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Review

Versatile Gas-Transfer Membrane in Water and Wastewater Treatment: Principles, Opportunities, and Challenges

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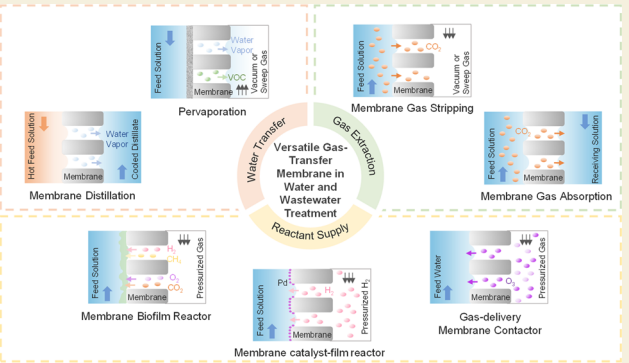
ABSTRACT: Technologies using liquid-transfer membranes, such as microfiltration, ultrafiltration, and reverse osmosis, have been widely applied in water and wastewater treatment. In the last few decades, gas-transfer membranes have been introduced in various fields to facilitate mass transfer, in which gaseous compounds permeate through membrane pores driven by gradients in chemical concentration or potential. A notable knowledge gap exists among researchers working on these emerging gas-transfer membranes as they approach this subject from different angles and areas of expertise (e.g., material science versus microbiology). This review explores the versatile applications of gas-transfer membranes in water and wastewater treatment, categorizing them into three primary types according to the function of membranes: water vapor transferring, gaseous reactant supplying, and gaseous compound extraction. For each type, the principles, evolution, and potential for further development were elaborated. Moreover, this review highlights the potential knowledge transfer between different fields, as insights from one type of gas-transfer membrane could potentially benefit another. Despite their technical innovations, these processes still face challenges in practical operation, such as membrane fouling and wetting. We advocate for research focusing on more practical and sustainable membranes and careful consideration of these emerging membrane technologies in specific scenarios. The current practicality and maturity of these emerging processes in water and wastewater treatment are described by the Technology Readiness Level (TRL) framework. Particularly, ongoing fundamental progress in membranes and engineering is expected to continue fueling the future development of these technologies.



KEYWORDS: *Gas-transfer membrane, membrane distillation, pervaporation, membrane biofilm reactor, MBfR, membrane-aerated biofilm reactor, MABR, membrane contactor, membrane gas stripping, MGS, membrane gas absorption, MGA*

1. INTRODUCTION

Membrane-based technology has been widely applied in environmental engineering, particularly in water and wastewater treatment using microfiltration, ultrafiltration, and reverse osmosis. These processes typically use membranes to extract clean water from feed solutions through membrane pores, which are also known as liquid-transfer membranes. Differently, the concept of using membranes for gas transfer in water/wastewater treatment, in which gaseous compounds pass through membrane pores from one side to the other, emerged in the 1960s and 1970s (Figure 1).[1−5] Investigations on gas-transfer membranes focused on addressing various needs in water/wastewater treatment.[6,7] Despite varying applications, a similar principle is shared, that is, to leverage the gradient between the two sides of membranes to drive gas transfer across the membranes. These gradients can refer to the differences in substrate concentration and chemical potential.[8−10] By regulating the applied gradient, the mass transfer can be controlled effectively to fit different requirements.[6] Moreover, the gas transfer efficiency through membranes is generally superior to traditional bubbling gas transfer methods.[6,11] Additionally, these processes also benefit from the general advantages of membrane-based technologies, such as high selectivity and a compact design.[12]

The properties of gas-transfer membranes are quite different from those of conventional liquid-transfer membranes. Early stage studies of gas-transfer membranes focused on identifying suitable materials that could effectively separate gases from

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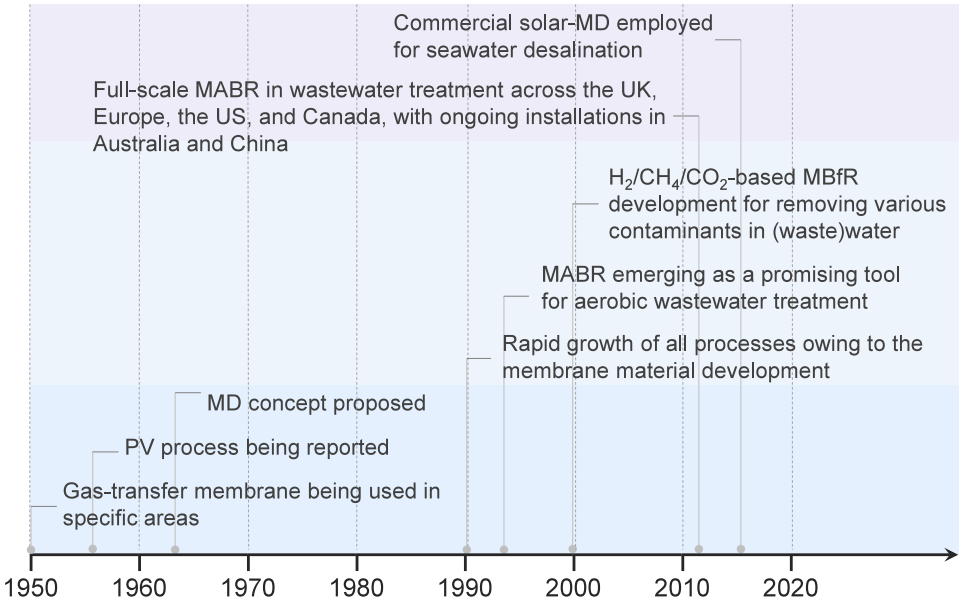

Figure 1. A brief history of the development of gas-transfer membranes. The timeline and arrangement of items are indicative.

