normally observed in these systems [25]. In general, AnMBRs are capable of producing biogas of excellent fuel quality, in some cases, having a composition of 80% and even 90% CH_4 [36–38], which can then be burnt to produce electric power, being in some cases able to cover all energy demand required for membrane filtration [27] and produce net energy for the WWT plant [25]. During the case study of Van Zyl et al. in 2008, 2.02 kW h $kg_{CODremoved}$ from synthetic wastewater were produced, an amount being ~7 times higher than the amount of electricity required for the operation of their system [25]. However, biogases less rich in CH_4 (but still of very high content), i.e. 70%, have also been reported in the literature [10]. This difference in CH_4 composition percentages comes from the fact that the CO_2/CH_4 ratio varies substantially depending on the characteristics of the organic compounds to be degraded. [32]. Organic wastes rich in carbohydrates, such as biowaste and corn silage, can improve the biogas production and the proportion of CH_4 [39]. In addition, the overall composition of the biogas varies according to the conditions prevailing in the MBR [32]. The higher CH_4 content achieved in AnMBRs, if compared with conventional anaerobic treatment processes, is due to the shorter HRTs that can be achieved by applying membranes for sludge separation. This leads to a larger removal of CO_2 than CH_4 from the effluent because of its much lower gas solubility (~10 times), according to the Henry's law.

2.3. Treated wastewaters

Since 2006, there have been only few scientific studies that dealt with pilot scale AnMBRs, namely that of Saddoud et al. [40] or that of Wong et al. [41] and no authors have produced any scientific article with respect to industrial-scale trials. Almost all authors worked with bench-scale (laboratory-scale) apparatuses [15,36,42–44]. In the studies reviewed, AnMBRs were used to treat a wide variety of wastewater types ranging from municipal wastewaters [3] and raw domestic wastewaters [36,40,42,45], to white waters from pulp and paper mills [11,46] or petrochemical effluents [25] – more details in Tables 1–3. Regarding municipal wastewaters in particular, both conventional MBRs and AnMBRs being operated under similar conditions ended up producing similar soluble COD removal efficiencies with AnMBRs avoiding at the same time all costs for aeration [45]. However, AnMBRs cannot respond properly when considerable fluctuations in wastewater composition occur, or when toxic compounds exist in the influent, as biomass may end up being unable to adapt itself to the environment; hence, under such conditions, steady state within the system may never be reached [47]. Toxicity is generally discussed in terms of toxic levels and not in terms of toxic material as any compound present in sufficiently high concentration is toxic. However, toxicity impact can be minimised by some design measures, such as application of long SRTs [32], which is the case for AnMBRs. In general, for wastewaters with compounds concentrations over the toxic level, the use of aerobic MBRs may be more sensible; however, application of some control methods, i.e. dilution below the toxic level [32], or removal of the toxic compounds before the application of the anaerobic treatment [32,37] can lead to a safe operation of AnMBRs as well.

Moreover, with respect to wastewaters with low organic content, it is advisable to operate at temperatures close to the ambient ones because the low methane production that is achieved may not be able to cover the heating costs; however, even though operation at ambient temperatures appears to be technically feasible, SRTs need to be lengthened [3]. On the other hand, AnMBRs can be successfully operated at high MLSS concentrations, as demonstrated with swine manure of 49 g L^{-1} [48] or municipal waste of 50 g L^{-1} [49].

2.4. Operating conditions

A large number of different combinations of operating conditions have been reported. Hydraulic retention time (HRT) values ranged from a few hours, i.e. ~2 h [6] to a few days, i.e. 20 d [50], whereas solids retention time (SRT) values ranged from a few days, i.e. 18 d [45] or 30 d [23] to about a year, i.e. 300 d [51] or even more, indicating that no sludge purging took practically place during the MBR operation [52] (Tables 1–3); most of researchers worked at SRT values higher than 150 d. As a general rule, the operation of AnMBRs at longer SRTs results in the generation of greater quantities of biogas [39] as any decrease in the SRT decreases the extent of the reactions and vice versa; so, short SRTs are insufficient for a stable digestion [53]. For example, Huang et al. in 2008 reported that 0.023 L_{CH4} $g_{MLVSS}{}^{-1}$ d^{-1} and 0.028 L_{CH4} $g_{MLVSS}{}^{-1}$ d^{-1} were produced at an SRT of 30 d and an SRT of 60 d respectively [23].

