

Anaerobic dynamic membrane bioreactor applied to wastewater treatment: a review

Jacob Fortuna José Chimuca*, Catarina Simone Andrade do Canto, José Tavares de Sousa, Valderi Duarte Leite, Wilton Silva Lopes

State University of Paraíba (UEPB). Baraúnas Street, 351, University District, Campina Grande, Paraíba, Brazil. CEP: 58429-500

Biorreactor dinámico anaerobio de membrana aplicado al tratamiento de aguas residuales: una revisión

Bioreactor de membrana dinàmica anaeròbica aplicada al tractament d'aigües residuals: una revisió

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ABSTRACT

Membrane bioreactors have been widely used in biological wastewater treatment. The membranes used in this type of technology are produced from organic or inorganic materials. However, membranes can also be formed from the deposition of solid particles, colloids, and polymeric materials, as well as microbial cells and flocs, on an inert support during the filtration process. When coupled to a bioreactor, they establish a unique system called a dynamic membrane bioreactor (DMBR). This type of bioreactor, while retaining the solids and microorganisms present in the system, removes both easy and difficult-to-degrade organic material, which reduces treatment costs and makes it advantageous compared to conventional membrane bioreactors (MBRs). In Brazil, this technology is relatively new and still little explored. Therefore, the present study aims to evaluate the DMBR's performance in anaerobic wastewater treatment systems. In addition to the advantages and disadvantages presented by this type of system compared to conventional MBRs (micro and ultrafiltration), the fouling phenomenon, its implications, and the theories that explain the formation of the dynamic layer are described. Finally, some challenges that still need to be overcome in the use of this technology are pointed out in order to be affirmed as a safe and robust tool for the biological treatment of domestic and industrial wastewater.

Keywords: dynamic membrane, fouling, wastewater treatment challenges, bioreactor performance.



RESUMEN

Los biorreactores de membrana se han utilizado ampliamente en el tratamiento biológico de aguas residuales. Las membranas utilizadas en este tipo de tecnología se fabrican a partir de materiales orgánicos o inorgánicos. Sin embargo, las membranas también se pueden formar a partir de la deposición de partículas sólidas, coloides y materiales poliméricos, así como células microbianas y flóculos, sobre un soporte inerte durante el proceso de filtración. Cuando se acoplan a un biorreactor, establecen un sistema único llamado biorreactor de membrana dinámica (DMBR). Este tipo de biorreactor, al tiempo que retiene los sólidos y los microorganismos presentes en el sistema, elimina tanto el material orgánico fácil como el difícil de degradar, lo que reduce los costos de tratamiento y lo hace ventajoso en comparación con los biorreactores de membrana (MBR) convencionales. En Brasil, esta tecnología es relativamente nueva y aún poco explorada. Por lo tanto, el presente estudio tiene como objetivo evaluar el desempeño del DMBR en sistemas anaeróbicos de tratamiento de aguas residuales. Además de las ventajas y desventajas que presenta este tipo de sistemas frente a los MBR convencionales (micro y ultrafiltración), se describe el fenómeno del fouling, sus implicaciones y las teorías que explican la formación de la capa dinámica. Finalmente, se señalan algunos desafíos que aún deben superarse en el uso de esta tecnología para afirmarse como una herramienta segura y robusta para el tratamiento biológico de aguas residuales domésticas e industriales.

*Corresponding author: jacobfortuna66@gmail.com

Palabras clave: membrana dinàmica, ensuciamiento, desafíos del tratamiento de aguas residuales, rendimiento del biorreactor

RESUM:

Els bioreactors de membrana s'han utilitzat àmpliament en el tractament biològic d'aigües residuals. Les membranes utilitzades en aquest tipus de tecnologia es produeixen a partir de materials orgànics o inorgànics. Tanmateix, les membranes també es poden formar a partir de la deposició de partícules sòlides, col·loides i materials polimèrics, així com cèl·lules microbianes i floculs, sobre un suport inert durant el procés de filtració. Quan s'acoblen a un bioreactor, estableixen un sistema únic anomenat bioreactor de membrana dinàmica (DMBR). Aquest tipus de bioreactor, tot i que reté els sòlids i microorganismes presents en el sistema, elimina tant material orgànic fàcil com de difícil degradació, fet que redueix els costos de tractament i el fa avantatjós en comparació amb els bioreactors de membrana (MBR) convencionals. Al Brasil, aquesta tecnologia és relativament nova i encara poc explorada. Per tant, el present estudi pretén avaluar el rendiment del DMBR en sistemes de tractament anaeròbic d'aigües residuals. A més dels avantatges i inconvenients que presenten aquest tipus de sistemes en comparació amb els MBR convencionals (micro i ultrafiltració), es descriuen el fenomen de l'encrassement, les seves implicacions i les teories que expliquen la formació de la capa dinàmica. Finalment, s'assenyala alguns reptes que encara cal superar en l'ús d'aquesta tecnologia per afirmar-se com una eina segura i robusta per al tractament biològic d'aigües residuals domèstiques i industrials.

Paraules clau: membrana dinàmica, contaminació, reptes de tractament d'aigües residuals, rendiment del bioreactor

INTRODUCTION

Membranes are barriers used both in separation and in biocatalytic processes, in which they usually act as supports for enzymes, cells or microorganisms. Since the processes of separation and biocatalysis are closely related to biochemical processes, membranes can be coupled to bioreactors, becoming the so-called membrane bioreactors (MBRs). MBRs perform the important function of separating the biocatalyst from reagents and products and, thus, carry out the separation and biocatalysis processes in a single step¹.

Membranes are generally produced from synthetic materials of organic or inorganic nature. Organic membranes are based on cellulose and modified organic polymers, such as cellulose acetate, polypropylene, polyethylene, and polycarbonate². The inorganic ones use ceramics and metals as raw materials. Ceramic membranes are typically used in industrial applications; however, when it comes to wastewater treatment,

they are economically impracticable due to their high commercial value. The metallic membranes have very specific applications, which can also be related to biotechnological treatment processes³.

However, membranes can also be formed by deposition of colloids or solid particles in suspension, such as microbial cells and flocs, on a porous material, which acts as a support for the formation of a biologically active layer during the filtration process⁴⁻⁷. In this case, there is the so-called dynamic membrane (DM) which, like synthetic membranes, can also be coupled to bioreactors and used in separation and biotransformation processes.

