

Review

# Membrane-based biological waste gas treatment

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## Abstract

This article presents a literature review on developments of membrane reactors for biological waste gas treatment as well as examples of applications to different compounds. The use of membranes combines selective separation of compounds from a waste gas stream followed by biological removal. Gas transport phenomena and different types of membranes used in biological waste gas treatment are discussed. So far, membrane-based biological waste gas treatment has only been tested on laboratory scale. If the long-term stability of these reactors can be demonstrated, membrane bioreactor technology can be useful in the treatment of gas streams containing poorly water-soluble pollutants and highly chlorinated hydrocarbons, which are difficult to treat with conventional methods for biological waste gas treatment.

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## 1. Introduction

Membrane bioreactor (MBR) technology is advancing rapidly, and different MBR configurations have evolved during last 30 years [1]. MBR systems have mostly been used to treat industrial, domestic, and specific wastewaters, where a small footprint, water reuse, or stringent discharge standards are required. In this review, we will focus on transport and biodegradation of pollutants in membrane bioreactors for waste gas (MBRWG) treatment. In a MBRWG, gaseous pollutants diffuse through the membrane and are subsequently degraded by the microorganisms in the biofilm attached to the membrane surface [2–4]. Biomass may also be suspended in the liquid phase. MBRWG are especially favorable for poorly water-soluble compounds. Membrane materials can be dense, microporous, porous or composite. Dense materials are more selective, while microporous materials are more permeable but susceptible to plugging by biomass [5]. Passage of the pollutants contaminated air across the membrane allows passive diffusion of contaminants through the membrane into the liquid biofilm phase on the other side, driven by the concentration gradient [5]. The mass transfer coefficient through a dense membrane depends on the solubility and diffusivity of the contaminant. By careful selection of membrane

material, pollutants can therefore be extracted selectively from the gas phase [5,6]. While membrane bioreactors for volatile organic compounds (VOCs) treatment have yet to be tested at full scale, a number of features have emerged from the lab and pilot-scale work carried out to date. Compared with the traditional biological waste gas treatment techniques such as biofiltration, MBRWG have several advantages: presence of a discrete water phase allows optimal moistening of the biomass and removal of degradation products, avoiding inactivation of the biomass. Moreover, in MBRWG the gas and liquid flow can be varied independently without problems of flooding, loading or foaming [7]. A MBR also have the high construction cost disadvantage. Furthermore, their long-term operational stability still has to be demonstrated.

In this review we summarize the state-of-the-art of membrane based biological waste gas treatment. In addition, transport phenomena through membranes and development of MBRWG for biological waste gas treatment are summarized.

## 2. Membrane bioreactor configurations for waste gas treatment

Different membrane bioreactor configurations have been used, all on lab-scale: hollow fibre (i.d. < 0.5 mm), capillary (0.5 mm < i.d. < 10 mm), tubular (i.d. > 10 mm), flat sheet and spiral-wounded membrane type reactors [8]. A schematic

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### Nomenclature

$A$	membrane surface area ( $\text{m}^2$ )
$C_g, C_l$	gas and liquid phase concentrations ( $\text{g m}^{-3}$ )
$D$	diffusion coefficient of substrate ( $\text{m}^2 \text{s}^{-1}$ )
$EC$	elimination capacity ( $\text{kg m}^{-3} \text{day}^{-1}$ )
$EC_{\text{max}}$	maximum elimination capacity ( $\text{kg m}^{-3} \text{day}^{-1}$ )
$F$	mass flux ( $\text{g s}^{-1}$ )
$H$	Henry's law coefficient
$J$	flux of compound in membrane reactor ( $\text{g m}^{-2} \text{s}^{-1}$ )
$k_g, k_m, k_b, k_l$	gas, membrane, biofilm and liquid mass transfer coefficients ( $\text{m s}^{-1}$ )
$K_{\text{ov}}$	overall mass transfer coefficient ( $\text{m s}^{-1}$ )
$L$	length of reactor channel membrane (m)
$LR$	mass loading rate ( $\text{kg m}^{-3} \text{day}^{-1}$ )
$LR_{\text{max}}$	maximum mass loading rate ( $\text{kg m}^{-3} \text{day}^{-1}$ )
$P$	permeability through a dense membrane ( $\text{m}^2 \text{s}^{-1}$ )
$P_c$	permeability through a composite membrane ( $\text{m}^2 \text{s}^{-1}$ )
$P_i, P_o$	inlet and outlet reactor pressure (Pa)
$Q$	flow rate of gas phase ( $\text{m}^3 \text{s}^{-1}$ )
$Q_f$	feed flow rate ( $\text{m}^3 \text{s}^{-1}$ )
$Q_p$	permeate flow rate ( $\text{m}^3 \text{s}^{-1}$ )
$R_a^{-1}$	additional interfacial resistance ( $\text{s m}^{-1}$ )