Most of the AnMBRs were operated either at around 35 °C in the mesophilic range [54,55] or at around 55 °C in the thermophilic range [54,56,57]; even though psychrophilic temperatures of around 20 °C were also tested [58]. The temperature of the mixed-liquor affects the COD removal efficiencies; higher temperatures lead to better COD removal efficiencies. For example, Ho and Sung in 2010 operated two AnMBRs, one at 25 °C and another at 15 °C and they concluded that total COD removal efficiency achieved was over 95% and over 85% respectively [33]. In addition, it was found that AnMBRs operating in the thermophilic range of temperatures managed to cope with higher volumetric loading rates than AnMBRs operating in the mesophilic range. Jeison et al., in 2008 claimed that a value of 14 g_{COD} L^{-1} d^{-1} could be maintained by a thermophilic AnMBR, whereas at the same time a mesophilic one could not maintain values higher than 10 g_{COD} L^{-1} d^{-1} [55]. In general, when the organic loading rate increase, the risk of a deteriorated performance due to volatile fatty acids (VFAs) accumulation may be run, and lower COD removal efficiencies are achieved due to inhibition of microbial activity [41,59,60].

2.5. Removal efficiencies

The most important target to achieve during an AnMBR operation is to reduce the organic carbon content in the influent before its discharge or re-use as final effluent. To assess the achievement of this purpose, most research groups measured the organic carbon concentrations both in the influent and in the effluent to estimate the overall removal efficiencies. Depending on the wastewater tested, COD concentrations in the influent ranged from low values in the range of about 162 mg L^{-1} [3] to high values in the range of 10,000 mg L^{-1} for kraft evaporator condensate [54] or even 18,000 mg L^{-1}, for high-strength petrochemical effluent, mainly loaded with short-chain (C_2 to C_6) fatty acids [25]. Removal efficiencies have varied from 76% [40] up to 99% [23,54] (Tables 1–3). Regarding 5-day biochemical oxygen demand (BOD_5) removal efficiencies, values higher than 99% were reported [10,54]. Furthermore, it may be worth mentioning that the biomass attached on the membrane does not play a significant role in the biological removal of organics compared to the suspended biomass in the mixed-liquor [44].

Regarding TSS removal efficiencies, very high values were reported in the references; in particular, values higher than 99% were reported [10,42]. Also, regarding pathogens, namely *Escherichia coli* and *Enteroccoci*, total removal can be achieved; so, most of the time the effluents can be suitable for re-use in unrestricted crop irrigation [40], which is officially defined as the use of treated wastewater to grow crops that are normally eaten raw [61].

Finally, it is worth mentioning that pH shocks cause significant long-lasting negative impacts on the COD removal efficiency [11]. Gao et al. in 2010 carried out an experiment during which pH shocks were applied to the reactor liquor resulting in changes both in biogas production and membrane filtration performance [11]. It was observed that a pH 8.0 shock had a minor impact on COD removal, whereas a pH 9.1 and a pH 10 shock had a significantly negative effect. During the pH 9.1 shock, the COD removal efficiency was rapidly reduced from almost 90% to less than 75%, whereas, during the pH 10 shock, it diminished from 90% to values less than 30% [11].

2.6. Energy consumption and costs

During the time period considered in this review, no significant attention was paid to energy consumption issues, despite the fact that the use of AnMBRs can lead to energy-sustainable operation. The authors of the present work believe that further research on energy consumption aspects of AnMBRs would be beneficial. An interesting study on energy issues of AnMBRs is the one by Kim et al. who after operating an AnMBR preceded by a conventional anaerobic reactor, concluded in 2011 that the total energy required for fluidisation in both reactors was equal to 0.058 kW h m^{-3}, an energy amount that could be covered by using only 30% of the methane produced, with AnMBR consuming 0.028 kW h m^{-3} [6].

Also, Jeison and van Lier, after making a series of rational assumptions, concluded in 2007 that membrane costs were a lot higher than the energy consumption costs in AnMBRs – values of 0.5 € $m^{-3}_{\text{treated water}}$ and 0.046 € $m^{-3}_{\text{treated water}}$ were respectively estimated [62]. Thus, despite the fact that membrane costs have dramatically decreased over time, they can still be an issue restricting potential applications of AnMBRs.