The use of dynamic membrane bioreactors (DMBRs) in ultrafiltration processes started in the mid-1980s⁸. A decade later, research focused on the treatment (aerobic/anaerobic) of wastewater was initiated⁵⁻⁶. Over the past 30 years, the results of numerous pieces of research have shown efficiencies in the removal of suspended solids, biochemical oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon (TOC), nitrogen, and phosphorus comparable to those presented by reactors operating with conventional ultra and microfiltration membranes, thus confirming the viability of the use of DMs in biological wastewater treatment⁹⁻¹⁶.

The interest in anaerobic dynamic membrane bioreactors (AnDMBR) has been growing since 2007 and, although these systems have shown promising results in terms of treatment, research on the field is generally focused on optimizing the system's operating conditions^{8, 11, 17}. Despite the good results already found, much still needs to be clarified about this type of technology, which has shown itself as a viable alternative to the already established systems.

Therefore, the present literature review aims to evaluate AnDMBR performance in wastewater treatment systems, describe the advantages and disadvantages of using this type of system and the implications of the fouling formation, as well as the theories explaining the formation of the dynamic layer during the filtration process. Finally, some challenges that must be overcome to improve the functionality of this technology and make it more robust and reliable are described.

Dynamic Membrane Bioreactors

The dynamic membrane, also known as secondary or biological membrane, is usually formed on a cheap inert material, with pore size ranging from 10 to 200 μm and with almost negligible intrinsic resistance¹⁸⁻²¹. The support material can be polyester, polypropylene, polyethylene terephthalate (PET), stainless steel, cotton, nylon, non-woven fabric (TNT), activated carbon sponge, and even a synthetic membrane—usually used when the solution to be filtered contains suspended solid particles such as microbial cells, flocs, and colloids^{16, 22-23}.

Due to the support's pore sizes being relatively large, these materials do not act as filtration media, but, in fact, as supports. Organic and colloidal particles, which usually result in pore blockage in micro and

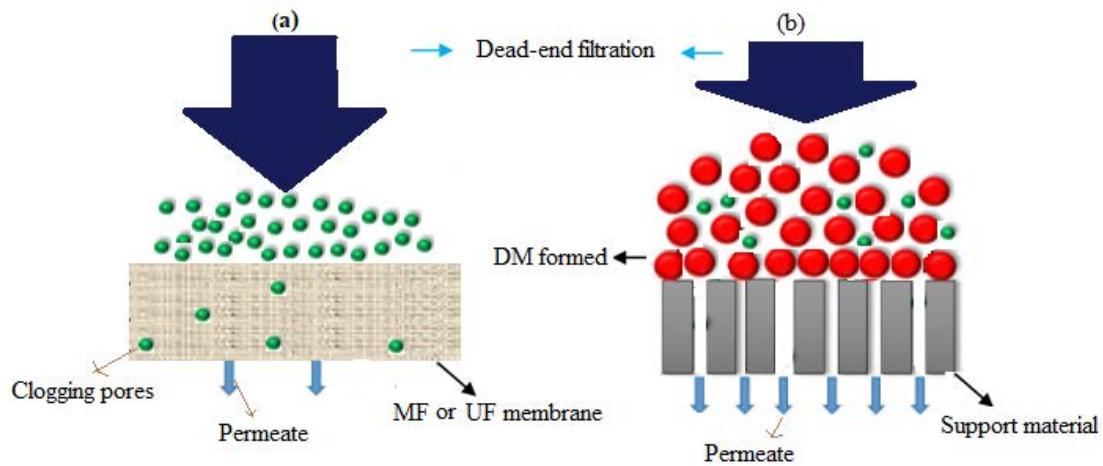


Figure 1. Demonstration scheme of membrane filtration under dead-end: (a) MF or UF membranes; and (b) DM formed on macroporous support material. Source: adapted from Pajoooh (2018)²⁵.

ultrafiltration membranes, are retained in the DM filtration layer, thus preventing the formation of scale (fouling) on the support material (Figure 1). Therefore, for the filtration process through DM, modules with cheap and macroporous materials are used, rather than relatively expensive membranes with extremely small pores^{18, 24}.

Compared to MF and UF membrane bioreactors, DMBRs have advantages including (i) 1 to 4 times greater flow; (ii) lower investment costs, since the support material used for the deposition of the dynamic layer is relatively cheap; (iii) high anti-pollution potential, which allows scale to be easily removed and the flow restored; (iv) low filtration pressure, allowing the effluent to flow by gravity, without the need for pumps, and therefore, saving energy; and (v) the ability to retain helminth eggs^{7-8, 18, 23-24, 26-29}. DM is also a promising technology for the removal of low-density, non-biodegradable microparticles such as microplastics, which are highly present contaminants in wastewater. These microparticles are not easily removed by conventional sedimentation, resulting in higher operating and maintenance costs in the unit processes subsequent to secondary treatment³⁰.

Despite having advantages, including economic ones, DMBRs present instability in their practical application. In addition, parameters such as cell retention time (CRT), hydraulic retention time (HRT), extracellular polymeric substances (EPS), permeate flow, and the sludge morphological properties (sedimentation, dehydration, flocculability, and hydrophobicity)—which are functions of feed, temperature, and concentration of dissolved oxygen in the medium—can negatively affect the bioprocess if not strictly controlled²⁸⁻²⁹. However, one of the main disadvantages observed in membrane systems is the phenomenon called fouling, defined as the unwanted deposition of particles, colloids, macromolecules, and salts on the membrane surface (external fouling) and/or inside its pores (internal fouling), which causes the reduction of flow through

the membrane, and consequently, a reduction in performance of the process^{2, 31-32}. Despite being an extremely complex phenomenon—as it involves a number of factors—examples of how it can be controlled include modifying the membrane surface, or optimizing the operating conditions, depending on the bioprocess in progress³³⁻³⁴.

It can be stated, however, that the continuous replacement of fine particles causing fouling on the surface of micro and ultrafiltration membranes is responsible for more than 80% of the filtration resistance, resulting from the formation of irreversible fouling. On the other hand, these same particles, as well as soluble microbial products (SMP) (proteins and polysaccharides) and the inorganic substances present in the medium, help the active biomass to form the dynamic layer, which has the dual function of both filtering and biologically treating wastewater, thus becoming an important and promising tool in the area of sanitation. In addition, through this technology it is also possible to obtain an effluent with quality similar to that of bioreactors that use MF and UF membranes, but significantly reducing process costs³⁵⁻³⁹.

The DM appears to be more suitable for anaerobic processes, due to the slow growth of microorganisms and the milder hydrodynamic condition resulting from the absence of aeration. The configuration, which can be internal or external, plays an important role in the effectiveness of the process. However, studies have shown that the internal configuration seems to be more beneficial, particularly in terms of the rapid DM formation/regeneration, which directly affects the treatment quality and efficiency^{17, 24, 40-41}.