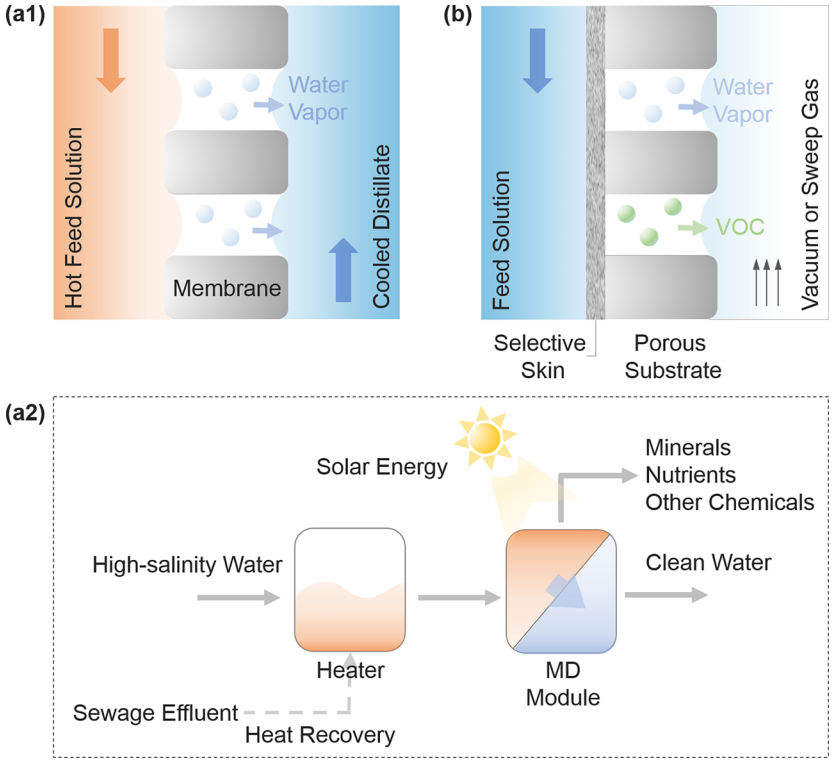

Figure 2. Schematic of a direct contact membrane distillation process: (a1) working principle and (a2) application scenarios. (b) Schematic of the principle of a pervaporation process. The PV process can also transfer volatile organic compounds (VOCs) in addition to water vapor.

liquids.[13,14] Hydrophobic polymers such as polytetrafluoroethylene (PTFE), polypropylene (PP), and poly(vinylidene fluoride) (PVDF) were among the first tested due to their high permeability and resistance to water. In the 1990s and early 2000s (Figure 1), significant technological innovations in membrane fabrication techniques, such as phase inversion, electrospinning, and stretching, led to the production of membranes with improved structural integrity, increased gas transfer rates, and cheaper prices.[15,16] The development of composite membranes, which combine different materials to enhance overall performance, opened new possibilities for more efficient and cost-effective solutions.[17,18]

Currently, gas-transfer membranes are extensively used in water and wastewater treatment through physical, chemical, and biological processes, showing considerable versatility. However, the considerably different applications of these membranes have led to relatively independent research efforts. For instance, researchers focusing on gas-transfer membranes for physical or chemical treatments typically concentrate on transmembrane mass/heat transfer and membrane modifica-

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tion. In contrast, those working on membrane-based biological processes are more interested in microbial communities but often neglect the role of the membrane itself. Consequently, a notable knowledge gap exists among researchers working on these emerging gas-transfer membrane processes, as they approach this subject from different angles and areas of expertise (e.g., material science versus microbiology).

In this Perspective, we seek to bridge this gap by fostering a more integrated understanding of gas-transfer membranes. This study categorizes the numerous applications of gas-transfer membranes into three distinct types, namely, water vapor transfer, gaseous reactant supply, and gaseous compound extraction. The principles, application contexts, and development of membranes used in each technology are briefly reviewed. We aim to elucidate the role of gas-transfer membranes in each scenario and highlight their advancements driven by the limitations and challenges encountered during operation. Given the similarity in gas transfer via membrane pores, we highlight the opportunity for cross-disciplinary knowledge transfer between fields as insights from one type of gas-transfer membrane technology could potentially benefit another. Additionally, the application scenarios and practical maturity of each process using gas-transfer membranes are also identified.

2. TRANSFERRING WATER VAPOR USING A GAS-TRANSFER MEMBRANE

In recent decades, processes that combine phase change with membrane separation have obtained wide attention. In these processes, liquid water typically transforms into a gaseous or molecular form at the membrane surface before transporting through the membrane pores. This implies that the driving force required for these processes is considerably lower than the pressure needed to drive liquid water through the membrane pores. Membrane distillation (MD) and pervaporation (PV) are two typical processes that utilize gas-transfer membranes for water vapor transfer.

2.1. Membrane Distillation

MD is a thermally driven process, which integrates membrane-based separation and thermal distillation. Direct contact membrane distillation (DCMD) is one of the most common MD configurations (Figure 2a1). In DCMD, a hydrophobic membrane is generally employed to separate the feed and distillate, allowing only water vapor to transport through its pores while retaining the nonvolatile foulants on the feed side (Figure 2a1). The driving force in MD is the vapor pressure difference between the heated feed solution and the cooled distillate. Therefore, the transmembrane pressure in MD is less affected by the feed salinity compared to that in pressure-driven membrane processes. This makes MD particularly advantageous in high-salinity wastewater desalination and zero liquid discharge processes.[19−21] Besides, MD requires lower operating temperatures (<80 °C)[22] and can be combined with alternative heat sources to significantly reduce energy consumption, showing potential for sustainable water treatment.

Specifically, in the MD process, water from the hot feed solution evaporates on the feed-facing side of the membrane. The resulting vapor is then driven by the transmembrane pressure gradient to pass through the membrane pores to the distillate-facing side, where it condenses back into liquid water. The vapor flux (J) can be described by Fick's law:[23]

$$J = B_m(p_{mf} - p_{mp})$$

where B_m is the membrane distillation coefficient, p_{mf} and p_{mp} are the partial pressures of water at the feed and permeate sides. The expression for the B_m varies under different flow regimes, but it is consistently related to membrane properties such as the porosity, pore tortuosity, pore radius, and membrane thickness. This indicates that these membrane characteristics significantly influence the mass transfer efficiency in MD.