To conclude, it is worth mentioning that the AnMBR technology offers the potential to be combined with other treatment technologies in a flexible way. AnMBRs have been combined with a hydrolytic reactor [51] aiming at the hydrolysis of the wastewater organic matters; a hydrogen delivery system [63] aiming at the de-nitrification of a municipal effluent; they have also been integrated in a acidogenesis optimisation via a central composite design aiming at the enhancement of methane production [50], or were operated in line with conventional anaerobic digesters [6], etc.

3. Membrane fouling issues

Membrane fouling is one of the main disadvantages of MBRs, because it hinders the operation of the systems in a constant, reliable way. Although the deposition of solids on AnMBR membrane

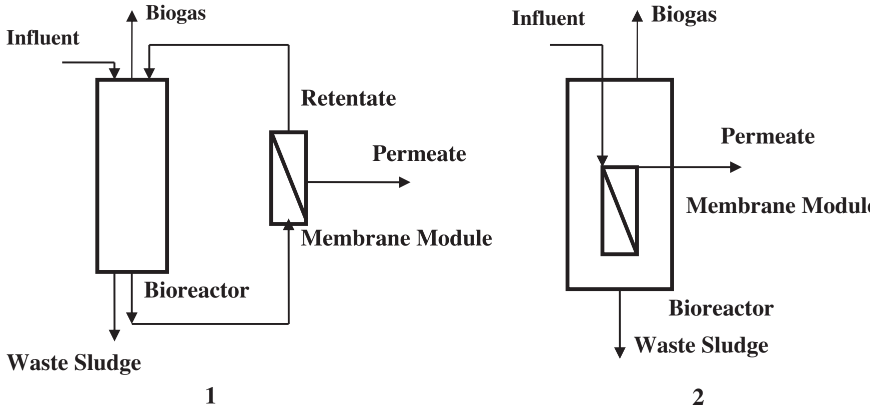

Fig. 1. A schematic of AnMBR configurations – (1) Side stream (external) AnMBR, (2) submerged (immersed) AnMBR.

Table 4
Membrane characteristics for flat sheet membranes.

Case study	Filtration range	Pore size (μm)	Area (m^2)	MPF[A] ($L\ m^{-2}\ h^{-1}$)	TMP[A] (kPa)	Gas flow rate ($L\ min^{-1}$)
[15]	MF[a]	0.4	0.1	2–10	0.23 bar	5
[24]	MF	0.4	0.1	1.5–2 (Max)	–[b]	2–5
[14]	MF	0.4	0.1	10–20	0–0.3 bar	5
[11]	MF	70,000 Da[c]	0.03	4.7–5.7	<30	0.75
[29]	MF and UF[a]	MF: 100,000 Da UF: 30,000 Da	260 cm^2	4–12	–	–
[75]	UF	70,000 Da	0.03	4.35–4.85	43.5	–
[78]	MF	0.4	0.1	10–20	0.1–0.3 bar	5
[23]	–	0.45	0.236	5.3	–	–
[52]	MF	0.45	0.118	4.3–7.9	<30	–
[42]	–	0.2	0.003	80–450	20–125	–
[37]	MF	0.3	0.03	1.6–6.3	30	0.25–0.75
[54]	MF	70,000 Da	0.03	1.8–8.1	<30	0.75
[85]	MF	70,000 Da	0.03	5.3–9.2	35	0.15–0.75
[46]	–	70,000 Da	0.03	4.8–9.1	<40	0.75
[30]	MF	0.4	0.12	2–12	<50	–
[51]	MF	0.4	0.1	3–7.2	175[d] mbar	5
[58]	MF	0.4	0.1	–	–	5
[25]	–	–	0.351	20–30	–	–
[2]	–	0.4	0.1	5–8	0.33–0.41 bar	5.5
[38]	MF	0.3	0.03	4.6–10.1	<30	0.30–0.75

[A] MPF: Membrane Permeate Flux, TMP: Transmembrane Pressure.
[a] MF: Microfiltration, UF: Ultrafiltration.
[b] Value not reported.
[c] Da: Dalton.
[d] Value corresponding to critical flux.

surfaces is lower than on aerobic MBR membrane surfaces, as AnMBRs are usually operated at lower membrane permeate fluxes [45], AnMBRs are characterised by lower sludge filterabilities, which favour membrane fouling [30].