It is also worth mentioning that the use of DM can increase the substrate retention capacity in an anaerobic dynamic membrane bioreactor (AnDMBR) and, subsequently, stimulate the increase of microorganisms responsible for the hydrolysis step in the digestion process due to the increase in the amount of hydrolytic enzymes (such as protease and β -glyco-

sidase). This can result in changes in the pathways of organic matter degradation. In addition, bacteria that are essential for the degradation of refractory organic matter, and which normally exhibit a low growth rate, can be retained and accumulated in the dynamic layer, allowing further degradation of refractory substances into more easily biodegradable by-products, such as volatile fatty acids²⁶.

Most studies have been carried out at bench and pilot scale, and although these systems have shown promising results comparable to systems using synthetic membrane bioreactors (commercial), research has generally been aimed at optimizing the operating conditions of bioreactors. Therefore, studies aiming, for example, at the elimination of pathogens and the recovery of products still need to be better investigated^{8, 11, 17, 27-28, 42}.

Dynamic membrane formation

There are several theories to explain how solids and other particles contained in the liquid medium agglutinate and settle on a support material. Among them, there is the extended Derjaguin-Landau-Verwey-Overbeek theory (XDLVO), whose objective is precisely to explain the adhesion mechanisms of biocatalysts on the surface of a support⁴³⁻⁴⁵. This theory can also be applied to the DM formation process that occurs according to the processes of adhesion and cohesion.

At the initial stage of DM formation, sludge flows from the bioreactor to the clean surface of the support. This process is dominated by the interactions between the support and the sludge flocs, and is defined as being the sludge adhesion behavior (adhesion process). In the next stage, named as the DM maturation stage, most of the membrane surface is covered by the sludge flocs. The interactions between the sludge flocs approaching the support and the already deposited flocs replace the interactions that predominate at the initial stage and begin to control the formation of the dynamic layer. Thus, the interactions between the sludge flocs, i.e. floc-floc interactions, are defined as being the sludge cohesion behavior (cohesion process)⁴⁶.

DM formation can also be described as being a dynamic process developed under hydrodynamic and thermodynamic forces. Hydrodynamic forces include permeate drag, inertial, shear, and gravitational forces. On the other hand, the thermodynamic forces are represented by the physicochemical interactions that occur in the short distance (d) between the support surface and the sludge flocs, and are defined as the Lifshitz-van der Waals (LW), Lewis acid-base (AB), and electrostatic double layer (EL) forces⁴⁷. Normally, the permeate drag force, which is induced by the flow through the membrane, can transport the sludge flocs to the surface of the support. The total forces (XDLVO forces), which are the sum of the components LW, AB, and EL, are attraction forces that may be responsible for fixing the sludge flocs on the surface of the support. Figure 2 shows the analy-

sis of the microcosmic force of a sludge floc located very close to the surface of the support. In general, from the XDLVO theory it is possible to predict, in a practical way, how the DM formation process takes place, also allowing to optimize the sludge properties, as well as the operation of the system.

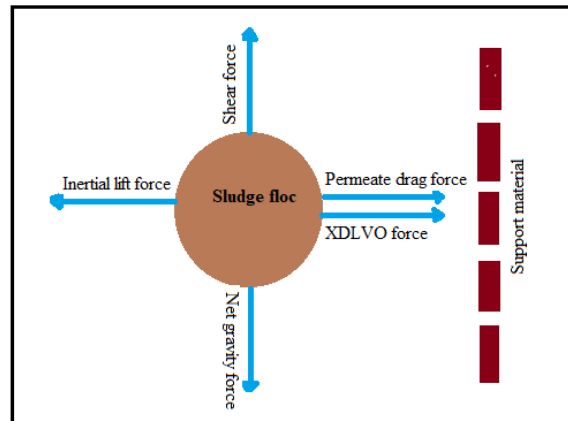


Figure 2: Analysis of the microcosmic force of a sludge floc located very close to the surface of the support. Source: adapted from YU *et al.* (2019)⁴⁶.

Considering the fluid mechanics fundamentals, the permeate drag force plays a function of great importance in the DM formation process when the support material, in addition to having relatively large pores, has hydrophilic characteristics. In this case, when the initial permeate flow is high, sludge flocs of larger size and with low relative hydrophobicity are quickly deposited on the surface of the support, causing an increase in filtration resistance, despite the dynamic layer having low specific resistance. However, as the membrane forms, the pores of the support and the flow through the membrane decrease, while the adhesion forces increase due to accumulation of the smaller sludge flocs and the production of the EPS. It is at this stage that the dynamic layer becomes more compact and with higher specific resistance. Thus, the movement of the sludge flocs depositing on the support material includes the permeate drag force and the shear force on the surface of the support material; which control, respectively, the convection speed and the reverse transport speed, both dependent on the permeate flow. In this way, the sludge floc deposition rate is governed by the liquid speed in the direction of the support material, since there is a balance between the convection speed and the reverse transport speed, although the role of the shear force is negligible in the process⁴¹.

After complete DM formation, good holding capacity of the small flocs is achieved and the permeate flow is substantially reduced, associated with a decrease in the DM porosity, and consequently, the formation of a more compact membrane. These factors contribute to a higher holding capacity, an increase in transmembrane pressure (TMP), and specific resistance during the filtration process.

Prior to the formation of a stable DM, the support material cannot induce solid-liquid separation, as the DM consists mainly of a cake layer formed from the

bioreactor sludge and a gel layer formed from the sludge metabolites, mainly EPS, which develop during filtration. In addition, the DM bioreactor's effluent quality in the first hours or even days is extremely low, and the sludge loss is significant. However, the quality gradually increases with prolonged operation. Sufficient time should be allowed for the filtration process to take place properly, so that a stable DM can form and the solid-liquid separation is effective, as is sludge retention by the system⁴⁸.

However, the macroscopic indicative of DM formation is a sharp reduction in permeate turbidity, along with a drastic decrease in the flow through the membrane^{46,49}. The DM maturation stage is characterized by a continuous decrease in permeate turbidity, a reduction in permeate flow, and an increase in filtration resistance. According to Hu *et al.* (2020)⁵⁰, the time required for the formation of a stable DM layer is in the range of 10-25 days during long-term operation (approximately 30-50 days), and under flow from 1.0 to 8.0 L.m⁻².h⁻¹.

Thus, there are many theories based on forces, velocities, microbial flocs and supporting material properties, as well as the operation characteristics and final effluent. However, it seems that there is consensus that a DM is formed in two stages: initial, and maturation.