In addition to the intrinsic physical properties of the membrane, the performance of MD is heavily dependent on the chemical characteristics of membranes, including hydrophobicity and chemical stability. The key to the successful operation of MD is to facilitate efficient vapor transport while preventing liquid penetration through membrane pores (i.e., membrane wetting). Thus, early research mainly focused on the selection and development of hydrophobic membrane materials, especially polymeric materials such as PTFE, PP, PVDF, and polysulfone (PSU).[13,14,24] However, the usage of hydrophobic polymer alone is insufficient to avoid wetting when treating different kinds of wastewater. Based on Wenzel and Cassie−Baxter theories, roughness plays a vital role in rendering the membrane surface hydrophobic.[25] Accordingly, for further enhancement of the MD performance, membrane fabrication techniques such as phase inversion,[26] electrospinning,[15] and stretching[27] have been introduced to optimize the surface morphology and interior structure of the membranes. Among them, incorporating nanoparticles into the membrane structure through electrospinning has obtained wide attention over the past decade.[28] Numerous studies have successfully deposited nanoparticles (e.g., ZnO, TiO_2, SiO_2, Al_2O_3) to render the membrane superhydrophobic or even omniphobic.[25] The omniphobic membranes are reported to repel not only water but also oils and other low-surface-tension liquids.[29] This can be attributed to their typically microscopic reentrant structure and very low surface energy. The reentrant structure imparts surprising liquid repellency to the membrane surface by trapping air between the liquid and solid surface to decrease the total contact area. Additionally, the low surface tension often makes the membrane surface slippery (i.e., sliding angle <10°), enhancing its ability to repel low-surface-tension matter such as surfactants. Furthermore, to enhance the flux and robustness of the membrane, graphene oxide (GO) and carbon nanotubes (CNTs) have also been widely used due to their high porosity and abundant functional groups.[30,31] However, the current methods for fabricating nanocomposite membranes via electrospinning still face challenges such as low productivity, potential environmental issues posed by materials, long-term stability of the membranes, and high costs. Consequently, current research mainly focuses on improving membrane fabrication efficiency, developing facile and one-step fabrication methods, and using environmentally friendly materials.[25] Additionally, to enhance the resistance to fouling by hydrophobic organics, Janus membranes comprising a hydrophilic surface and a hydrophobic or omniphobic substrate have been developed. Specifically, hydrophobic fouling can be repelled through the formation of a hydration layer on the hydrophilic surface. A recent study has reported the construction of a cellulose nanocrystal composite hydrogel layer, which exhibits robust resistance to surfactant-induced wetting, oil fouling, and even gypsum scaling at the same time.[32] Compared to polymers

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(e.g., polyamide (PA), poly(acrylic acid) (PAA), poly(vinyl alcohol) (PVA), poly(ethylene glycol) (PEG)) commonly used to engineer a hydrophilic surface layer, this highly hydratable surface not only improves antiscaling capabilities but also potentially has a relatively lower impact to the environment.

Energy consumption also remains a critical consideration in MD. The energy required to heat the feed solution can significantly reduce the economic viability of the MD process. Additionally, during long-term operation, heat loss from the feed solution further exacerbates heating costs. In recent years, some studies have introduced conductive materials during membrane fabrication to locally heat the liquid on the membrane surface via Joule heating,[33,34] without the requirement of heating the entire bulk solution. This approach significantly reduces heat loss in the feed solution and also mitigates the decline in mass transfer efficiency caused by the temperature polarization on the membrane surface. However, the economic feasibility of the energy consumption required for Joule heating in practical applications, as well as the water production rate of conductive membranes, still warrants further analysis and evaluation as the technology scales up. In recent years, photothermal materials have obtained wide attention in the fabrication of photothermal membranes, as the efficient use of solar energy presents a promising alternative for desalination. Chen et al.[35] reported the development of an omniphobic−photothermal nanocomposite membrane with carbon black nanoparticles deposited. Their study demonstrated that the solar energy utilization efficiency of this system is more than 1 order of magnitude higher than the energy efficiency of conventional MD. However, current research on solar MD usually overlooks the photothermal conversion efficiency during operation and the impact of the instability of sunlight on the system output. Despite these issues, solar MD and MD utilizing other low-grade heat sources (e.g., industrial waste heat) remain viable strategies for addressing water scarcity and the energy crisis.

Abundant studies have successfully advanced MD from bench to pilot scale. Compared to DCMD, vacuum membrane distillation (VMD) and air-gap membrane distillation (AGMD) have been more extensively studied at the pilot scale due to their higher overall energy efficiency and stability. Pilot-scale trials have evaluated MD's productivity, selectivity, and energy consumption in treating various feeds, including surface water, reverse osmosis brine, radioactive wastewater, and produced water.[36,37] They also focus on the rejection of certain viruses and pharmaceutical residues, as well as the recovery of valuable resources like ammonia and boron.[38,39] Compared to bench-scale experiments, pilot-scale studies allow for more reasonable adjustment and optimization of operational parameters as well as the design of membranes and membrane modules. Additionally, MD using low-grade heat has been tested at the pilot scale for desalination, aiming for sustainable full-scale application.[40,41] Integrated processes, such as forward osmosis-AGMD (FO-AGMD), have also been explored at the pilot scale, to further improve seawater desalination technologies.[42] Insights from these studies are essential for identifying potential challenges and solutions before commercial application. Although full-scale trials are limited, several studies have employed MD in large-scale modules to evaluate the technical feasibility and thermal available for desalination.[43] Moreover, models or conducted computational fluid dynamics (CFD) simulations have been developed to explore different factors with a wide range.[44] Data from bench- and pilot-scale experiments help simulate the feasibility of MD in specific scenarios. Although the MD process has not yet reached a mature commercial application, successful pilot tests and simulations are advancing it toward real-world implementation.

