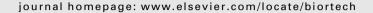
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The membrane biofilm reactor (MBfR) for water and wastewater treatment: Principles, applications, and recent developments

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ABSTRACT

The membrane biofilm reactor (MBfR), an emerging technology for water and wastewater treatment, is based on pressurized membranes that supply a gaseous substrate to a biofilm formed on the membrane's exterior. MBfR biofilms behave differently from conventional biofilms due to the counter-diffusion of substrates. MBfRs are uniquely suited for numerous treatment applications, including the removal of carbon and nitrogen when oxygen is supplyied, and reduction of oxidized contaminants when hydrogen is supplied. Major benefits include high gas utilization efficiency, low energy consumption, and small reactor footprints. The first commercial MBfR was recently released, and its success may lead to the scale-up of other applications. MBfR development still faces challenges, including biofilm management, the design of scalable reactor configurations, and the identification of cost-effective membranes. If future research and development continue to address these issues, the MBfR may play a key role in the next generation of sustainable treatment systems.

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1. Introduction

Certain dissolved gases serve as electron donors or acceptors for microbial treatment processes. For example, dissolved oxygen is used by aerobic microorganisms to oxidize chemical oxygen demand (COD) and ammonium in the activated sludge process; hydrogen drives the microbial reduction of oxidized contaminants and halogenated organics; and methane supports the cometabolic oxidation of a wide range of organic compounds. Despite the versatility and efficiency of many gaseous substrates, low aqueous solubility limits their practical use. In the activated sludge process, the solubility bottleneck is resolved by continuously bubbling air to avoid oxygen depletion. Unfortunately, gas bubbling requires large amounts of energy and can strip volatile organic compounds (VOCs) and greenhouse gases (e.g., methane and nitrous oxide) from the water. Furthermore, this approach is infeasible for more expensive or combustible gases, such as hydrogen or methane. A more effective means of supplying dissolved gases to microorganisms is through membranes. The membrane's lumen can be pressurized with a gas, which diffuses through the membrane wall to a biofilm attached at the membrane's outer surface. When used to deliver air or oxygen, the process is often called a membraneaerated bioreactor (MABR). However, more general terms are membrane-supported biofilm reactor (MSBR) (Stricker et al., 2011) and membrane biofilm reactor (MBfR) (Rittmann, 2006). For consistency, the term "MBfR" will be used in this paper.

Past reviews have addressed MBfRs in regard to specific applications, such as air or oxygen-based MBfRs (Syron and Casey, 2008) or hydrogen-based MBfRs (Celmer-Repin et al., 2010; Hwang et al., 2009b; Rittmann, 2006). This paper provides a comprehensive review of MBfRs for water and wastewater treatment by presenting key aspects of MBfR behavior, summarizing applications, discussing design considerations, and identifying future research needs and directions for the technology.

2. Fundamental behavior of MBfRs

MBfR biofilms behave differently than conventional biofilms, and this should be considered when interpreting past studies or developing new applications. The main differences are described below.

2.1. Gas-liquid exchange

2.1.1. Gas transfer from membrane to biofilm

The driving force behind gas transfer in an MBfR system is the concentration gradient across the membrane wall. The flux through an MBfR membrane can be described by:

$$J = HS'_{MG}K_M\left(\frac{p}{H} - C_{MB}\right) \tag{1}$$

where,
$$S'_{MG}K_M = \frac{P}{t_M}$$
 (2)

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J is the gas flux (M L^{-2} T^{-1}), S'_{MG} is the gas/membrane partition coefficient (L^{-2} T^2), K_M is the mass transfer coefficient of the membrane (L T^{-1}), p is the partial pressure in the gas phase (M $L^{-1}T^{-2}$), H represents Henry's law constant ($L^2 T^{-2}$), and C_{MB} is the dissolved gas concentration at the membrane-biofilm interface ($M L^{-3}$). P is the permeability of the membrane (T^{-1}) , and t_M is the membrane thickness (L), though for hollow-fiber membranes, the equivalent thickness should be used (Cote et al., 1989). Resistance to mass transfer is introduced by the gas film in the lumen of the membrane and the membrane wall itself, but gas film resistance is usually considered negligible. Cote et al. (1989) may be referenced for a more in-depth discussion of gas transfer in clean membranes with no biofilm. When biofilm is present, C_{MB} depends on both the diffusional resistance imposed by the biomass and the activity of the gas-consuming microbial reaction. Compared to clean membranes, the concentration at the membrane surface is typically lower in the presence of a biofilm, resulting in a greater gas transfer rate (Casev et al., 1999; Gilmore et al., 2009).

For the MBfR, the liquid diffusion layer (LDL) resides on the outer edge of the biofilm, introducing resistance to gas transfer from the biofilm to the bulk. Mass transfer to the bulk liquid is represented by:

$$J = K_{LDL}(C_{LB} - C_L) \tag{3}$$

where,
$$K_{LDL} = \frac{D}{t_{LDL}}$$
 (4)

 $K_{\rm LDL}$ is the mass transfer coefficient imposed by the LDL (L T $^{-1}$), C_{LB} represents the dissolved gas concentration at the biofilm-liquid interface (M L $^{-3}$), and C_L is the bulk liquid concentration (M L $^{-3}$). D is the diffusivity of the dissolved gas in water (L 2 T $^{-1}$) and $t_{\rm LDL}$ is the thickness of the LDL (L).