### Greek symbols

$\delta$	membrane thickness (m)
$\delta_s$	thickness of porous support layer of composite membrane (m)
$\delta_t$	top layer thickness of composite membrane (m)
$\varepsilon$	porosity
$\eta$	removal efficiency (%)
$\varphi$	packing density ( $\text{m}^2 \text{m}^{-3}$ )
$\rho$	density ( $\text{g m}^{-3}$ )
$\tau$	gas residence time (s)
$\tau_m$	pore tortuosity
$\phi$	specific gas–liquid contact or membrane area ( $\text{m}^2 \text{m}^{-3}$ )

representation of a flat composite membrane bioreactor for the treatment of waste gas is shown in Fig. 1. In this concept, one side of the membrane is dry and acts as a surface for uptake of pollutants from the air flowing along the membranes, while the other side is kept submerged in a flowing nutrient solution and covered by a biofilm.

### 3. Mechanism of membrane-based biological waste gas treatment

Mass transfer and microbial kinetics of a gaseous pollutant within a MBRWG module can be described as follows:

- (1) Bulk mixing of the contaminant in the air entering the bioreactor.

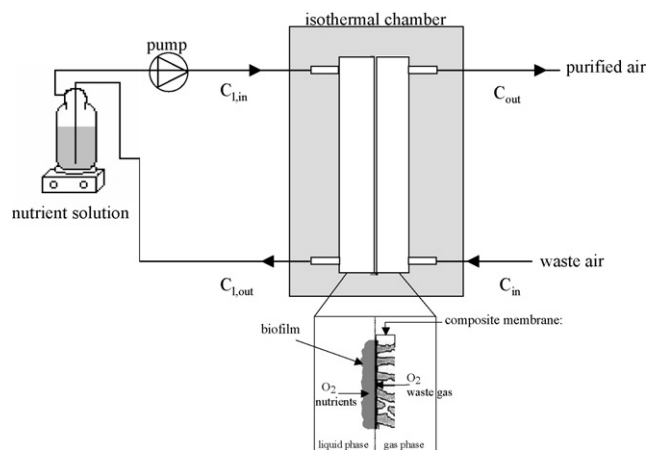


Fig. 1. Membrane bioreactor for removal of waste gas [2].  $C_{\text{in}}$  is the compound's concentration to be treated ( $\text{g m}^{-3}$ ),  $C_{\text{out}}$  the purified air ( $\text{g m}^{-3}$ ), and  $C_{l,\text{in}}$  and  $C_{l,\text{out}}$  are the concentration of nutrients inlet and outlet respectively.

- (2) Air boundary layer transport.
- (3) Transport through the membrane.
- (4) Transfer from the membrane, dissolution and diffusion into the biofilm.
- (5) Diffusion through and degradation within the biofilm.
- (6) Boundary layer transport through the liquid phase.
- (7) Subsequent mixing and degradation within the cell suspension.