2.2. Pervaporation

PV is another gas-transfer membrane technology mainly for the separation of water and organic liquids. In a typical air gap/sweep PV process, vacuum or sweeping gas is applied on the permeate side to enhance a chemical potential gradient as the driving force. As the liquid is brought into contact with the feed side of a selective membrane, the better permeating component preferentially permeates and vaporizes on the other side, allowing for effective separation based on differences in affinity and permeability (Figure 2b).[8] Thereby, the PV process can overcome the limitations of thermodynamic vapor−liquid equilibrium (VLE) and decrease energy consumption with only latent heat required,[45] compared to distillation technologies. Accordingly, the PV process can work without the need for entrained additives to alter the VLE behavior, thus avoiding contamination of the final products. Therefore, in addition to transferring water vapor, PV is also advantageous in separating azeotropic mixtures, thermally sensitive components, and organic mixtures, as well as in removing and recovering dilute organic components in wastewater.[8,46]

PV and MD share similarities in maintaining the partial vapor difference across the membrane and in the phase change of the gaseous permeate, which may lead to confusion between the two processes. The primary difference between PV and MD is the function of membranes in the process.[47,48] Membranes act merely as a medium for vapor transport in MD and make no contribution to the separation. In contrast, membranes act as a molecule-scale selective barrier in PV and actively participate in mass transfer and separation due to their specific chemical properties. Therefore, the performance of PV is theoretically more dependent on the membrane properties.

The gas-transfer membranes used in the PV process are often made with a selective layer and a porous substrate. The generally accepted mechanism of mass transport through the dense layer is based on the dissolution−diffusion or adsorption−diffusion model,[8,49] which consists of three steps: (1) sorption of components from the liquid mixture onto the feed side of the dense layer, (2) diffusion of the absorbed species across the free volume of the membrane matrix, and (3) evaporation of the absorbed species as vapor from the permeate side of the dense layer. The diffusion step is regarded as the rate-limiting process, which can be generally given by Fick's law. Thus, the flux J can be described as follows:[8]

$$J_i = \frac{D_i K_i}{l} \cdot c_{if}$$

where D_i is the diffusion coefficient of component i in the membrane (m^2/s), K_i is the absorption coefficient, l is the membrane thickness (m), and c_{if} is the concentration of component i at the membrane-feed interface (kg/m^3).

Unlike hydrophobic MD membranes, which are prone to wetting issues, the PV membranes are inherently hydrophilic and thus free of such problems. However, the PV process faces challenges related to the trade-off between the selectivity and permeability as well as maintaining stability during long-term

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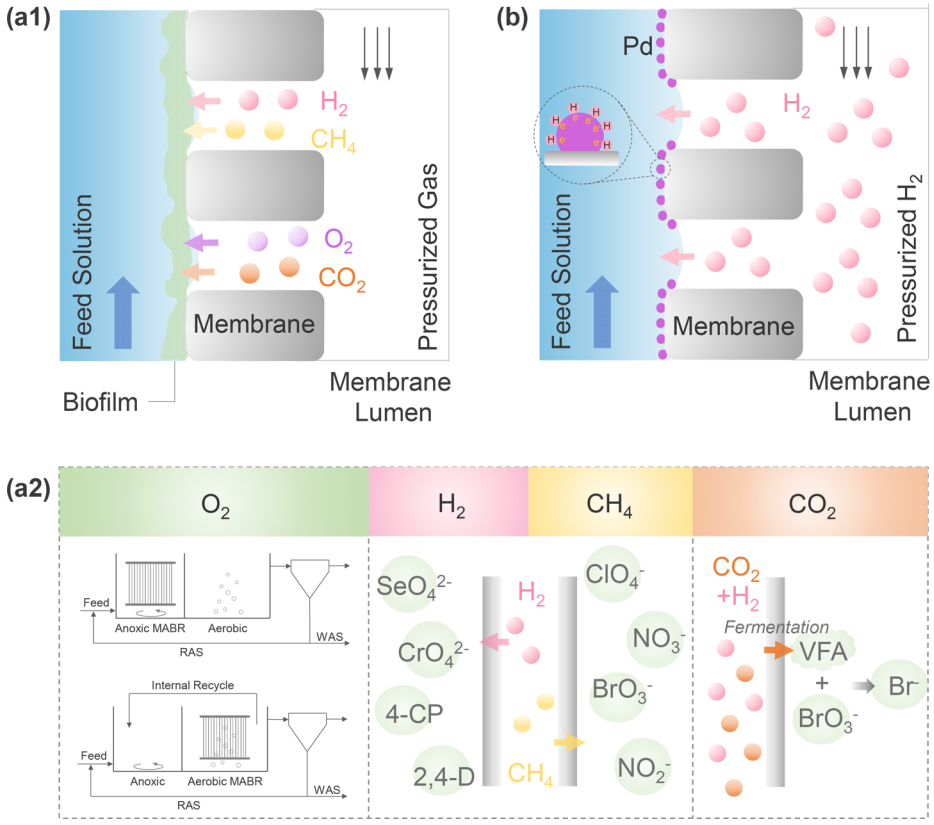

Figure 3. Schematics of a membrane biofilm reactor: (a1) working principle and (a2) application scenarios with different gases supplied. The O_2-based MBfR (MABR) can be incorporated into anoxic and aerobic tanks of wastewater treatment. H_2/CH_4-based MBfRs can be used to remove oxidized contaminants (e.g., NO_3^-, ClO_4^-, BrO_3^-, CrO_4^{2-}, and SeO_4^{2-}) and halogenated organic compounds (e.g., 4-chlorophenol [4-CP] and 2,4-dichlorophenoxyactic acid [2,4-D]). (b) Schematics of working principle of a membrane catalyst-film reactor. H_2 can be activated by palladium deposited on the membrane surface to remove oxidized contaminants and halogenated organic compounds.