Fouling in dynamic membrane bioreactors

Fouling can be understood as the set of phenomena capable of, over time, provoking a drop in membrane performance. Such phenomena arise when working with a solution or mixed liquor and its consequences can be completely or partially irreversible. Fouling causes the restriction and occlusion of the membrane pores through the deposition of organic and inorganic particles, which results in an increase in resistance to filtration, and consequently, reduction of the permeate flow⁵¹. Furthermore, the phenomenon has distinct characteristics and natures, i.e. it can be reversible and removable or non-reversible and non-removable and present an organic, inorganic or biological nature, as shown in Table 1.

Regarding DMBRs used in wastewater treatment, fouling can be affected by factors such as support material characteristics (material type and pore size), biomass characteristics (solids concentration, EPSs, soluble microbial products, floc structure and size), and operating conditions (TMP, permeate flow, HRT, CRT, cleaning periodicity)⁵².

The type of material from which the support is made strongly affects the DM bioreactors, although clear information about the real influence of this parameter on the performance of the dynamic layer itself is missing. However, it was observed that supports made of cotton promote greater efficiency of removal of organic matter and suspended solids, in addition to favoring the formation of a thicker and stronger dynamic layer. On the other hand, nylon, in addition to its low cost, allows the complete removal of fouling and the restoration of the structure through a simple physical cleaning, such as a backwash^{16, 22-23, 28, 39}.

More severe fouling is observed in hydrophobic supports than in hydrophilic ones, although changes in

hydrophobicity often occur as a function of other characteristics of the support, such as pore size and morphology, which makes the correlation between hydrophobicity and fouling to be more difficult to evaluate⁵⁷. Since the biomass has hydrophobic characteristics at different levels, its affinity, and consequently its adhesion to the support material surface with hydrophobic characteristics, is more intense than that observed with the use of hydrophilic materials⁵⁸.

On conventional membranes, if the particle size is larger than the pore size, there is blockage or restriction to filtration due to the formation of fouling. In DM systems, the support material pore size is also a key parameter for filtration performance through the dynamic layer, even when operating conditions favor the rapid formation of a more stable DM^{23,59}.

Table 1: types of fouling and their features.

Type of fouling	Description	Reference
Removable	Arising from particles that weakly bind to the membrane and are associated with the dynamic layer formation .	2
Non-removable	Usually caused by strongly adhered scale during the filtration process.	2
Organic	Caused by the deposition—on the membrane or inside its pores—of proteins, polysaccharides, humic acids, and other organic substances from the sludge, such as EPS and SMP.	53
Inorganic	Caused by the deposition—on the membrane or inside its pores—of inorganic compounds such as struvite (MgNH ₄ PO ₄ ·6H ₂ O), ammonia and potassium phosphate (K ₂ NH ₄ PO ₄), and calcium carbonate (CaCO ₃).	54
Biofouling	Formed due to the deposition and growth of microorganisms on the surface of the membrane.	55
Reversible	Occurs when deposition occurs only on the surface of the membrane, therefore, easily removed through merely physical processes.	56
Irreversible	Occurs when deposition is due to the adsorption of organic and inorganic substances in the pores of the membrane.	56
Colloidal	Refers to the membrane's encrustation with suspended and colloidal particles, such as microorganisms, polysaccharides, slime, lipoproteins, lignin, sulfur, and sulphides.	2

Results indicate a good filtration performance when the DM is formed on a support material with pore size ranging from 10 to 100 μm^{9-10, 28, 60}. For materials with a pore size less than 5 μm, blockage of 99% of the pores may occur⁶¹. However, if the support material has pores in the order of 10-40 μm, the formation of the DM is relatively fast and the effluent generated is of high quality. However, the pores of the support are easily blocked, decreasing the flow through the membrane, thus increasing the need for more frequent backwashings. In contrast, when the support material pore size is greater than 100 μm, there is a larger filtration area, higher fluxes, and lower filtration resistance, but

DM formation requires more time. Also, with larger pore sizes, it may not be easy to obtain high-quality effluents due to increased membrane instability (sludge loss). Therefore, to determine the most appropriate support material, pore size is a critical success factor in terms of dynamic layer formation time, flow, and pollutant removal^{16, 62}.

The total solids present in a biological reactor can be divided as follows: dissolved solids (less than 0.001 μm), colloidal (0.001 to 1.0 μm), and suspended solids (greater than 1.0 μm), and affect membrane behavior differently. Colloidal and soluble particles, for example, contribute to the total or partial blockage of pores, which can irreversibly affect membrane performance. In general, keeping the other operating parameters constant, the increase in the concentration of total solids leads to a decrease in the permeate flow due to the increase in the dynamic layer thickness. However, the layer thickness can be minimized by improving the hydrodynamic conditions of the system and, thereby, reducing the deleterious effects of fouling⁶³.

EPS consist of a complex mixture of proteins, carbohydrates, polysaccharides, DNA, lipids, and humic substances that are constituents of floc and biofilm matrices. On the other hand, SMP are a mixture of organic compounds resulting from metabolism and cell death during the substrate mineralization process. Such products are easily absorbed by the dynamic layer, being able to block the pores and reduce the permeability of the support material, as well as influence the structure and porosity of the formed dynamic layer⁶⁴⁻⁶⁶.

Total blockage of pores by EPS and SMP is the main factor of fouling in micro and ultrafiltration membrane bioreactors, which usually does not occur in DMBRs due to the fact that the pores of the support are relatively large, and therefore more difficult to fully block⁶⁷⁻⁶⁸. However, in DMBRs, the blockage of support material pores can be caused by larger microbial flocs, constituted mainly of bacteria covered by EPS and SMP, which causes the microbial flocs to form a dense structure with few narrow channels for the permeate passage¹⁹⁻²⁰.

Biological flocs, in turn, consist of heterogeneous structures consisting mainly of microorganisms (bacteria, protozoa, fungi, among others), organic and inorganic particles, EPS, and SMP. It is noteworthy that the EPS and SMP function in floc formation is to increase the viscosity of the liquid, favoring extracellular enzymatic activity, and consequently, cell aggregation⁶⁹. The floc structure is strongly related to the operating conditions in DMBRs. Thus, if tangential velocity conditions are too high, they can promote a decrease in floc size, causing the formation of more compact cake layers on the surface of the support material. It is important, therefore, to optimize the tangential flow rate on the support material so that it favors the deposition of the cake on the support, but does not cause a high increase in the concentration of solutes and colloids of comparable or smaller size than the size of the support material pore. This is a

strategy capable of minimizing the effects of fouling and the deterioration of permeate quality²¹.