3.1. Membranes

In this review, the working volumes of bench-scale MBRs ranged from 0.6 L [50] to 3 L [15] up to values as high as 45 L [64] and 50 L [42], even though most of them were operated at volumes between 5 L and 10 L, (Tables 1–3). At the same time, values in the range of 850 L were reported for pilot-scale systems [65]. AnMBRs that were trialled were either of side stream (external) configuration [15,23,25,66] or of immersed (submerged) configuration [48,51,54,67], (Fig. 1). The membranes used were mainly flat sheet (FS), like KUBOTA [30,36,68] (Table 4), or hollow fibre (HF), like Mitsubishi Rayon [36], (Table 5) or tubular [48,67] (Table 6). Tubular membranes were usually located externally. The materials used for their construction were mainly polymers, like poly-ethersulphone [48], poly-ethylene [15], poly-vinylidene fluoride [29], poly-tetrafluoroethylene [44], etc.; however, non-polymeric materials like ceramics were also tested [56], but not as widely. Ceramic membranes may be a good choice for AnMBR applications due to evidence of less membrane fouling and the ability to clean them without affecting their life span negatively [69]. Also, Jeison et al. in 2008 tried to operate AnMBRs without installing a real membrane, but by generating a self-forming dynamic membrane requiring only a support material made either of woven or non-woven materials, over which a cake layer made of microbiological material in the mixed-liquor was formed [22]. In general, the non-woven material, which acts as a dynamic membrane, is low-cost material leading to the development of cost-effective membranes, [3,70]. However, operation of the dynamic membrane was unstable; with sudden increases in filtration resistance and low membrane permeate fluxes, due to incapability of properly controlling the formation of the cake layers [22].

Regarding filtration, both microfiltration (MF) [42] and ultrafiltration (UF) [25,48,67] membranes were used, with membrane

Table 5
Membrane characteristics for hollow fibre membranes.

Case study	Filtration range	Pore size (μm)	Area (m^2)	MPF[A] ($L\,m^{-2}\,h^{-1}$)	TMP[A] (kPa)	Gas flow rate ($L\,min^{-1}$)
[59]	UF[a]	0.1	0.46	–	–	–[b]
[65]	UF	100,000 Da[c]	5.02	–	–	–
[6]	–	0.1	0.091	7–10	0.1–0.35 bar	–
[63]	–	0.04	0.094	–	–	–
[76]	–	0.4	0.05	24 (Initial)	–	–
[43]	–	0.4	0.1	–	0.045 MPa	–
[49]	–	0.4	0.12	1.3–3.5	–	–

[A] MPF: Membrane Permeate Flux, TMP: Transmembrane Pressure.
[a] UF: Ultrafiltration.
[b] Value not reported.
[c] Dalton.

Table 6
Membrane characteristics for tubular membranes.

Case study	Filtration range	Pore size (μm)	Area (m^2)	MPF[A] ($L\,m^{-2}\,h^{-1}$)	TMP[A] (kPa)	Gas flow rate ($L\,min^{-1}$)
[3]	–[a]	0.64	0.98	5	<30	–
[45]	–	0.1	0.1	–	<0.1–15 psi	–
[44]	MF[b]	1	0.09	5	6.9–55.2	–
[79]	–	–	0.042	6–17 (Critical)	<25[c] mbar	–
[57]	MF	0.2	0.042	15–25	<400 mbar	60–65
[62][d]	MF	0.2	0.042	<10 (Critical)	–	70
[55]	MF	–	0.042	4–20	0.2 bar	70–75
[66]	–	0.2	0.013	20–50	–	–
[74]	–	0.2	0.013	6.5–20	–	–
[56]	–	0.14–0.2	1	–	1 bar	–
[67]	UF[b]	20,000 Da[e]	0.0377	5–10	0.3–0.7 bar	–
[72]	–	0.2 30 nm	–	4 to >20	–	–
[41]	–	–	0.126	–	–	–
[48]	UF	20,000 Da		5 to >100	20–70	–

[A] MPF: Membrane Permeate Flux, TMP: Transmembrane Pressure.
[a] Value not reported.
[b] MF: Microfiltration, UF: Ultrafiltration.
[c] Value corresponding to critical flux.
[d] A UF membrane was also tested.
[e] Dalton.

pores ranging from 0. 45 μm [52] or 0.4 μm [36,47] in the MF region to values as low as 20,000 Da in the UF region [67], (Tables 4–6). Cross flow filtration was also preferred, [42].