The flux of a volatile component over the membrane in a gas–liquid membrane extractor can be described by the following formula [5]:

$$F = K_{\text{ov}} A \left( \frac{C_g}{H} - C_l \right) \quad (1)$$

where  $F$  represents the mass flux through the membrane ( $\text{g s}^{-1}$ ),  $K_{\text{ov}}$  the overall mass transfer coefficient ( $\text{m s}^{-1}$ ),  $A$  the membrane surface area ( $\text{m}^2$ ),  $H$  the dimensionless air–water partition coefficient ( $(\text{g m}^{-3})/(\text{g m}^{-3})$ ) and  $C_g$  and  $C_l$  the concentrations in gas and liquid phase ( $\text{g m}^{-3}$ ), respectively. The concentration difference between the gas and liquid phase provides the driving force for diffusive transport across the membrane. A pressure difference is not applied. The driving force strongly depends on the compound's air–water-partitioning coefficient. For components with a high  $H$ -value, the driving force for mass transfer is small. The concentration in the liquid phase, which depends on the microbial activity of the membrane attached biofilm and/or cells in suspension, also affects the driving force. The surface of the membrane forms the contact area. The overall mass transfer resistance ( $K_{\text{ov}}^{-1}$ ,  $\text{s m}^{-1}$ ) for gaseous pollutants in a membrane bioreactor is a combination of several resistances in series: gas phase ( $k_g^{-1}$ ), membrane phase ( $k_m^{-1}$ ), biofilm ( $k_b^{-1}$ ) and liquid phase ( $k_l^{-1}$ ) (Fig. 2). For a gas filled microporous membrane it is defined by:

$$\frac{1}{K_{\text{ov}}} = \frac{1}{k_g H} + \frac{1}{k_m H} + \frac{1}{k_b} + \frac{1}{k_l} \quad (2)$$

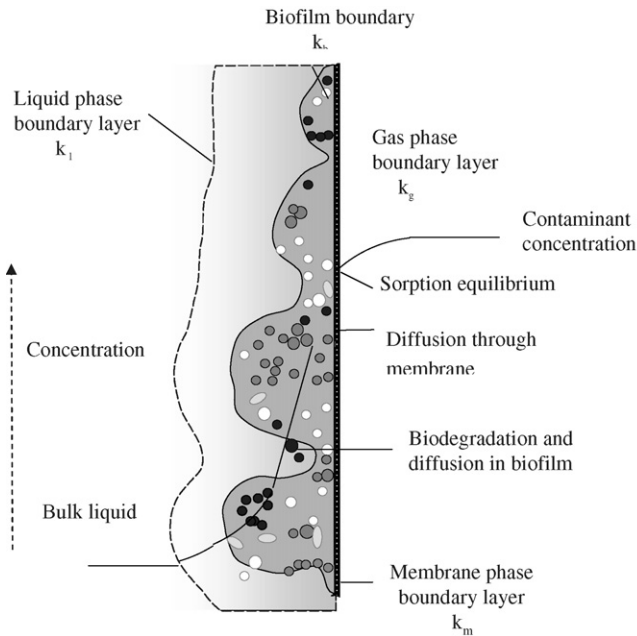


Fig. 2. Mass transfer resistance in a biofilm attached on a flat membrane.

Both  $k_g$  and  $k_l$  are function of feed flow velocity, the compounds diffusion coefficient, the viscosity, the density and the module geometry and dimensions. Several semi-empirical relationships for mass transfer coefficient in pipe and channels are reported in literature [8]. For the mass transfer resistance in the biofilm Lewandowski developed a method for calculating the thickness of the diffusive boundary layer (DBL) from substrate concentration profiles [9]. According to the thin film theory, the flux of substrate to a biofilm can be calculated using finite differences in Fick's diffusion equation:

$$J = D \frac{\Delta C}{\text{DBL}} \quad (3)$$

where  $J$  is the flux ( $\text{g m}^{-2} \text{s}^{-1}$ ),  $D$  the diffusion coefficient of substrate in stagnant water ( $\text{m}^2 \text{s}^{-1}$ ),  $\Delta C$  the difference in the solute concentration ( $\text{g m}^{-3}$ ) between the bulk liquid and at the reacting surface, and DBL is the thickness of the effective diffusive layer (m). From this definition, the mass transfer coefficient to the thickness of DBL,  $k_l = D/\text{DBL}$ . The value of the mass transfer coefficient depends on many factors, with hydrodynamics being the most significant, because flow velocity influences the thickness of the DBL. The higher the flow velocity, the thinner the DBL.