Sludge with stable structures forms and protects large flocs from being broken or broken down by external forces (e.g. shear force) or by turbulence. In addition, the structure-stable sludge flocs potentially reduce the release of EPS from the biological flocs, which alleviates blockage and promotes the formation of reversible *fouling* in DM. In other words, the good sludge aggregation capacity in dynamic membrane bioreactors presents lower *fouling* potential and higher permeability through the formed dynamic layer⁷⁰.

Other parameters that have a strong influence on the fouling in dynamic membrane bioreactors treating wastewater are permeate flow and TMP. Permeate flow, defined as the volume that permeates through the membrane per unit time and per unit area, is influenced by the affluent quality, stability of the operation, the properties (such as the type of material and the size of the pores), and cleaning frequency of the support material. It is also influenced by operating conditions such as TMP (pressure gradient that serves as the driving force for the transport through the membrane) and the mixing regime in the bioreactor. It should be mentioned that low TMP values allow much higher flows in dynamic membrane bioreactors, when compared to those observed in synthetic membrane bioreactors^{23, 28 71}.

Comparing the filtration between DMs and synthetic membranes, it has been found that higher TMP values are reached only when the dynamic membrane porosity is reduced by excessive accumulation of solids or by other phenomena related to sludge growth, such as SPM or EPS overproduction, which usually results in the occurrence of fouling^{23, 49, 72}.

Another important concept is that of critical flow, which consists of the highest permeate flow value below which fouling is not observed or its occurrence is negligible⁷³. Above the critical flow value, the formation of fouling is due to the greater tendency of particles to drag during filtration, causing them to deposit more quickly, not allowing hydrodynamic conditions to prevent blockage of the membrane pores.

It is important to note that the critical flow value can be determined in two distinct ways. One is keeping the flow constant and observing the TMP behavior. Thus, the critical flow will be established when the TMP begins to increase. Another way is to keep the TMP constant and measure the permeate flow. In this way, the critical flow will be set when the permeate flow begins to decrease⁷³.

From a practical point of view, for the treatment of wastewater in DMBRs, a high CRT associated with a low HRT promotes a higher sludge concentration and enables efficient treatment in a short time. However, such conditions can generate more intense fouling problems, which implies that the variables of HRT and CRT may be indirectly linked to the impacts caused by fouling in this type of system⁷⁴. However, the time required for the formation of a stable dynamic layer can take up to 50 days, depending on the permeate flow used^{18, 75}. Therefore, CRTs and HRTs must be

rigorously defined so that a stable and efficient DM is formed in a short time.

Studies have shown that very low HRTs (1h, for example) lead to an increase in TMP due to an increased concentration of the soluble microbial product, which leads to poor process performance and poor quality effluent. In addition, a very low HRT allows a greater accumulation of inorganic substances in the DM, which results in severe fouling caused by shorter reaction time³⁹.

The reduction of CRT however can cause an increase in filtration resistance, and consequently, in TMP. Huang *et al.* (2019)⁷⁶ found that, for a 5-day CRT, a thin and compact gel-like biofilm layer was formed (porosity = 27.5%), presenting a relatively high filtration resistance (approximately $4.9 \times 10^{11} \text{ m}^{-1}$), while a thick and porous biofilm layer (porosity > 60%), with lower filtration resistance ($< 2.5 \times 10^9 \text{ m}^{-1}$), was formed with CRTs greater than or equal to 20 days. Furthermore, it was observed that larger CRTs decrease the EPS production, but increase the protozoa population in the biofilm, which increases the biofilm porosity and reduces the biofouling on the material-support.

Although the increase in filtration resistance and the reduction in permeability in DMBRs are much lower compared to those observed in synthetic membrane bioreactors (approximately two orders of magnitude lower), the fouling control is even more necessary when a severe decline in flow or a rapid increase in TMP occurs. To minimize such deleterious effects during the wastewater bioprocess treatment it is necessary, first of all, to define suitable operating and hydrodynamic conditions, or even perform a pre-treatment of the suspension that will feed the membrane system. However, among all the factors that control the fouling, cleaning techniques are the only ones that have the function of recovering the permeate flow and can be carried out by chemical, physical or biological methods, whenever necessary²³.

Chemical cleaning is carried out using strong acids, caustic products, and/or oxidizing agents that recover the membrane's performance almost completely. However, the cleaning solution must have a pH value compatible with the pH range supported by the support material⁷⁷⁻⁷⁹.

Schneider & Tsutiya (2001)⁸⁰ emphasize the importance of the formulation of the cleaning solution, since fouling is rarely caused by a single type of material that settles on the membrane support. For the removal of inorganic deposits, acidic cleaning is usually carried out, while for the removal of organic deposits and biofilms, the solution formulation must be alkaline. It is, therefore, recommended to use different formulations alternately, as repeated applications of the same solution may result in the selection of a resistant biofilm. In general, supporting material manufacturers indicate commercial formulations compatible with their products.

In the case of the possibility of secondary contamination caused by chemicals, physical cleaning is resorted to, such as backwashing, a method widely used in membrane systems. Backwashing pushes the particles adhered to the pore structure into the liquid

and partially removes the cake formed on the surface of the mesh-support. The frequency and flow rate of the backwash are related to the operating conditions and characteristics of the effluent to be treated. Usually the method uses air, water or the permeate itself, which is pumped in the opposite direction of the permeation⁸¹.

It is worth mentioning that the backwashing frequency does not significantly affect the DMBR's performance. Therefore, an automated support material cleaning protocol can be easily applied at full scale for DMBRs^{16, 21}.

Cleaning can also be carried out by (1) using nitric oxide (in low concentrations) to induce the biofilm dispersion; (2) enzymatic hydrolysis of the extracellular polymers formed; and (3) rupture of the bond between the biofilm and the surface of the membrane, which can be promoted by the addition of bacteriophages. Although these techniques are still under development for bench scale use, there is the possibility that they can also be applied on a real scale⁵⁶.

Fouling can also be controlled depending on the module configuration where the membrane is located. In this sense, in external configuration systems, a high tangential flow velocity is maintained in order to limit the accumulation of organic and inorganic substances, responsible for the fouling. On the other hand, in the submerged configuration, the fouling control is carried out by spraying with gas, a backwash with tap water, or with the effluent itself⁸²⁻⁸³.

Mathematical modeling can define the mechanism of fouling formation during the filtration process through the DM. Ruth, B. F., in the mid-1930s, established the theory of filtration through the cake layer, providing the basis for the definition of modern filtration theory⁸⁴.

Despite the fact that in the processes of filtration through a DM the particles causing fouling play a decisive role in the development of the dynamic layer, few studies attempt to understand specifically how these phenomena occur by model-based analyses. In addition, the use of mathematical modeling to elucidate the mechanism of DM formation, together with the effect of changes in operating conditions (TMP and concentration of suspended solids) on the model's response and its interpretation, is still limited.