Transmembrane pressure (TMP) values were found to be higher across HF membranes than across FS membranes when operated under similar conditions, making HF membranes more susceptible to fouling [36]. Even though, under normal conditions, there should not be any difference in TMP values in membranes having the same surface area, material and pore size [36], Hu and Stuckey in 2006 showed that HF membranes, during air sparging, may experience insufficient hydraulic shear on each of their fibres, whereas FS membranes have higher-shear slugs passing across their surface [36].

3.2. Parameters influencing membrane fouling

With respect to MBR configuration, membrane permeate fluxes in side-stream AnMBRs treating municipal wastewaters are mostly influenced by TMP values and cross-flow velocities, whereas, in submerged anaerobic membrane bioreactors (subAnMBRs), they are influenced by TMP values, gas sparging intensities, and the duration of membrane relaxation [71]. However, gas sparging applied in external configurations with tubular membranes led to an increase in the membrane permeate fluxes; so, introduction of gas/liquid two-phase flow inside tubular membranes can additionally be a way of controlling fouling in such applications [72].

External AnMBRs are, in general, expected to perform differently to submerged anaerobic membrane bioreactor (subAnMBR) systems, as the magnitude of shear forces to which their biomass is exposed is significantly greater. Shear forces play a really important role in membrane fouling, both positive and negative. High shear stress reduces both the microbial activity and the size of the bioflocs, increasing at the same time the release of soluble microbial products (SMP) in the mixed-liquor [54,66], and, hence, more membrane fouling is expected. On the other hand, high shear stress increases particle back-transport on the membrane, a principle used during membrane gas-sparging with a positive effect on membrane fouling [65,71]. Feed toxic shock can also cause sludge de-flocculation; thus, entailing a decrease in the membrane performance [37]. Membrane material can also affect membrane fouling [29,71]. For example, Gao et al. operated in 2010 two different kind of membranes, namely one made of poly-vinylidene fluoride coated with PEBAX and another made of poly-etherimide, both treating wastewater in the UF region, and they concluded that the latter became fouled faster than the former [29].

Finally, it is worth mentioning the relationship between membrane fouling and membrane nominal pore size. The optimal membrane pore size is related to the specific mixed population that is being filtered. Initial membrane permeate fluxes are usually greater for membrane with larger pore sizes; however, their rate of fouling is higher. That was mainly due to internal pore fouling as cake formation is independent from pore size [71].

3.3. Foulants, types of fouling and the role of SRT/HRT

Potential foulants in AnMBRs are inorganic precipitates, or biological–organic material like SMP (in particular, anaerobic SMP were found to be more complex than the aerobic ones), extracellular polymeric substances (EPS) mainly bound protein-based EPS, biopolymer clusters (BPC) and microbial cells, with biomass composition playing an important role in membrane fouling [3,11,23,54]. EPS are generally defined as polymeric material bound to cell surface which are extracted by using different chemical and physical methods. SMP are defined as microbial products released into the bulk solution as a result of the cell lysis, the hydrolysis of EPS as well as of the interaction of the micro-organism with its surroundings. Therefore, while EPS are, by definition, of extracellular origin, SMP originate from cell lysis and decay [14]. Finally, BPC are a solute independent of the biomass and they are much larger than SMP in the sludge suspension [73]. The main components of the organic matter in the membrane foulants were identified by Fourier transform infrared spectroscopy as proteins, polysaccharides and humic acids [11].

Membrane fouling can be either internal, due to membrane pore clogging/blocking, or external, due to cake formation with cake layers being defined as porous layers rejected on the membrane surface [27]. Internal fouling is usually irreversible, compared to cake formation, which is usually reversible [74]. However, even reversible fouling can never be completely removed [65]. Finally, even though high molecular weight (MW) protein and carbohydrate compounds and EPS can cause internal membrane fouling, it was showed that in the inner pores of membranes only inorganic material was deposited [2,14,48].