Several important features distinguish gas transfer in the MBfR from that in conventional biofilms:

- In conventional biofilms, the dissolved gas must traverse the LDL in order to penetrate the biofilm. In an MBfR, no LDL exists between the gas-supplying membrane and biofilm.
- The LDL, located at the outer edge of the biofilm, helps to resist the loss of gas to the bulk liquid, thus contributing to higher gas utilization efficiencies.
- Gas transfer flux can be controlled through adjustment of the gas supply pressure.
- The gas flux is self-regulating, in that biochemical demand for dissolved gases increases the concentration gradient in the biofilm, which thereby increases the driving force for gas supply.

2.1.2. Gas back-diffusion

Dissolved gases present in the biofilm, but not in the supply gas, are able to diffuse from the biofilm back into the membrane. For example, in applications where hydrogen gas is supplied, dissolved nitrogen gas can back-diffuse from the bulk liquid to the membrane lumen. The effect is increased for denitrification applications, where nitrogen forms within the biofilm. When hollowfiber membranes are used in dead-end mode, where the distal end of the membrane is sealed, back-diffusion can significantly dilute the supply gas and consequently decrease the effectiveness of the MBfR. This topic is further discussed in Section 4.2.1.

2.2. Microbial metabolism and ecology

MBfR biofilms behave differently than conventional biofilms due to the counter-diffusional delivery of substrates. For conventional, co-diffusional biofilms, both the electron donor and acceptor concentrations are greatest at the outer edge of the biofilm

near the bulk liquid. Here, the relative microbial degradation activity is highest (Fig. 1a). Relative activity is defined as the product of the Monod terms for the electron donor and acceptor:

relative activity =
$$\frac{[S_d]}{K_{Sd} + [S_d]} \cdot \frac{[S_a]}{K_{Sa} + [S_a]}$$
 (5)

where S_d and S_a are the concentrations of electron donor and acceptor (M L⁻³), respectively, and K_{Sd} and K_{Sa} are the half saturation constants (M L⁻³). For counter-diffusive biofilms, one substrate (i.e., either donor or acceptor) enters the biofilm from the bulk liquid, while the other is supplied from the attachment surface (i.e., the membrane) (Fig. 1b). Once either the donor or acceptor becomes depleted, the activity decreases to zero. Unlike conventional biofilms, where the highest activity typically occurs at the outer biofilm edge, the highest activity in an MBfR occurs at any location within the biofilm depending on the donor and acceptor concentrations. For example, when the membrane side of the biofilm is limited by the donor and the bulk liquid side by the acceptor, the highest activity occurs in the biofilm's middle section (Essila et al., 2000; Rishell et al., 2004). Consequently, knowledge of the dissolved gas concentration within the biofilm is required for understanding its behavior.

The unique behaviors of counter-diffusional biofilms benefit MBfR performance. For instance, MBfR biofilms can exhibit greater resistance to toxic shocks or inhibitory compounds since they maintain high activity in the inner portions of the biofilm (Misiak et al., 2011; Syron et al., 2009). Counter-diffusional biofilms also support unique microbial niches. Fig. 1c provides an example, an MBfR biofilm conducting concurrent removal of COD and total nitrogen (i.e., nitrification and denitrification). Like in conventional biofilms, nitrifying bacteria reside deep within the biofilm due to their slower growth rates. However, in conventional biofilms, the inner regions of the biofilm experience the lowest oxygen concentrations, while in MBfR biofilms, the aerobic, nitrifying bacteria are positioned near the membrane, where oxygen concentrations are highest. Concurrent COD and total nitrogen removal is further discussed in Section 3.1.3.

Counter-diffusion also presents unique challenges. For both conventional and MBfR systems, excessively thin biofilms result in low fluxes due to biomass limitation. For MBfRs, excessively thick biofilms also provide low fluxes. In this case, the inner and outer portions of the thick biofilm introduce significant mass transfer resistance to the substrates, so that metabolic activity is limited to the center of the biofilm where concentrations are less. Thus, effective biomass management is critical for the optimal performance of MBfRs. This is discussed at length in Section 4.3.

3. Applications

3.1. Oxygen-based MBfRs

Air or oxygen-based MBfRs offer several advantages compared to conventional bubble aeration:

- The MBfR achieves high gas transfer rates, especially when high gas supply pressures or pure oxygen is used. Consequently, smaller tank sizes are required.
- In comparison to conventional bubble aeration, the MBfR's high gas transfer efficiencies can provide significant energy savings.
- The MBfR supports unique microbial community structures that allow for the simultaneous removal of COD and nitrogen from wastewater.
- The elimination of bubbling prevents the stripping of VOCs and greenhouse gases and can also avoid foaming when surfactants are present.

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