The laws of filtration blockage describe both the membrane pore blockage and the dynamic layer formation, in addition to allowing that the increase in filtration resistance be evaluated, as this is directly responsible for the flow decline over time and under constant pressure conditions, or for the increase in pressure over time under constant flow conditions. The fact is that fouling can be described by more than one model that explains the different mechanisms of membrane pore fouling (pore constriction, pore clogging, and the formation of the cake layer on the membrane surface), which can occur successively or simultaneously. In this sense, Iritani and Katagiri (2016)⁸⁴ defined the four models shown in Table 2 and Figure 3.

Where: P is the TMP (Pa); P_0 is the initial TMP (Pa); J_0 is the flow ($\text{L} \cdot \text{h}^{-1}$); K_i is the complete blockage constant; K_s is the standard blockage constant; K_i is the intermediate blockage constant; K_c is the filtration constant through the cake layer.

Table 2. Mathematical models to assess the evolution of fouling formation over time.

Filtration model	Model equation	Blockage constant
Complete blockage	$\frac{P}{P_0} = \frac{1}{1 - K_b \cdot t}$	$K_b \text{ (1.s}^{-1}\text{)}$
Standard blockage	$\frac{P}{P_0} = \left(1 - \frac{K_s \cdot J_0 \cdot t}{2}\right)^{-2}$	$K_s \text{ (1.m}^{-3}\text{)}$
Intermediate blockage	$\frac{P}{P_0} = \exp(K_i \cdot J_0 \cdot t)$	$K_i \text{ (1.m}^{-3}\text{)}$
Filtration through the cake layer	$\frac{P}{P_0} = 1 + K_c \cdot J_0 \cdot t$	$K_c \text{ (s.m}^{-6}\text{)}$

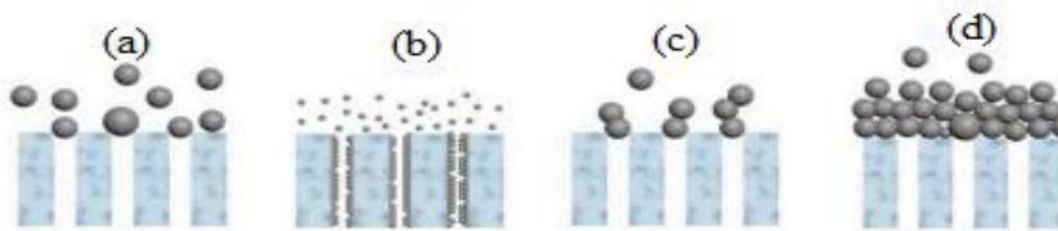


Figure 3. Schematic representation of fouling: (a) complete blockage; (b) standard blockage; (c) intermediate blockage, and (d) blockage through cake layer. Source: Wang et al. (2020)21.

In the complete blockage model it is assumed that each particle that reaches the support material surface obstructs a pore's passage, i.e., completely closes a pore on the surface of the support material. Furthermore, a particle never settles on another particle that has been previously deposited, i.e., there is no overlapping of the particles. The permeate flow through the non-blocked pores is not affected; thus, the reduction in the permeate flow is proportional to the reduction in the surface area of the support material corresponding to the non-blocked pores. It is worth stressing that this type of fouling occurs when the particle size is larger than the supporting material pore. Consequently, pore clogging occurs only on the surface of the supporting material and not inside its pores (Figure 3a).

The standard blockage model considers that the obstruction occurs inside the pores of the support material and, in this way, gradually reduces the pore volume until its complete blockage. The decrease in pore volume is proportional to the decrease in permeate volume (Figure 3b). It is also considered that the pore has a constant diameter and length along the thickness of the support material. Furthermore, as this type of obstruction is caused by particles smaller than the pores of the support material, the fouling becomes independent of the transverse flow velocity. Thus, the flow in the steady state is equal to zero, and the counter-diffusion of solutes from the surface of the supporting material into the solution does not occur.

The intermediate blockage model considers that the pore of the support material is not necessarily ob-

structed by a single particle, i.e., several particles are equally likely to deposit on each other causing the pore to block. Once more the blockage inside the pores is not considered. This type of fouling occurs when the particles are similar in size to the pore size of the support material (Figure 3c).

Finally, in the cake layer filtration model, the particles are larger than the pore size of the support material and the particle concentration is high. Therefore, there is no blockage of the pore. In this case, particles settle on the material-support surface, forming a first layer. Subsequently, a new layer is formed over the first one, and thus successively (Figure 3d), characterizing the adhesion process that occurs during the dynamic layer formation.

Performance of dynamic membrane anaerobic bioreactor in wastewater treatment

The first research on the application of AnDMBRs in wastewater treatment occurred in the mid 1990s⁶. However, since 2010, research has been developed with the objective of evaluating the performance of the bioreactor in anaerobic systems for the treatment of synthetic and domestic wastewater, emphasizing the efficiencies of removal of organic matter and nutrients, the production of biogas, and also carrying out an analysis of the various factors that influence the formation of the DM and the efficiency of the bioprocess.

Table 3 shows some studies involving the use of AnDMBR in bench and pilot scales. In general, the

results show that the efficiency of the organic matter removal is independent from the system configuration (external module or submerged), but is related to the applied organic load, the type of substrate, and the operating temperature.

For applied organic loads ranging from 0.66 to 5.5 kgCOD.m⁻³.d⁻¹, the removal efficiency of organic matter in terms of total COD (COD_t) reached values between 63 and 90%, for domestic wastewater, and between 75 and 99%, for synthetic wastewater. For removal efficiency in terms of soluble COD (COD_s), there were variations between 39 and 78% and between 65 and 98.5% for domestic and synthetic wastewater, respectively. Thus, comparing these values to those presented by other anaerobic systems such as a UASB reactor treating domestic sewage, it can be considered that the AnDMBR presented satisfactory results. However, for applied organic loads equal to or greater than 6.0 kgCOD.m⁻³.d⁻¹, the total and soluble COD removal efficiencies of domestic wastewater were considerably reduced (60 and 15.2%, respectively), to the point where the system was considered to be of low efficiency³⁹. These results can be explained by the fact that systems with high organic loads result in high concentrations of

SMP in the liquid phase, and inorganic and protein-like substances in the dynamic layer, resulting in a denser and more compact membrane (severe fouling). In these cases, a significant reduction in the degradation and filtration of pollutants in the environment can occur, leading to the loss of microbial metabolites and increase in the concentration of organic matter in the effluent.

