The Membrane-Biofilm Reactor (MBfR) as a counter-diffusional biofilm process

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Abstract

The membrane-biofilm reactor (MBfR), sometimes known as the membrane-aerated biofilm reactor (MABR), is an emerging treatment technology based on gas-transferring membranes. The membranes typically supply a gaseous electron donor or acceptor substrate, such as oxygen, hydrogen, and methane. The substrate diffuses through the membrane to a biofilm naturally forming on the membrane outer surface. The complementary substrate (electron donor or acceptor) typically diffuses from the bulk liquid into the biofilm, making MBfR counter diffusional. This paper reviews the unique behavior of counter-diffusional biofilms and highlights recent research on the MBfR. Key advances include insights into the microbial community structure of MBfRs, applying the MBfR to novel contaminants, providing a better understanding of biofilm morphology and its effects on MBfR behavior, and the development of methane-based MBfR applications. These advances are likely to further the development of the MBfR for environmental applications, such as energy-efficient wastewater treatment and advanced water treatment.

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Introduction

Biofilms processes are of increasing interest in environmental biotechnology, mainly due to their ability to accumulate high biomass densities and retain biomass within the reactor [1]. Most existing biofilm processes are based on inert attachment surfaces, such as stone, pumice stone, dense plastic, or plastic foams. These can be called “co-diffusional” biofilms, as both electron donor and acceptor substrates are supplied from the bulk liquid (Figure 1a). However, biofilms can also develop on “reactive” surfaces that release an electron donor or acceptor substrate into the biofilm (Figure 1b). Because the electron donor and acceptor enter the biofilm from opposite sides, these are known as counter-diffusional biofilms.

Fig. 1. a) Substrate gradients in a conventional, co-diffusional biofilm. b) Substrate gradients in a counter-diffusional MBfR biofilm. The bold line is the substrate released by the reactive surface (electron acceptor, S_a , in this case), while the dashed line indicates the substrate from the bulk liquid (electron donor, S_d , in this case).

Counter-diffusional biofilms can be found in natural and engineered systems. In natural systems, they can be found on gas-liquid interfaces, such as air bubbles or on plant roots, where the roots may provide organic exudates or oxygen to biofilms growing on their surface. Counter-diffusional biofilms also can develop on inorganic solids, such as elemental sulfur and pyrite, or organic solids, such as chitin and biodegradable polymers.[2[•]-4] Even microbial electrochemical systems, such as microbial fuel cells, can behave as counter-diffusional biofilm processes.[2[•]]

An important counter-diffusional biofilm process in environmental biotechnology is the membrane-biofilm reactor (MBfR). The MBfR is based on gas-permeable membranes that deliver a gaseous substrate to biofilms naturally forming on the membrane outer surface.[2[•],5^{••}] When supplying air or oxygen, they often are called Membrane Aerated Biofilm Reactors

(MABRs).[5•,6] This technology has been studied since the 1970s, but in recent years has received intense interest. Also, several MBfR and MABR commercial technologies have been launched in recent years, spurring further interest.

This paper reviews the basic principles of counter-diffusional biofilm behavior and the most recent developments on the MABR and MBfR processes.

Special behavior of counter-diffusional biofilms

With co-diffusional biofilms, the most metabolically active region of the biofilm is typically the exterior, where the electron donor and acceptor substrates are at their highest concentrations. In counter-diffusional biofilms, in contrast, one of the two substrates diffuses into the biofilm from the base, while the complementary substrate diffuses from the bulk liquid.[5•] Thus, the most active zone is typically located in the interior of the biofilm. This counter diffusion of donor and acceptor leads to unique behavior, including three key differences: development of unique microbial community structures, greater sensitivity to biofilm accumulation, and reduced susceptibility to liquid diffusion layer (LDL) resistance. Each item is discussed below.

- **Development of unique microbial community structures.** The counter diffusion of substrates can lead to unique microbial community structures in MBfRs. For example, in most biofilms for wastewater treatment, aerobic nitrifying microorganisms develop in the deeper biofilm regions, where the organic carbon concentration is lowest. While this location has minimized competition from heterotrophic microorganisms, it also is where the oxygen concentration is lowest, leading to low nitrification activity.[7] In an MABR system, oxygen is supplied from the base of the biofilm, so the nitrifiers experience low organic carbon and high oxygen concentrations, promoting high nitrification activity. Additionally, if the bulk liquid is left anoxic, then both nitrification and denitrification can take place within the same biofilm (Figure 2).[8] This unique stratification may favor suppression of nitrite-oxidizing organisms (NOB).[7]
- **Greater sensitivity to biofilm accumulation.** In conventional biofilms, the initial contaminant transformation fluxes are low, due to low biofilm thicknesses. The fluxes then increase as the biofilm thickness increases, until the biofilm growth is balanced by decay and detachment. In counter-diffusional biofilms, fluxes increase up to a point, but then decrease as the thickness increases further. This is because of donor and acceptor counter diffusion. The biofilm interior has low activity due to the limitation of one substrate, while the exterior has low rates due to the limitation of the other. Thus, careful management of biofilm accumulation is needed to maintain high fluxes.[5•]
- **Lower susceptibility to liquid diffusion layer resistance.** In a conventional biofilm, the LDL limits substrate fluxes into the biofilm. As the biofilm thickness and flux increase, the LDL provides mass transfer resistance and limits further biofilm growth. Higher bulk substrate concentrations are needed to overcome the LDL resistance. In contrast, in a counter-diffusional biofilm, the LDL provides a barrier to loss of the internal substrate to