Applied SRTs play an important role in irreversible membrane fouling. MBR operation at longer SRTs is able to lead to worse internal pore blocking, possibly due to higher concentrations of responsible foulants [23]. Also, longer SRTs may lead to higher carbohydrate and protein concentrations in SMP as well as they can result in less flocculation of particulates and smaller particle sizes, and, hence, to accelerate membrane fouling [52]. Regarding HRTs, Huang et al., concluded in 2011 that a decrease in their values enhances growth of biomass leading to accumulation of SMP within the MBR tank leading to acceleration of membrane fouling [52]. Longer SRTs combined with shorter HRTs lead to both higher MLSS and SMP concentrations within the MBR tanks speeding up particle deposition and causing cake formation.

3.4. Cake formation

Small flocs, bound-EPS and inorganic materials played an important role in the cake formation process, with the cake layer being found to have a highly heterogeneous structure [46]. Cake sludge was found to have smaller particle size distribution, much higher specific filtration resistance, 1.5 times more bound-EPS and significantly different microbial community than the bulk sludge, [46].

Cake formation has been found to be the main factor governing both the applicable membrane permeate fluxes [57] and the critical fluxes [62]. Regarding critical fluxes, the small-sized particle concentration in the mixed-liquor is considered to be the main parameter inducing their low values [62]. Operating an AnMBR in the short-term at a membrane permeate flux close to the critical value leads to reversible cake formation [57,62] that can be easily removed by flushing the membrane [48]; on a long-term basis, however, cake consolidation occurs, with back flushes being unable to remove it. Despite the fact that small flocs have a higher tendency to accumulate on the membrane surfaces, consolidated cake usually results from large flocs and it mainly exists at the bottom cake layers. In addition, bound-EPS density increases from top to bottom cake layers [75]. Once membrane permeate fluxes higher than the critical value are applied, cake formation proceeds fast, [57,62].

3.5. The effect of temperature

Under similar hydrodynamic conditions, thermophilic subAnMBRs were found to have a filtration resistance 5–10 times higher than that of mesophilic AnMBRs [54]. Thermophilic subMBRs were shown to lead to the production of more BPC, more SMP and a larger portion of fine flocs as well as thermophilic sludge was found to have a higher protein/polysaccharide ratio in EPS; thus, their filtration resistance increases [46,54]. In addition, sludge cake layers in a thermophilic subAnMBR were found to be more compact and less porous [54]. Finally, at lower temperatures of 20 °C, higher concentrations of SMP are present in the mixed-liquor leading to a reduction in the membrane permeate fluxes [58,71].

3.6. Measures for membrane fouling mitigation

Different measures can be applied to mitigate membrane fouling, i.e. a short-term operation of the AnMBR as a conventional anaerobic reactor during the start-up process, to waste some of the fine particles of the feed, can be an effective strategy to reduce membrane fouling [37].

Cross-flow operation of membranes of AnMBRs is also able to reduce particle deposition on the membranes [57], with membrane performance being benefitted each time an increase in cross flow velocities occurs [67]. However, even though the resistance resulting from concentration polarisation and cake formation can be significantly decreased by increasing the cross-flow velocity, a plateau is reached at a Reynolds number of about 2000, for which no further decrease in the resistance can be achieved each time an increase in the cross-flow velocity happens [71]. Also, increases in cross-flow velocities come at a cost [71]. On the other hand, application of high shear stresses can also affect negatively the membrane permeate flux in the AnMBR [71].

Application of ultrasonic irradiation can be effectively used to control membrane fouling [49,76,77], with total filtration resistance being only 30% of that without the application of ultrasonic irradiation [76]. Higher sludge concentrations require longer ultrasound irradiation times, or higher amounts of ultrasound energy, for removing membrane fouling successfully [49,76].

Gas sparging is a way of in situ membrane cleaning with biogas being recycled back to the MBR. The efficiency of this technique will depend on the biogas sparging rate applied, as the critical flux increases and the membrane fouling rate decreases when biogas sparging rate is increased [38]. However, it would be more preferable to maintain membrane permeate fluxes at low values, as membrane scouring with gas may not be very efficient at high values [55]. In addition, membrane fouling can be promoted when gas sparging is turned from a continuous mode into an intermittent one. Vyrides and Stuckey in 2009 concluded that reduction of continuous biogas sparging to intervals of 10 min on and 5 min off resulted in a slight increase in the TMP values by 0.025 bar during their experiments [2].