Regarding the removal of nutrients such as phosphorus and nitrogen, the low values presented (Table 3) may have been due to the fact that nitrogen and phosphorus are soluble in water, and small soluble molecules are weakly adsorbed by the sludge particles, thus resulting in a low adhesion^{75, 85}. However, Ma *et al.* (2013)¹³ verified nitrogen and phosphorus removal efficiencies of 30 and 62%, respectively. The authors used additional equipment for nutrient recovery in order to optimize the efficiency of the bioprocess.

Where: EM: external module; SM: submerged module; DWW: domestic wastewater; SWW: synthetic wastewater; COD_t: total chemical oxygen demand; COD_s: soluble chemical oxygen demand; P_t: total phosphorus; N_t: total nitrogen; NA: not available.

Analyzing these results, Hu *et al.* (2020)⁵⁰ associated the AnDMBR with good performance, especially in the

Table 3. Performance of anaerobic dynamic membrane bioreactors in wastewater treatment.

Setting	Substrate	System volume (L)	Temperature (°C)	Organic load (kgCOD.m ⁻³ .d ⁻¹)	Permeate flow (L.m ⁻² .h ⁻¹)	Organic matter removal efficiency (%)	Nutrient removal efficiency (%)	Biogas production [mLCH ₄ .(gCOD removed) ⁻¹]	Reference
SM	SWW	11	NA	NA	137.1	89.5 - COD _t	21.5 - NH ₃	NA	89
SM	DWW	45	10 to 30	NA	65	63.4 - COD _t	NA	NA	90
SM	DWW	2.0	7.5 to 30	NA	60	79.4 - COD _t	60 - P _t	NA	12
SM	DWW	2.0	7.5 to 30	NA	60	81.6 - COD _t	30.3 - NH ₃ 62 - P _t	NA	13
EM	SWW	0.898	35 ± 1	0.73 to 5.0	1.0 to 7.2	75±8 - COD _t	NA	NA	18
EM	SWW	0.898	20 to 24	0.16 to 3.3	1.4 to 28.0	80 - COD _t 90 - COD _s	NA	85	42
SM	SWW	6.8	35.7 ± 1	2.0	2.6	99 - COD _t 63 - COD _s	20 - N _t 13 - P _t	310	75
SM	SWW	7.4	35.7 ± 0.1	2.0	2.2	99 - COD _t 65 - COD _s	18 - N _t 16 - P _t	280 to 310	14
SM	SWW	7.4	35.5 ± 0.2	2.0	2.2	99 - COD _t 65 - COD _s	NA	NA	15
SM	DWW	3.5	20	0.88 to 3.01	22.5	70-90 - COD _t 50-70 - COD _s	NA	NA	91
SM	SWW	14.0	25	NA	50 to 150	80 - COD _t	10 - N _t 30 - P _t	NA	85
SM	SWW	4.52	35	2.34 to 5.2	36 to 51.5	75 to 89 - COD _t	NA	260	92
EM	SWW	10.0	37 ± 1	1 to 5.5	14 to 28	93 - COD _t 98.5 - COD _s	NA	NA	93
EM	DWW	62.8	19 to 30	1.43	780	86 to 88 - COD _t 74 to 78 - COD _s	20.3 - N _t 16.4 - P _t	78 to 91	27
EM	DWW	62,8	24 to 32	1.87 to 2.90	1,170 to 1,755	79 to 86 - COD _t 60 to 75 - COD _s	19 to 21 - N _t 15 to 18 - P _t	104.4 to 111	28
SM	DWW	3.6	20 to 25	0.82 to 3.01	22.5 to 90	71 to 75 - COD _t 39 - COD _s	NA	80 to 120	39
SM	DWW	3.6	20 to 25	6.8	180	60 - COD _t 15 - COD _s	NA	50	39
SM	SWW	6.0	33 ± 2	NA	5 to 15	95 - COD _t	NA	NA	62

treatment of wastewater with low solids concentration, at room temperature. Under these conditions, once the phosphorus is in the particulate form, it can be easily assimilated by the sludge and/or accumulated in the stable dynamic layer. Nitrogen removal efficiencies are relatively lower because ammonia nitrogen, the main component of total nitrogen present in wastewater, cannot be oxidized by anaerobic microorganisms, therefore, it leaves the reactor dissolved in the liquid effluent.

Although it is not possible to achieve high efficiencies of nutrient removal in anaerobic systems, in AnDMBRs, removal can occur through chemical and physical mechanisms. The membrane cake layer can present high concentrations of phosphorus and nitrogen, making it feasible to recover these elements in the form of struvite (natural fertilizer), which occurs in the presence of appropriate concentrations of ions such as calcium, magnesium, and potassium in alkaline conditions. Precipitation occurs when the combined concentrations of Mg^{2+} , NH_4^+ and PO_4^{3-} exceed the solubility of struvite, which, in turn, is controlled by the pH of the medium. As the pH increases, the solubility decreases, which makes pH one of the most important factors for precipitation of this material^{24, 86-88}.

The orthophosphate concentration in the medium is another important factor. According to Britton *et al.* (2005)⁹⁴, only for an orthophosphate concentration equal to or greater than 40 mg.L^{-1} is it possible to maintain high efficiency in the forming of struvite. However, the product of the molar concentrations of struvite-forming constituents present in the liquid phase of the anaerobic digester is not sufficient to induce the formation of a substantial amount of the product, limiting its efficiency to phosphorus recovery. Therefore, in the AnDMBR system the removal happens as a result of the growth of the dynamic membrane and the reduction of the pores, causing the particulate material to be retained in the membrane itself. This fact makes dynamic membrane bioreactors such an interesting alternative, since, in addition to efficiently performing secondary treatment, they still allow the recovery of nutrients contained in the cake, thus avoiding a possible tertiary treatment.

Regarding biogas production, it was observed that when the organic load was between 0.66 and $5.5 \text{ kgCOD.m}^{-3}.\text{d}^{-1}$ in synthetic wastewater (except ALIBARDI *et al.*, 2016)⁴², methane yields were high, ranging between 260 and $310 \text{ mLCH}_4.(\text{gCODremoved})^{-1}$, which amounted to, respectively, 66 and 86% of the theoretical value ($395 \text{ mLCH}_4.(\text{gCODremoved})^{-1}$, at 35°C). In domestic wastewater, methane yields were relatively lower and ranged between 80 and $120 \text{ mLCH}_4.(\text{gCODremoved})^{-1}$, reaching, respectively, 22.9 to 34.3% of the theoretical value equivalent to $350 \text{ mLCH}_4.(\text{gCODremoved})^{-1}$, which was measured under normal temperature and pressure conditions (NTP). For organic loads equal to or greater than $6.0 \text{ kgCOD.m}^{-3}.\text{d}^{-1}$, it was found that biogas production was even lower ($50 \text{ mLCH}_4.(\text{gCODremoved})^{-1}$, i.e., 14.3% of the theoretical value measured at NTP, since the degradation efficiency of organic matter was also lower.