#### 4. Physical transport: membranes for mass transfer

A membrane may be simply defined as an interphase between two bulk phases of a system allowing the selective transport of compounds from one phase to other [10]. In waste gas treatment applications, gases are most often blown through the lumen of the membrane materials. Pollutants from the gas phase diffuse through membranes to a liquid phase on the shell side of membranes. The membrane also serves as a support for the microbial

population. Transport through the membrane takes place as a result of driving force acting on the compounds in the feed.

Gas separation in membranes occurs due to differences in permeabilities of the species flowing through the membrane. Membranes used for gas separation can be broadly categorized into porous, dense and composites. For successful application, membrane materials must strike a balance between reasonable mechanical strength, high permeability and selectivity [11].

##### 4.1. Microporous membranes

Microporous hydrophobic membranes are most often used in gas transfer applications because they provide high gas permeability, while not allowing transport of water across the membrane. Microporous hydrophobic membranes are available with pore diameters between 1000 and 10,000 Å [11]. The membrane pores remain gas filled and compounds transfer from the gas stream through the membrane pores by gaseous diffusion, usually the ratio between gas and liquid diffusivity is about  $10^4$ . At excess liquid side pressure above the critical pressure ( $\Delta P_{\text{cr}}$ ), water enters the pores of the membranes, significantly decreasing mass transfer rates [12]. Gas side pressure greater than the bubble point results in bubble formation in the liquid phase [13]. Within the excess pressure range of  $0-\Delta P_{\text{cr}}$ , the gas-liquid interface is immobilized at the mouth of the membrane pore on the liquid side [10].

##### 4.2. Porous membranes

Porous membranes have a well-defined static pore structure; it can be highly connected, non-connected or straight. Membranes can be classified according to their pore size as macroporous ( $>500$  Å) and mesoporous (500–20 Å) [14]. The mass transfer coefficient for the porous membrane type can be calculated as follows:

$$k_m = \frac{D\varepsilon}{\delta\tau_m} \quad (4)$$

with  $D$  being the diffusion coefficient in the gas phase ( $\text{m}^2 \text{s}^{-1}$ ),  $\varepsilon$  the porosity,  $\delta$  the membrane thickness (m) and  $\tau_m$  the tortuosity. The tortuosity is a measure for the shape of the pores.

Across these pore size regimes, gas transport in membranes may occur via different mechanisms such as Knudsen diffusion, viscous and surface diffusion [8]. Porous membranes have lower mass-transfer resistance than dense ones, but a disadvantage of these is biofouling [15].

##### 4.3. Dense membranes

Dense membranes rely on physical-chemical interactions between the permeating compounds and the membrane material. In dense polymeric materials, solution-diffusion is widely accepted to be the main mechanism of transport [16–19]. The mass transfer rate through a dense membrane depends on the solubility and the diffusivity of the permeating compound in the dense matrix [5,8]:

$$k_m = \frac{P}{\delta} = \frac{SD_m}{\delta} \quad (5)$$

where  $P$  is the permeability of the dense matrix ( $\text{m}^2 \text{s}^{-1}$ ),  $S$  the solubility coefficient or gas-membrane partition coefficient ( $\text{g m}_{\text{membrane}}^{-3} / (\text{g m}_{\text{gas}}^{-3})$ ) and  $D_m$  is the diffusion coefficient through the membrane ( $\text{m}^2 \text{s}^{-1}$ ). For each compound, the solubility and diffusivity are different, depending on the specific interactions between the compounds and the membrane. The transport mechanism is generally considered to be a three-step process. In the first step the gas molecules are absorbed by the membrane surface on the up-stream end. This is followed by the diffusion of the gas molecules through the polymer matrix. In the final step the gas molecules evaporate on the down-stream end. Dense membranes are limited to polymeric materials, such as latex, silicon rubber, polypropylene, and polyethylene, etc. They can be operated at high gas pressure, and are resistant to chemical as well as mechanical abrasion [11,20]. Dense membranes have also been shown to be more resistant to biofouling than porous membranes [21,22], possibly because of the hydrophobic nature of membranes resists attachment of microorganisms. The diffusion of gas through a dense membrane can be expressed by Fick's first law:

$$J = -D \left( \frac{dC}{dx} \right) \quad (6)$$

where  $J$  is the flux of the gas through the membrane,  $D$  the diffusion coefficient in the membrane, and  $dC/dx$  is the concentration gradient of the gas across the membrane. At steady state, the flux is a constant. If  $D$  is assumed to be constant, Eq. (6) can be integrated to give:

$$J = D \left( \frac{C_0 - C_1}{l} \right) \quad (7)$$

where  $C_0$  and  $C_1$  are the concentration of the gas on the upstream and down stream ends, respectively, and  $l$  is the thickness of the membrane. At low pressure, concentration of the gas in the membrane:

$$C = SP \quad (8)$$

where  $S$  is the solubility constant and  $P$  is the pressure of the gas. By substituting Eq. (8) into Eq. (7) we can get:

$$J = DS \frac{P_0 - P_1}{l} = P \frac{P_0 - P_1}{l} \quad (9)$$

where  $P$  is the permeability of the gas and according to Eq. (9) can be defined as:

$$P = DS \quad (10)$$

The permeability is therefore, a product of the diffusivity and solubility coefficient of the gas species. The diffusion coefficient ( $D$ ) and the solubility coefficient ( $S$ ) may both be function of concentration, so the theoretical analysis becomes more complicated. The idea of permeability being the product of a solubility term and diffusivity term is quite general. In Table 1, permeability and solubility coefficient of gases and vapour through PDMS are summarized. In gas separation with membranes, selectivity is defined as the ratio of individual gas permeabilities. The selectivity can therefore be viewed as a function of differences in both the diffusivity and solubility coefficient of the two gases.

#### 4.4. Composite membranes

A composite membrane combines the best characteristics of both dense (better interface) and porous materials (better mass transfer). Mass transfer characteristics for composite membrane are:

$$\frac{1}{k_m} = \frac{\delta}{P_c} = \frac{\delta_s \tau_m}{D\varepsilon} + \frac{1}{R_a} + \frac{\delta_t}{P} \quad (11)$$

where  $P_c$  is the permeability through a composite membrane ( $\text{m}^2 \text{s}^{-1}$ ),  $\delta$  the membrane thickness (m),  $\delta_s$  and  $\delta_t$  represent the thickness of porous support layer and dense top layer of the composite membrane (m), respectively,  $\tau_m$  the membrane tortuosity,  $R_a^{-1}$  the additional interfacial resistance ( $\text{s m}^{-1}$ ),  $k_m$  the mass transfer rate in membrane ( $\text{m s}^{-1}$ ),  $D$  the diffusion coefficient of compound in gas ( $\text{m}^2 \text{s}^{-1}$ ), and  $P$  the permeability through dense membrane ( $\text{m}^2 \text{s}^{-1}$ ). In a composite membrane bioreactor, a porous layer is used as support, while the thin

Table 1  
Permeability and solubility coefficient of gases and VOC in polydimethylsiloxane membrane arranged in order of decreasing value for Henry's law coefficient

Compounds	$H$ at 25 °C	$P$ ( $\text{m}^2 \text{s}^{-1}$ )	$S$	Reference
O <sub>2</sub>	32 <sup>a</sup>	$0.00691 \times 10^{-7}$	n.r.	[24]
CO <sub>2</sub>	n.r.	$0.036 \times 10^{-7}$	1.43	[24]
ET	n.r.	$0.028 \times 10^{-7}$	2.53	[24,25]
DMS	0.087 <sup>b</sup>	$0.561 \times 10^{-7}$	92.8	[24,25]
TCE	0.35 <sup>b</sup>	$1.43 \times 10^{-7}$	360	[24,25]
TOL	0.22 <sup>b</sup>	$2.1 \times 10^{-7}$	902	[24,25]
DCM	0.1 <sup>a</sup>	$5.6 \times 10^{-9}$	n.r.	[26]
DCE	0.05 <sup>a</sup>	$3.8 \times 10^{-9}$	n.r.	[26]
PROPAN	0.00028 <sup>a</sup>	$3.7 \times 10^{-10}$	n.r.	[27]
EOH	0.00021 <sup>a</sup>	$1.1 \times 10^{-11}$	n.r.	[27]
MeOH	0.00018 <sup>a</sup>	$4.9 \times 10^{-12}$	n.r.	[27]

Compounds: ET: ethylene; DMS: dimethylsulfide; TCE: trichloroethylene; TOL: toluene; DCM: dichloromethane; DCE: dichloroethane; PROPAN: propanol; EOH: ethanol; MeOH: methanol; n.r.: not reported or not sufficient data to calculate.