Addition of activated carbon has also a positive effect on membrane fouling as operation with powdered activated carbon (PAC) leads to remarkably lower TMP values and less membrane fouling. Vyrides and Stuckey showed in 2009 that the addition of PAC inside their MBR resulted in a decrease in the TMP by 0.07 bar [2]. Both PAC and granular activated carbon (GAC) can provide a solid support for biomass growth; so, its use implies a reduction in floc breakage [2]. PAC seems to have a better performance than GAC as it leads to greater absorbance of low and high MW biodegradable matter and fine colloidal particles, on the carbon surface [14,15,78]. As addition of activated carbon remarkably reduces the number of bioflocs on the membrane surface and the SMP inside the MBR tank, less membrane fouling is expected [2], and, hence, lower TMPs can be recorded or higher membrane permeate fluxes can be sustained [14,36]. This conclusion is also supported by the fact that PAC is also characterised by high back transport velocities [15].

Specifically for membrane scaling caused by the precipitation of inorganic species, the use of a dialyzer–zeolite unit can be useful, as precipitation can be significantly reduced. However, the dialyzer–zeolite unit was successful only with ceramic membranes, whereas, with polymeric membranes, no significant improvement in membrane permeate flux was observed. This was due to the fact that fouling of the polymeric membranes was mainly due to deposition of biomass than the struvite precipitation [56].

Finally, it is worth mentioning the attempt of Jeison and Van Lier, in 2006, who, by operating two subAnMBRs, presented a new operation strategy based on a continuous critical flux determination; hence, avoiding excessive cake-layer accumulation on the membrane surface [79]. Therefore, each time cake-layer formation was detected, a decrease in membrane permeate flux or an increase in cross flow velocity was immediately applied. The proposed approach then allows the MBR to operate around the critical flux all the time, minimising the maintenance and maximising the efficiency of the performance [79].

4. Microbiological issues

AnMBRs are excellent systems for cultivating slow-growing micro-organisms, [80]. Biomass can be both suspended and attached with suspended biomass increasing over time, playing the most significant role with respect to microbial activity, and being the main contributor to biological removal of organics [44]. Suspended biomass also led to a significantly higher methanogenic activity compared to attached biomass. Methanogenic activity also increases with temperature. Ho and Sung in 2010 concluded that working at 25 °C was more efficient in producing methane than working at 15 °C. Also, longer SRTs or shorter HRTs have a positive effect on methanogenic activity, as biogas production, due to the increase in organic loading rate, was also found to increase [52]. However, the SRT increase in AnMBRs could result in inert solids accumulation [68] as for aerobic MBRs [81]. In addition, Padmasiri et al., in 2007 concluded that increases in cross flow velocity resulted in poor anaerobic digestion performance [67]. VFAs were found to have been accumulated, and the soluble COD both in the reactor and in the effluent was increased, as well as the biogas production was decreased [67].

Regarding micro-biological cultures within AnMBRs, there seems to be a significant difference between the microbial populations on the cake and in the bulk, as well as along the cake layer depth, with the outer cake surface being a lot looser than the inner surface of the cake. As the populations, which cause membrane fouling are of the utmost importance, it was found that their main populations of bacteria were affiliated to *Firmicutes* at a percentage of 42.3%, and *Alpha-proteo-bacteria*, at a percentage of 30.8%, while their populations of *archaea* were mostly affiliated to the *methanosarcinales* and *methano-spirillaceae*. *Sphingo-monadaceae*-related bacteria and *Methanogenic archaea* were then found to be components of biofouling, even after the application of chemical cleanings [65].

Regarding the effects of the application of ultrasonic irradiation on microbial activity, opinions are slightly divided. Sui et al., in 2008 found that it may have a slightly negative effect on the anaerobic bacterial activity, but without leading to obvious decrease in COD concentration removal rates [76], whereas, on the other hand, Xu et al., in 2011 claimed that it does not affect the anaerobic micro-organisms as well as it slightly improves their digestion performance [49].