Hu *et al.* (2018)⁹⁵ state that the lower yields of biogas for domestic sewage, when compared to those obtained for synthetic and industrial wastewater, can be explained by the fact that in domestic wastewater particulate substances represent more than 50% of total organic matter. In contrast, synthetic wastewater contains dissolved and easily biodegradable organic substances (such as glucose and acetate). These substances can be effectively degraded by anaerobic biomass under well-controlled operating conditions, thus contributing to high removal of organic matter and consequently higher biogas production. Moreover, domestic wastewater may contain some refractory organic compounds (even inhibitory and toxic substances) or of moderate biodegradability, which reduce COD removal rates and may inhibit biogas production, further compromising the operating stability of AnDMBRs^{39, 50}.

Alibardi *et al.* (2016)⁴² found a relatively low methane yield value— $85 \text{ mLCH}_4.(\text{gCODremoved})^{-1}$ —even when treating synthetic waste water. The authors found that, due to the AnDMBR configuration, part of the produced biogas was dissolved in the liquid medium and, thus, left the bioreactor with the effluent.

Similar behavior was verified by Chimuca *et al.* (2020)²⁷ treating domestic wastewater. The authors attributed the low yield to the dissolution of biogas in the liquid medium, due to the supersaturation conditions of the AnDMBR system as the DM was reaching irreversible fouling. The authors also found that the occurrence of system supersaturation conditions is considerable when backwashing is not applied.

When analyzing biogas production in terms of temperature (Table 3), it was found that the high methane yields (260 and $310 \text{ mLCH}_4.(\text{gCODremoved})^{-1}$) were obtained at higher and constant temperatures (around 35°C). In contrast, all low methane yields were obtained at relatively low and variable temperatures (between 19°C and 30°C) during the operation of the respective systems (including ALIBARDI *et al.*, 2016)⁴². This proves that temperature influences the gas-liquid mass transfer significantly, since the solubility of biogas in the liquid is inversely proportional to the operating temperature of the bioreactor. The gas-liquid mass transfer coefficient changes significantly according to reactor configuration and operating conditions, and can lead to methane concentrations in the liquid phase up to 12 times higher than thermodynamic equilibrium values⁹⁶.

The diversity of possibilities that the application of DMBRs in anaerobic treatment of wastewater promotes is noticeable, since, in addition to the stabilization of the affluent organic matter and not needing a secondary decanter, it also allows the sustainable and economically profitable use of the generated by-products.

Prospects for the use of AnDMBRs for wastewater treatment: future challenges

Studies carried out over the years show that the AnDMBR system has shown excellent results in relation to the removal of organic matter (expressed in COD terms) and total suspended solids, providing low turbidity to the permeate (effluent) equivalent to

that which is characteristic of ultra and microfiltration membrane bioreactors. However, nutrient removal (N and P) has been relatively low in some studies^{15, 18, 24, 42, 75, 85, 91, 95}, in the same way that biogas production and average methane yield are normally lower than their theoretical values, due to the considerable output of dissolved biogas in the system effluent and the operating conditions that are adopted^{15, 42, 95}. Therefore, there is still a need to improve the configuration and operating conditions of the system to at least optimize biogas production and methane yield, so as to make AnDMBRs more sustainable.

So far, the numerous pieces of research developed using AnDMBRs have been focused on the operational parameters (TMP, permeate flow, critical flow, HRT, and CRT), the structure of the dynamic layer (physicochemical and biological properties of the cake layer), on the morphological properties of the sludge (sedimentation, dehydration, flocculability, hydrophobicity, EPS, and SPM), the quality of the permeate produced (COD, turbidity, solids, N, and P), the modeling of DM formation, and the mechanisms responsible for fouling, such as the XDLVO theory^{12-15, 18-20, 24, 26, 42, 45, 48-49, 70, 75, 85, 91, 95, 97-98}. However, there are still challenges to be overcome and answers to be achieved, such as obtaining a more complete mathematical model, which can aggregate (concomitantly or not) operational and physico-chemical parameters, thermodynamic and hydrodynamic forces, and biological properties of the sludge, to accurately determine the critical flow and backwash frequencies. In this way, AnDMBR operation would have greater control in the medium and long term, and problems such as fouling could be avoided. Moreover, there are very few studies that evaluate biological parameters such as helminth eggs^{27, 28}.

Most of the research has been carried out in bench scale and with temperature control. Thus, it is still a great challenge to develop, build, and monitor full-scale stations operated at room temperature, with the objective of applying the AnDMBR in individual systems and in collective domestic and industrial wastewater treatment systems. In addition, much still needs to be clarified in terms of removal and reuse of resources. Therefore, the development of basic research and real-scale projects need to be continuously carried out so that increasingly optimized systems are obtained.

CONCLUSION

AnDMBRs have provided results that are comparable to those obtained in conventional MF and UF membrane systems, presenting effluents with low turbidity and with high removal efficiencies of organic matter, suspended solids, and helminth eggs. Furthermore, the DM is formed on a low-cost support material, and filtration can occur by gravity, which further reduces treatment costs.

Regarding fouling, despite causing an increase in TMP and a reduction in permeate flow, it is an easy problem to solve, through backwashing using the treated effluent itself, for example. The scale-free sup-

port material allows the DM to be re-formed and the effectiveness of the process recovered.

The formation of the DM, crucial for the good performance of the process, can be explained through several theories, which can be based on hydrodynamic and thermodynamic forces, as well as the speeds with which the sludge flocs reach the support material. The process, which usually takes place in two stages (initial and maturation), should allow the formation of a robust, dense, and stable membrane with adequate permeability and filterability characteristics.

However, despite the numerous positive points, much still needs to be investigated so that existing obstacles and doubts regarding the use of AnDMBRs in wastewater treatment can be eliminated. The necessary advances should seek measures that further reduce process costs, making this type of bioreactor become increasingly competitive.

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ORCID

Jacob Fortuna José Chimuca: <https://orcid.org/0000-0001-7143-7443>

Catarina Simone Andrade do Canto: <https://orcid.org/0000-0002-6078-8560>

José Tavares de Sousa (*in memoriam*): <http://orcid.org/0000-0002-1056-1771>

Valderi Duarte Leite: <http://orcid.org/0000-0001-5861-7407>

Wilton Silva Lopes: <http://orcid.org/0000-0002-0151-7664>

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