operation. Researchers have designed and synthesized a wide range of membrane materials.[50] Among these, polymers remain the largest family of membrane materials, for the ease of fabrication and relatively lower costs than inorganic and hybrid materials.[8] Microporous inorganic membranes often show better PV performance than polymeric membranes due to their highly connected and relatively rigid transport channels. For example, zeolite membranes have attracted interest in both research and industry over the last decades, for the unique properties of zeolite in a film-like configuration, and consequently better resistance toward harsh chemicals, pressure, and thermal conditions.[51,52] To combine the strengths of both polymeric and inorganic membranes, the concept of mixed-matrix membranes was proposed. Numerous inorganic fillers (e.g., zeolites,[53] multiwalled carbon nanotubes, silica,[54] graphene oxide,[55] metal−organic frameworks,[56,57] and covalent-organic frameworks[58,59]) have been incorporated into polymeric matrixes for enhanced PV performance.[60] Compared with conventional polymeric membranes, the emerging hybrid nanoporous membranes are promising due to their robustness and diverse structure. Inorganic 2D materials (e.g., GO and MXene) have been reported to able to facilitate both high permeability and selectivity for molecule separations due to their interlayer channels or in-plane pores for molecule transport, and the atomic thicknesses.[55,61] Additionally, zeolite and metal−organic frameworks have also obtained increasing attention due to their unique merits such as well-defined pore structures, high porosity, and diverse building blocks, which facilitate higher flux and robustness of the membranes.[62,63] Recently, a novel type of porous material with skeleton-tunable nature and compatibility with the organic matrix, known as covalent-organic frameworks, has shown great potential in PV with enhanced separation performance.[64] However, the flux of mixed matrix membranes still needs to be improved.

Currently, the industrial applications of PV primarily focus on the dehydration of organic solvents.[45,65] Although the PV process has not yet achieved commercial application in water treatment, it shows promise due to its high selectivity and lower energy consumption. Niche applications in water treatment, such as removing volatile organic compounds (VOCs) and desalinating highly saline water and seawater,[18,61,66] have demonstrated good performance in pilot-scale tests. However, the cost and long-term performance of PV membranes remain key factors limiting their large-scale application. For instance, membrane materials that simultaneously exhibit satisfying hydrophobicity and organic stability are limited. Ongoing research is dedicated to the discovery and application of novel membrane materials and structures to enhance the PV process.

3. SUPPLYING GASEOUS REACTANTS USING A GAS-TRANSFER MEMBRANE

Certain gaseous compounds function as electron donors, electron acceptors, or carbon sources for biological and chemical reactions employed in water and wastewater treatment. For example, oxygen is utilized by aerobic microorganisms to remove organics and ammonium in

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(waste)water; hydrogen and methane are electron donors to facilitate the removal of oxidized contaminants and halogenated organics, including emerging per- and polyfluoroalkyl substances (PFAS); and carbon dioxide can be directly assimilated by autotrophic bacteria and microalgae used in wastewater treatment. To transfer these gaseous reactants into water and wastewater efficiently, gas-transfer membranes offer an efficient alternative to traditional bubbling gas-delivery methods.

3.1. Membrane Biofilm Reactors

The membrane biofilm reactor (MBfR) is an emerging technology in wastewater treatment, which delivers gaseous electron acceptors (e.g., O_2 and CO_2)[6,67] or donors (e.g., CH_4 and H_2)[68,69] directly via membranes to the biofilms attached and growing on the membrane surface.[70] In this case, membranes also serve as a substratum for biofilm growth and development in the MBfR. In a MBfR process, pressurized gas diffuses across the membrane wall under the concentration gradient without forming bubbles (Figure 3a1). This bubble-free aeration mode offers several advantages for MBfRs compared to traditional processes that rely on mechanical aeration. First, the gas transfer efficiency in the dead-end mode can reach nearly 100% compared to 5% to 50% with conventional bubbling aeration.[6,7] Second, stripping of the volatile compounds and greenhouse gases is mitigated under the bubble-free mode.[71] Third, the biofilm can grow and develop easily without scraping and disturbance by bubbles. In a MBfR, gas and the substrate in the bulk liquid diffuse relative to each other from two sides of the biofilm, which is also known as counter-diffusion.[71] Different from the co-diffusion in conventional biofilm systems, the counter-diffusion of gas reactants and contaminants determines the unique, stratified structure of the biofilm in the MBfR process.[5]

By supply of different gaseous reactants, MBfRs can be employed in various biological processes. For example, an O_2-based MBfR is used to oxidize contaminants in reduced states, such as organic carbon and ammonium.[7,72] In contrast, CH_4- and H_2-based MBfRs are applied to facilitate the microbial reduction of oxidized contaminants (e.g., NO_3^-, ClO_4^-, BrO_3^-, CrO_4^{2-}, SeO_4^{2-})[5,73,74] and removal of halogenated organic compounds (e.g., 4-chlorophenol and 2,4-dichlorophenoxyacetic acid)[75,76] (Figure 3a2). Moreover, CO_2, a known greenhouse gas, can also be supplied in the MBfR to produce volatile fatty acids (VFAs) (Figure 3a2).[67] Recent studies have demonstrated the coupling of CO_2 fermentation to VFAs and *in situ* utilization of VFAs for removing various oxidized contaminants in a MBfR.[77−79]

As the core to both the operation and cost of MBfR systems, design of membrane and module have been intensively discussed in previous reviews.[80] Here, we focus on the development and recent advancement of gas-transfer membranes in MBfRs. Membrane selection needs to be concerned with different aspects, including gas transfer resistance, mechanical strength, chemical stability, specific surface area, and cost. In previous studies, MBfRs have been tested using microporous, dense, and composite membranes.[5,81] Microporous membranes, made from hydrophobic polymers (e.g., PVDF, PTFE, and PP), are the most commonly used for their relatively higher gas transfer rates, lower costs, and ease of assembly.[82] However, the low bubbling point of microporous membranes limits the operational pressure,[5] while their porous structures also make them prone to clogging and wetting.[83] Dense membranes, in contrast, appear more suitable for wastewater treatment,[82] due to their ability to avoid membrane clogging and wetting.[5] The high diffusion resistance of dense membranes can be overcome by applying higher intramembrane pressure, which, however, usually corresponds to more energy consumption. For instance, silicone and polydimethylsiloxane, allowing for high intramembrane pressures, are popular materials in dense membrane fabrication.[84,85] In recent years, composite membranes with both high gas transfer rates and high bubbling points have attracted more attention. The composite membranes combine the advantages of microporous and dense materials through the addition of a thin, dense layer to a microporous layer. Owing to the low thickness, the thin dense layer can help raise the bubbling point without significantly increasing the diffusive resistance of the whole composite membrane, whether sandwiched between or on the top of the porous layer.[86,87] Unfortunately, composite membranes often require more sophisticated manufacturing processes and higher costs than single-layered membranes.[5] Therefore, membrane development for MBfR is still ongoing to obtain membranes suitable for large-scale applications with better economic performance.