<sup>a</sup> Ref. [28].

<sup>b</sup> Ref. [29].

dense layer prevents microbial growth through the membrane. Hydrophobic microporous membranes coated with a thin layer of silicone have also been investigated [10]. The thin silicon layer increases mass transfer resistance but also decrease bio-fouling. The membranes are manufactured as small diameter (200–400  $\mu\text{m}$  i.d.) hollow fiber bundles that provide surface area to volume ratios as high as 30–100  $\text{cm}^{-1}$  [10]. Different types of composite membranes have been proposed to enhance membrane performance. A flat sheet composite membrane consisting of a dense polydimethylsiloxane (1 or 2.5  $\mu\text{m}$ ) top layer on a polyvinylidene fluoride (210  $\mu\text{m}$ ) support layer has been used for toluene removal [2,4,23]. De Bo et al. [2] used a flat sheet composite membrane consisting of a porous zircon (polysulfone membranes containing  $\text{ZrO}_2$  filters) support layer (175  $\mu\text{m}$ ) coated with a thin dense polydimethylsiloxane top layer (17  $\mu\text{m}$ ) for dimethylsulfide removal.

## 5. Mass transport in biofilms

Biofilms are assemblages of single or multiple populations that are attached to abiotic or biotic surfaces through extracellular polymeric substances (EPS) [30]. Several studies have determined the composition of communities present in biofilms in various environments [31–37]. The diffusion processes that occur within a biofilm matrix are dependent on the water-binding capacity and mobility of the biofilm. The matrix displays a high degree of microheterogeneity because of the numerous microenvironments that co-exist within it [38]. The spatial distribution of the diverse dissolved and particulate components through the biofilm matrix and the shape of its external surfaces influence the rates of the occurring bioconversions and the stability of the biofilm in terms of resistance to mechanical stress [39]. Other morphological features such as biofilm thickness and voids are also important [40–43]. Thick biofilms have high mass transfer resistances which reduce the flux of pollutant across the membrane. The effect of biofilm thickness has been studied experimentally [43] and by modelling approach [44]. Biofilm activity may also be affected negatively by roughness in the biofilm shape, an effect studied both experimentally [42] and by modelling approaches [45,46]. The phenomenon of mass transport in biofilms is influenced by biofilm structure, which in turn depends upon the local availability of substrate. Solute transport in biofilms is driven by diffusive transport within the denser aggregates and potentially convective transport within pores and water channels [47]. Biofilm structure is of special importance in the operation of biofilm reactors and strongly influence mass transport mechanisms within biofilms. A quantitative understanding of how biofilm structure is linked to mass transport is essential for understanding of biofilms. Diffusion has been shown to dominate mass transport in many biofilm systems. Two main approaches can be used to relate biofilm structure to mass transport. One approach is to explicitly describe the complex three-dimensional structure of the different biofilm components, which can be obtained from direct imaging of biofilms [48,37] or from mathematical modelling [49,50]. Another approach is to relate the overall biofilm diffusion to the biofilm structure based on macroscale parameters such as overall biofilm density

or porosity. A disadvantage of the latter approach is that the spatial resolution of three-dimensional biofilm structure is lost. However, the advantage is that established methods are available to measure parameters describing the overall biofilm structure and the overall diffusion coefficients. Biofilms are mainly composed of water and the macroscale diffusion coefficient for the biofilm ( $D_F$ ) is often related to the diffusion coefficient in pure water ( $D_W$ ), where  $f_D$  is diffusivity ratio [51]:

$$D_F = f_D D_W \quad (12)$$

Three main approaches have been used to quantify diffusion coefficients in biofilms experimentally: (1) the two-chamber method [51], (2) microelectrode measurements [52–54] and (3) quantification of the overall substrate removal and assuming a substrate conversion rate inside the biofilm [55]. These three methods have been applied to a variety of biofilms ranging from biofilms grown directly on membrane surfaces [56] to detached biofilms or activated sludge filtered onto a membrane [51]. Several reviews on diffusion in biofilms have summarized the available data [57–61]. However, it is only during the last decade that transport in biofilm systems have become a focus of interest for researchers in the field of bioremediation.