5. AnMBR modelling

Little information is available with regards to AnMBR numerical simulations. Arros-Alileche et al., made in 2008 an attempt to model the operation of an AnMBR with respect to the assessment of the potential of controlled retention of solutes by the membrane [82]. Analysis of the data showed that low values of membrane retention in the MF or UF region require long HRTs to achieve good water quality, whereas high values of membrane retention in the nanofiltration (NF) or reverse osmosis (RO) region require short HRTs, or otherwise permeate of good quality could not be produced. Also, Jeong et al., in 2010 investigated the applicability of a system combining optimisation process and subAnMBR technology in the effective treatment of a high-strength organic waste; hence, ensuring satisfactory permeate quality and enhancing at the same time the amount of methane produced [50].

6. Summary of review and research needs

This review provides detailed information about the current applications of the AnMBR technology in the WWT sector. The research carried out during the period considered in this paper indicates that only the performances of bench-scale AnMBRs have been thoroughly studied, whereas pilot plant implementation is limited and no industrial-scale AnMBRs have been trialled.

In general, the literature proves that AnMBRs can treat very efficiently wastewaters of a variety of strengths and compositions producing a pathogen-free treated water of excellent quality. Also, they can produce biogases of good fuel quality, which can be further used within the AnMBR plants for the production of renewable energy. However, the adoption and commercialisation of this technology at industrial scale is still pending due to a number of reasons; membrane fouling and membranes sensitivity to toxicity are the main bottlenecks.

Membrane fouling in AnMBRs is more intense than in aerobic MBRs as AnMBRs experience lower sludge filterabilities. It is therefore imperative that further research is carried out to mitigate this problem. Cake formation on the membrane surfaces was found to be the key parameter that governs the applicable membrane fluxes. As AnMBRs are generally operated at lower membrane fluxes than the aerobic MBRs, it is critical to find ways of slowing down cake formation, as continuous reductions in the applicable membrane fluxes will make their operations be uneconomical.

As membrane foulants have been thoroughly identified, it is important to apply techniques that minimise their concentrations in the mixed-liquor as much as possible. Avoidance of toxic shocks, careful selection of the SRT and HRT values, good control of the mixed-liquor temperature at values never lower than 20 °C, avoidance of pH shocks, etc. should be always taken into account. In addition, proper selection of the membrane materials, cross flow operation of the membranes as well as application of moderate but still viable membrane permeate fluxes can lead to a stable long-term AnMBR performance.

As membrane fouling cannot be avoided, a number of techniques to mitigate or slow it down have been applied. In subAnMBRs, gas sparging with the biogas produced is the most-widely used technique as the high shear forces generated can remove particles off the membrane surface. However, the authors believe that more in-depth knowledge of the effects of shear forces on AnMBR systems would be needed, as high shear forces apart from cleaning the membranes can lead to reductions in microbial activity, flock size and to the release of SMP in the mixed-liquor, which as a result, increase the chances of membrane fouling. This need for research is especially critical for external membrane configurations as, by definition, higher shear forces are always present due to the applied high cross-flow velocities. Finally, the addition of active carbon, particularly the addition of PAC and application of ultrasound irradiation were found to significantly mitigate membrane fouling, when applied.

More research should also be carried out with respect to energy aspects of AnMBR operations. So far, little information is available regarding the energy that is consumed by AnMBRs as a whole or by each of their components, and almost no studies have been made regarding the optimisation of their energy consumption. Estimations of specific energy demand (SED) values at pilots will clearly show whether or not AnMBRs can be successfully shifted onto industrial-scale applications.

It must also be investigated in detail to what extent the biogas produced in AnMBR can lead to sustainable-energy operations. In addition, the authors have observed that the coupling of maximum biogas production with sustainable membrane performance in AnMBRs has not been the focal point in the above-mentioned research works. In line with this argument, more AnMBR modelling should also be available in the future, as, at the moment, only a few researchers have tried to numerically simulate AnMBR performances.

Finally, the high cost of membranes is still a significant issue impeding a faster commercialisation of both aerobic MBRs and AnMBRs. In AnMBRs, the membrane costs appear to be up to 10 times higher than the energy consumption costs per m^3 of treated water. So, even though the membrane costs have been dramatically reduced over the time, they are still a critical issue for the application of AnMBRs.

As an overall conclusion, it can be said that AnMBR technology, in certain cases, has been found to be a very promising alternative to the aerobic MBR technology. It was proven to be as efficient as the aerobic MBR technology with the additional ability of producing net energy within the MBR system due to the production of biogas, and without the added cost of aeration.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.cej.2012.05.070.

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