Notably, the O_2-based MBfR, also known as the membrane-aerated biofilm reactor (MABR), has been applied in full-scale wastewater treatment plants across the UK, Europe, the US, and Canada, with ongoing installations in Australia and China.[88−91] When MABR is coupled with conventional activated sludge, the hybrid process has proven feasible for intensifying existing water and wastewater treatment capacity and reducing greenhouse gas emissions.[92,93] The unique counter-diffusion mode of oxygen and contaminants creates an inner oxygen-abundant layer and outer oxygen-depleted layer in the biofilm, housing diverse microorganisms in the same biofilm. Thus, it is feasible to achieve, for instance, simultaneous nitrification and denitrification and partial nitritation and anammox (PNA), in one single MABR system.[94−96] However, using MBfR to deliver other gaseous reactants is mostly limited to laboratories and a few pilot demonstrations. Future scaling-up is highly expected to facilitate the adoption of this emerging membrane technology by industries.

3.2. Membrane Catalyst Film Reactor

Similar to the MBfR configuration, the membrane catalyst film reactor (H_2-MCfR) is a recently demonstrated technology,[97−99] which replaces the microbial biofilm of the MBfR with an abiotic catalyst film (Figure 3b). The membrane used in MCfR serves both as a robust catalyst-anchoring substratum and as a controllable H_2-delivering medium. In MCfR, H_2 acts as an electron donor and reacts with metal catalysts (e.g., palladium[97,98]) to remove oxidized pollutants and halogenated organic compounds (e.g., trichloroethene, 2,4-dichlorophenol) in water. Similarly, the bubble-free gas transfer in MCfR can avoid the gas transfer limitation from H_2 diffusion, minimize H_2 discharge to the liquid (with the majority consumed in the catalyst film), and prevent the stripping of volatile compounds. The membranes used in these previous studies were nonporous PP membranes, characterized by high hydrophobicity and bubbling point. As an emerging technology, the potential of MCfR systems in water and wastewater treatment needs further study. For instance, the design and selection of membrane materials should also consider the robustness of catalyst deposition, in addition to other general

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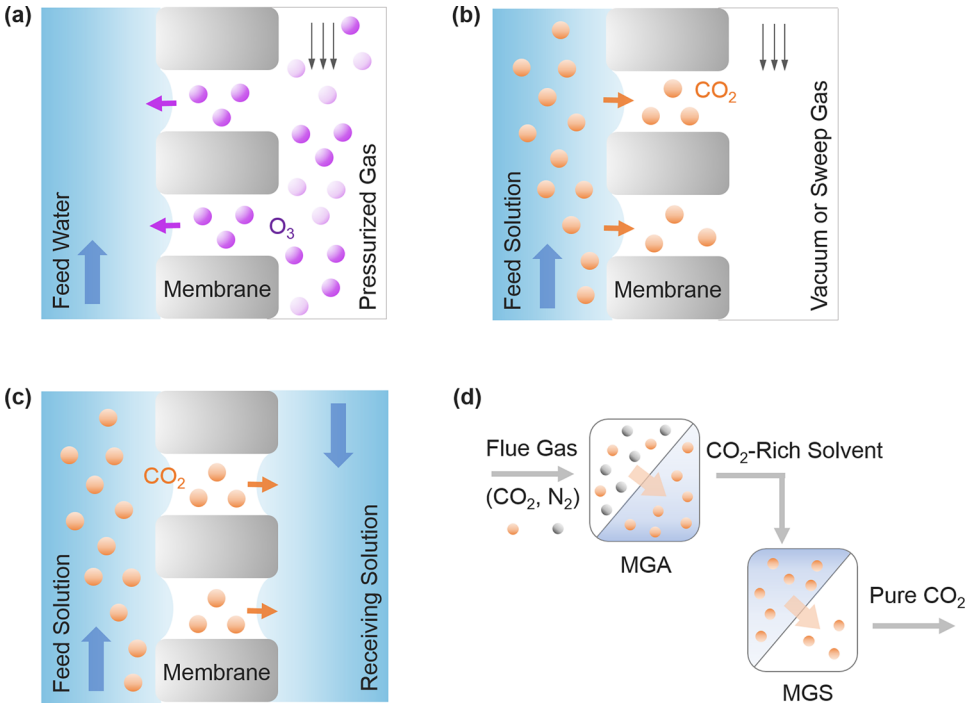

Figure 4. Schematics of working principles of (a) the gas-delivery membrane contactor (MC), (b) membrane gas stripping (MGS), and (c) membrane gas absorption (MGA). An exemplary application coupling MGA and MGS for CO_2 capture from flue gas is illustrated in panel d.

considerations aforementioned. Moreover, the MCfR can potentially be used to deliver different gaseous compounds other than H_2. This will require the selection of suitable catalysts to chemically activate these gaseous compounds under ambient conditions.

3.3. Gas-Delivery Membrane Contactor

The membrane contactor (MC) is a technology applied to facilitate mass transfer between different phases. While a MC can be applied for multiple purposes, including absorption, desorption, extraction, and even distillation,[9,100,101] one specific application is the gas-delivery MC, where membranes are utilized to deliver gaseous reactants in water treatment. Different from MBfRs and MCfRs in which the membranes also support biofilm and catalyst attachment, respectively, the membranes in gas-delivery MC are only used as the medium for gas transfer (Figure 4a). Thus, the MC membranes play a crucial role in maintaining a nondispersive contact between the gas and liquid phases, thereby enhancing the gas−liquid contact area and mass transfer efficiency through bubble-free operation.[11] In water and wastewater treatment, a gas-delivery MC is commonly used in the ozonation process, where the on-site produced O_3 mixture (O_2 and O_3) is transferred through membranes into water and wastewater to remove target pollutants.[102,103]

Hydrophobic polymeric and ceramic membranes have been used for gas-delivery MCs.[104,105] The durability of polymeric membranes made from PVDF and polydimethylsiloxane (PDMS) faces a significant challenge during long-term operation under the highly oxidative conditions of the ozonation process. Although ceramic membranes exhibit superior thermal, chemical, and mechanical stability, the hydrophobic modification of their surfaces is relatively expensive.[104] Therefore, the application of gas-delivery MC in ozonation treatment is mostly at the laboratory scale.[102] Further development in membrane materials is needed to obtain novel membranes that not only are easy and cost-effective to fabricate but also exhibit high mass transfer efficiency and extended service life.