the bulk liquid. Thus, as long as the substrate from the bulk is present at non-rate-limiting concentrations, the LDL will not limit, and may actually enhance, microbial activity.[5••]

Fig. 2. Substrate gradients in an MABR for Chemical oxygen demand (COD) oxidation, nitrification, and denitrification. Note that nitrifying bacteria are located in the interior, where they have access to high oxygen concentrations, and heterotrophic denitrifiers can grow using nitrate formed by nitrifiers and COD diffusing from the exterior of the biofilm. Thus, concurrent nitrification and denitrification is easily achieved.

In addition to the above, the mode of gas supply and biofilm development in MBfRs can lead to special behavior.[5••] For example, when gas is supplied via hollow-fiber membranes, other dissolved gases in the bulk liquid, or gases formed in the biofilm, can diffuse back into the membrane, diluting the supply gas. When operating an MBfR with sealed membranes, these gases concentrate at the distal end of the membrane, decreasing its effectiveness and leading to thinner biofilms. Also, several hollow-fiber membranes can clump together, effectively forming one large biofilm with individual membranes providing point gas sources in the biofilm interior. Finally, substrate counter diffusion can impact biofilm properties such as diffusivity and mechanical resistance. All these effects are distinct from conventional biofilm processes and can be important in understanding and predicting process behavior. Further research should assess these effects.

The following sections address the most recent research on the MBfR and MABR.

Biofilm morphology and MBfR performance

Biofilms can assume uneven or rough morphologies, which result from an interplay between cell growth rates, shear conditions, microbial cell types, and other factors. The roughness of counter-diffusional biofilms can impact their behavior. In particular, roughness increases the effective LDL thickness, which limits fluxes in conventional biofilms.[9] As explained above, in counter-diffusional biofilms, the LDL does not limit the substrate supplied from within the biofilm, so

roughness is not necessarily detrimental. Recent work has demonstrated this experimentally and with modeling.[10]

The fluid flow regime is important for all biofilm processes. Thus, computational fluid dynamics can be an important tool for MBfR studies, either to determine the flow regime around membranes, or to couple with a model of biofilm growth and deformation.[11,12] This can be especially important when biofilm-covered membranes contact each other, as they may bundle and cause dead zones without flow. As mentioned previously, the bundle may behave like a single biofilm with gaseous substrates sources in the interior.[13]

It may be important to consider the coupled effects of biofilm development and fluid flow and shear. Biofilm development can alter the fluid flow patterns, while flow-induced shear can determine the biofilm morphology.[12,14] This is important for mass transfer in and out of the biofilm, but morphology and detachment can also impact the microbial community structure.[15]

There is an increasing interest in biofilm mechanical properties, and new models are being developed to incorporate these properties to predict deformation and detachment.[16-18] Some recent studies have determined mechanical properties for MABR biofilms. These can differ significantly from conventional biofilms, but these have not yet been included in mechanical models.[12,19]

MABR

Much research has focused on MABRs for removal of COD and nitrogen.[5,6] Prior research has shown that concurrent nitrification and denitrification can be achieved in MABRs, that nitrification is less susceptible to BOD inhibition, and that short cut nitrogen removal, via nitrite, can be achieved.[8] Membrane fouling from excess biomass accumulation is a concern.[5] More recent research has confirmed these findings, and explored MAB performance for other types of matrices, such as landfill leachate, oil field wastewater, and pharmaceutical wastewater.[13,20-22, 23,24] Some research also confirmed the ability of the MABR to achieve biological phosphorus removal, when operated in sequencing batch mode with a biofilm removal stage. [25] Several types of new process configurations have been developed, for example combining MABRs with ozone, powdered activated carbon, and coagulation flocculation, to enhance removal of recalcitrant organics.[20,23,26] New membrane materials were also studied to improve cell adhesion and gas transfer.[24] Also, some studies addressed the microbial community structure of MABRs for removal of COD, nitrification and denitrification, as well as the need to model nitrite as an intermediate.[27,28]