4. EXTRACTING GASEOUS COMPOUNDS USING A GAS-TRANSFER MEMBRANE

As mentioned in Section 3.3, MC can also be integrated with gas extraction. Two primary forms of MC for gas extraction in water and wastewater are membrane gas stripping (MGS) and membrane gas absorption (MGA). MGS and MGA are both effective methods for extracting dissolved gases from liquids. In MGS, the concentration gradient across the membrane drives the dissolved gas to transfer from the liquid phase to the gas phase through membrane pores (Figure 4b). This gradient is maintained by applying a vacuum or sweeping gas on the gas-phase side (Figure 4b). In contrast, in MGA, the water or wastewater containing the target dissolved gas is placed on one side of the membrane, while a receiving liquid containing the corresponding absorbent is on the other side (Figure 4c). The target gas in water and wastewater diffuses through the membrane pores driven by the transmembrane concentration gradient and reacts with the absorbent in the receiving liquid (Figure 4c). This reaction reduces the vapor pressure of the target gas in the receiving liquid, thereby enhancing the driving force for mass transfer.

MGS and MGA have both similar and distinct applications in water treatment. MGS has been investigated in the degassing of water and wastewater, for example, the removal of O_2 and CO_2 from seawater and boiler feedwater, H_2S from natural water and wastewater, NH_3 from industrial wastewater, and dissolved CH_4 from anaerobically treated wastewater.[106−111] MGA processes have also been used to remove HCN and NH_3 from wastewater.[112,113] Notably, MGA is more commonly employed for separating and recovering acidic gases (e.g., CO_2 and SO_2).[114]

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In both MGS and MGA, membranes serve as tools for interphase mass transfer rather than selective barriers, providing a nondispersive contact and increasing the gas−liquid contact area. Consequently, membranes play a crucial role in enhancing mass transfer and mitigating issues such as flooding, foaming, channeling, loading, and weeping that are common in conventional contactors.[115,116] Microporous membranes made from hydrophobic polymers like PDMS, PVDF, and PP are frequently employed in MGS.[117−119] Notably, nanofiber membranes, with larger surface area and porosity, have demonstrated superior mass transfer coefficients compared to hollow fiber membranes.[117] Recently, the development of nonporous MCs has received much attention in specific applications, such as stripping CH_4 from anaerobic effluents, due to their excellent selectivity and resistance to membrane wetting.[120] Given the high mass-transfer resistance of dense membranes, composite membranes comprising a thin dense layer coated on a porous support have emerged as a promising alternative.[121] MGS holds promise as a replacement for conventional vacuum towers and forced draft degasifiers in water and wastewater treatment. In the past few years, the computational fluid dynamics (CFD) technique has been used to develop some machine learning (ML) models of MCs to simulate the carbon capture, biogas upgrading, and ozonation processes.[122,123] The theoretical removal of CO_2 or the ozone concentration under different operation parameters has been predicted and verified with experiment data. The models established can serve as a starting point for optimizing and scaling up the MC process.

Currently, several large-scale MGS modules have been tested in real-world applications,[124,125] which are approaching commercialization in specific applications such as the removal of VOCs and ammonia stripping. In contrast, the tests of MGA in water and wastewater treatment are mostly limited to laboratories. It should be noted that the commercialization progress of MGS and MGA processes is associated with factors such as membrane cost, durability, and membrane fouling. Therefore, further research and development are needed to create low-cost, fouling-resistant, and long-lasting membranes for the MGS and MGA processes. Additionally, it is essential to further evaluate the value of extracted gases.[126] For example, using MGS to extract and recover dissolved methane from wastewater usually yields a recovered gas containing highly diluted methane, which is not qualified for flaring or further gas upgrading.

5. FUTURE PROSPECTS

5.1. Knowledge Transfer among Diverse Gas-Transfer Membranes

The present study reviewed gas-transfer membranes based on their different functionalities, including water vapor transfer, gaseous reactant supply, and gaseous compound extraction. However, this classification may not fully cover their diverse functions in water and wastewater treatment. Particularly, each type of membrane may have additional applications beyond those discussed. For instance, PV membranes can also be used to extract gaseous compounds (e.g., chlorinated VOCs[66]) other than water vapor, depending on the chemical properties of the membranes. Additionally, some gas-transfer membranes share similar configurations but perform different functions. For example, the gas-delivery MC process involves membranes to supply gaseous reactants, while the other two types of MCs (i.e., MGA and MGS) use membranes to extract gaseous compounds from liquid. Therefore, using such general terminologies to indicate a membrane process without providing a specific context can be confusing in some cases, as the functionality of the same membrane can vary depending on how it is tuned and applied. Consequently, the specific functionality of membranes must be analyzed within the context of their intended application rather than only focusing on the membrane configurations.

It is essential to recognize that research on different gas-transfer membranes has largely been conducted independently, despite their shared foundational mechanism of using gradients across membranes for selective gas transfer. For instance, MD research primarily focuses on developing advanced materials and membrane modifications to address wetting issues but still encounters challenges of biofouling over time. In contrast, research on MBfRs predominantly emphasizes microbial community dynamics within biofilms, often with less focus on the role of membrane material properties. This separation is largely due to differences in research priorities and disciplinary expertise, which underscores a significant opportunity for interdisciplinary knowledge transfer. Insights from one membrane type could potentially offer valuable solutions for another, fostering mutual advancement in both fields. For example, advancements in antiwetting properties for MD membranes could potentially inform strategies to avoid wetting in MBfRs, enhancing the stability of long-term gas supply. Similarly, an in-depth understanding of biofilm formation in MBfRs could be beneficial in MD applications by guiding new approaches to biofouling prevention. Previous reviews and research papers have generally focused on individual membrane types without indicating these cross-functional similarities and differences. With recognition of the similarities across different gas-transfer membranes, collaboration and innovation within the broader membrane research community are highly recommended.

5.2. Critical Need for a Paradigm Shift in Membrane Development

Over the past two decades, research on membrane materials has advanced rapidly. In all of the processes covered in this review, the use of composite membranes has become a mainstream trend in recent years. The combination of a functional layer and a support substrate allows for improved performance while ensuring the mechanical strength and controlling the mass transfer resistance. In particular, the use of nanocomposite membranes has led to significant enhancements in the performance of various membrane-based processes owing to material diversity and optimized fabrication methods. For example, in the MD process, nanocomposite membranes have substantially alleviated membrane wetting and fouling challenges in lab-scale experiments.[29,30,32,35] In PV processes, nanocomposite membranes have significantly increased selectivity.[53,55,57,60] Regarding MBfR, MGA, and MGS processes, nanocomposite membranes have notably enhanced mass transfer efficiency.[117,120,121]

However, inherent material limitations have consistently constrained further improvements in the performance and energy efficiency of gas-transfer membranes.[127] For future research, we propose an urgent need to shift the research focus from exploring novel materials to enhancing the practicality and sustainability of membranes. First, enhancing membrane robustness is essential, particularly for complex real wastewater

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treatment, where long-term operational stability remains a critical challenge. Additionally, defects arising from the fabrication process must be further minimized. Second, the membrane costs should be further reduced. Although a dramatic decrease in membrane price has been observed in the past two decades, the capital and maintenance costs remain key barriers hindering the applications of gas-transfer membranes. Selecting readily available membrane materials and optimizing the fabrication methods are key to enabling cost-effective, large-scale production. Third, overall sustainability must be prioritized. In light of the global trend of seeking carbon neutrality, a critical and ongoing debate is whether the carbon emissions associated with membrane manufacturing should be included in the carbon footprint of water and wastewater treatment. Additionally, research efforts on recycling and repurposing waste membranes keep growing, as improper disposal of these polymers and organic materials could pose a significant threat to the environment.

5.3. Think before Acting: Specific Scenarios of Gas-Transfer Membranes

The placement of the right candidate in an appropriate context is pivotal for both the success of the technology and the overarching rationality of the design. Therefore, indiscriminately promoting the applications of gas-transfer membranes without a thorough evaluation of their suitability is not recommended. While the aforementioned processes offer certain technical advantages over traditional methods, each of them is often limited by different factors such as the complexity of membrane fabrication, energy consumption, and long-term operational stability. These constraints make some technologies more suitable for specific scenarios. Taking the MD process as an example, this process is generally not applicable to sewage treatment considering the high cost of heating large volumes of wastewater, especially in cold climates. Instead, MD stands out due to its minimal sensitivity to salinity compared to traditional pressure-driven membrane processes, such as reverse osmosis. This makes MD particularly promising for treating high-salinity water and wastewater (e.g., seawater, brackish water, reverse osmosis brine), recovering resources (e.g., valuable minerals and nutrients), and concentrating specific liquids (e.g., juice and acids).[19−21] Moreover, MD can become a sustainable water treatment option when alternative heat sources (e.g., solar energy and heat recovery) are available on-site or when dealing with high-temperature wastewater, such as textile effluents.[128] Similarly, the applications of all other gas-transfer membranes must be carefully evaluated before making the judgment to ensure both technological efficacy and economic feasibility.

5.4. Practicality and Maturity of Gas-Transfer Membranes in Real-World Scenarios

To evaluate the practicality and maturity of gas-transfer membranes in water and wastewater treatment, the Technology Readiness Level (TRL) framework is introduced.[129] Specifically, TRL 3−5 represents laboratory proof of concept and technology validation, TRL 6−7 indicates prototype development and pilot demonstration, and TRL 8−9 signifies full deployment and commercial use. Based on the previous discussion, MD and oxygen-based MBfR (MABR) appear to have the highest maturity level, both demonstrating excellent pilot-scale and even full-scale results, approaching or already achieving TRL 8−9. For example, technology developers such as Memsys, Aquastill, Scarab, Aquatech, and Memsift Innovation have successfully developed pilot-scale MD modules for seawater desalination, desalinating brines from thermal desalination plants, hypersaline groundwater desalination, oily wastewater treatment, and textile effluent treatment. Several commercial MABR products are also available on the market, including ZeeLung MABR by Suez Water Technologies & Solutions, OxyMem MABR by OxyMem Limited, and Fluence MABR by Fluence Corporation. These global efforts aim to facilitate the commercial application of MD and MABR technologies. In contrast, PV and MGS are nearly at TRL 6−7, showing promise in niche applications but with limited large-scale deployment. The overall costs remain a significant barrier hindering the further progress of such technologies.

Of note, in membrane science and engineering, fundamental research at low TRL levels focusing on tuning membrane materials and microscale structures is always an important research direction to address the challenges in their application, such as membrane fouling and wetting.[7,130] For example, the selectivity and permeability for specific gases need enhancement in certain processes, like using the MGS process in CH_4 recovery requiring avoiding the simultaneous extraction of H_2S and CO_2.[6] Therefore, fundamental research is essential to drive innovations in membrane materials, fabrication techniques, and configuration designs, which should also be paid sufficient attention.

It is important to note that this review focuses on the application of gas-transfer membranes in water and wastewater treatment, while their applications in other research fields may reach different readiness. Beyond water and wastewater treatment, gas-transfer membranes have extensive applications in other fields, such as chemical processing, biotechnology, and energy and food industries, demonstrating even higher adaptability. For instance, MGS and MGA have garnered significant attention in gas separation, particularly for CO_2 capture from flue gas (Figure 4d);[131] PV is more commonly applied in the dehydration of organic solvents in pharmaceuticals and fine chemical technology;[65] and MBfRs have been used in gas fermentation, converting gaseous compounds including biogas, syngas, and captured CO_2 into value-added products, such as fatty acids, single-cell proteins, bioplastics, and biofuels, among many others.[77−79]

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Notes

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