Gas transfer is key for MABRs. High oxygen transfer efficiencies (OTEs) can be obtained with closed end operation. However, closed end operation is subject to gas back diffusion, which results in lower oxygen transfer rates (OTRs).[5] Gas back-diffusion has been studied previously for clean, closed end membranes, but little has been done for membranes with biofilms. Recent research has addressed the effects of biofilm on oxygen transfer in an MABR,

and the interplay of OTE and OTR were addressed in pilot scale research.[13[•],29,30] Further research is needed to explore the effect of biofilm on back-diffusion in closed end membranes, as well as the effects of partially opened membranes or transient effects of opening and closing membranes. Optimal operational strategies may be developed from these approaches.

A major benefit of the MABR is savings in energy costs. It is difficult to quantify the savings without full scale data, but pilot scale data and modeling can be used as a first approximation. Studies suggest that the MABR can be up to 86% more energy efficient than conventional activated sludge.[13[•],31[•]] The major factors affecting cost savings are the cost of electricity, the membrane cost, membrane durability, and MABR removal fluxes. Future research should explore these items in more detail.

Some recent research has addressed the use of MABRs for autotrophic nitrogen removal via Anammox microorganisms. A challenges for Anammox processes is achieving nitrification, avoiding the proliferation of NOB. This is especially challenging for mainstream wastewater treatment systems. Research has shown nitrification can be obtained by cycling the oxygen supply, and that this operation also results in reduced emissions of nitric and nitrous oxide.[32-34[•]] However, others have shown it is possible to achieve nitrification in an MABR with continuous aeration.[35]

An exciting development is that several companies have developed commercial MABR applications, including Oxymem, GE, and Emefcy. This is likely to further catalyze research interest on the MABR.

H₂-based MBfR

One of the major applications of the MBfR is to supply H₂ as an electron donor for denitrification, and to a lesser extent to achieve the reduction of perchlorate and other oxidized contaminants.[5^{••}] When removing perchlorate or other trace contaminants concurrently with nitrate, a concern is that their reduction may be slowed or inhibited by nitrate.[36] Yet if no nitrate is present, there may not be enough primary electron acceptor to sustain the competent population. Also, if nitrate is driven to very low levels, sulfate-reducing bacteria and methanogens may proliferate, leading to undesirable byproducts[15^{••}]. A two-stage MBfR was studied as a means to reduce the nitrate concentration so that appropriate levels were obtained in the second stage.[37-41[•]] Sulfate-reducing bacteria were found to persist along with the denitrifying bacteria, even though sulfate reduction was minimal due to H₂ limitation. Modeling was also used to show that appropriate management of biofilm thickness and detachment were needed.[12,15^{••}] Multidimensional modeling revealed unexpected interactions between sulfate reducing bacteria and denitrifying bacteria, which in turn were affected by the geometry of the membrane assembly.[15^{••}] The curvature of the membranes provided protected niches with high H₂ concentrations and little detachment, allowing these undesirable species to proliferate

Several contaminants were shown to be effectively removed by the H₂-MBfR, including parachlorobenzene and tetrachloroethene, and other studies addressed the microbial communities carrying out selenate reduction.[42-44] Novel membrane configurations were also explored, either supplying CO₂ in separate membranes or placing H₂-supplying membranes separately

from a bioreactor to allow the membranes to be chemically cleaned without disrupting the biological process.[45,46]

A commercial version of the H₂-MBfR was launched by APTwater, although due to mergers and changes in management, it is not clear if they will continue to produce their MBfR technology.

CH₄-based MBfR

A novel MBfR application is to supply methane (CH₄) to drive reduction processes, in a similar fashion to the H₂-based MBfR. This process relies on recently discovered methane-oxidizing microorganisms known as denitrifying anaerobic methane oxidation (DAMO) microorganisms. The process is sometimes called “nitrate/nitrite-dependent anaerobic methane oxidation” (n-DAMO). Researchers have shown n-DAMO is achievable in a MBfR supplied with methane.[47-49] Methane has also been shown to serve as a donor for perchlorate reduction. Although the methane-based processes seem to be much slower than those based on H₂, this is an exciting new direction for MBfRs and is likely to receive much interest over the next few years.

Conclusions

The MBfR and MABR are technologies that are approaching maturity and are now available at the commercial scale. A surge in research, both fundamental and applied, has been seen globally in recent years. This is leading to a better understanding of the special behavior of the MBfR, as well as new applications. Future research should address the improved methods of biofilm management, possibly addressing the biofilm mechanical properties, the effect of back-diffusion on closed end membranes, membrane costs, membrane durability, MABR removal fluxes, and new applications, such as methane-based processes.

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Graphical Abstract