### 5.1. Biofilms in gas phase bioreactors

To date, little information exists about biofilm structure in bioreactors for waste gas treatment. Moller et al. reported on the structure of multispecies biofilms in a toluene-degrading biotrickling filter. *Pseudomonas putida*, the main primary pollutant degrader was present throughout the film, most probably because of large void channels in the biofilm allowing increased oxygen and toluene mass transfer. In situ toluene degradation activity of *P. putida* was found to be lower in biofilms than in suspension [37]. In another investigation reported by on biofilm structure of biotrickling filters and biofilters was determined in situ using computed axial tomography (CAT) scanning. The results show heterogeneous interfaces with air/water channels, image analysis allowed to calculate the gas/biofilm interfacial area [62].

However, such experience from the existing biofilm systems could lead to a better understanding of pollutants mass transfer in membrane bioreactors and ultimately to improve bioremediation process.

## 6. Development of membrane bioreactors in biological waste gas treatment

In Table 2 entries include reactor design, operation and performance parameters, observed range of individual pollutants, reactor dimensions, types of membrane, and inoculum type. Laboratory studies have demonstrated biodegradation of compounds with a broad range of air–water partitioning coefficients (five orders of magnitude). Efficient removal as single pollutants in synthetic waste air streams has been demonstrated for odorous sulfur, aromatic, and chlorinated compounds. The removal of poorly biodegradable compounds (such as DCM, DCE) and

Table 2  
Membrane bioreactors for biological waste gas treatment arranged in order of increasing values for EC<sub>m,max</sub> per compound

Compounds	H; Ref. at 25 °C	Time (days)	Inoculum (co-substrate); b = biofilm, s = suspend. cells	Reactor set-up			Experimental conditions		Reactor performance			Ref.			
				Configuration, type, material	A (m <sup>2</sup> )	φ (m <sup>2</sup> m <sup>-3</sup> )	φ (m <sup>2</sup> m <sup>-3</sup> )	C <sub>in</sub> (mg m <sup>-3</sup> )	τ (s)	EC <sub>m,max</sub> (g m <sup>-2</sup> day <sup>-1</sup> )	LR <sub>m</sub> (g m <sup>-2</sup> day <sup>-1</sup> )		η (%)		
MeOH	20 × 10 <sup>-5</sup>	n.r.	n.r.	b	C, NP, PDMS	12	n.r.	60	10–2600	n.r.	10	52	20	[75]	
BuOH	36 × 10 <sup>-5</sup>	1	n.r.	Activated sludge	b	C, NP, PDMS	0.31	1000	26	4–14	3.0–8.0	5.8	7.3	80	[76]
			135	Activated sludge	b	C, P, PSf	0.022	2070	155	917–1375	1.6/2.9	471	476	99	[70]
			90	Activated sludge	b	C, P, PSf	0.013	1500	92	2048–2315	1.6	1567	4148	38	[70]
NH <sub>3</sub>	66 × 10 <sup>-5</sup>	1	136	Activated sludge	b	HF, P, PO	0.063	20,000	126	42	0.4–1.3*	0.24	0.26	92	[69]
DCE	0.041	2	11	<i>Xa. autotrophicus</i> GJ10	b	SW, NP, PDMS	2.5	1250	n.r.	650	80–160	0.53	0.57	92	[77]
DCM	0.095	2	n.r.	Activated sludge	b	C, NP, PDMS	0.31	1000	26	4–21	3.0–14	0.098	0.25	38	[76]
			<1	Strain DM21	s	F, P, PP	0.0040	500	250	160	1.6–9.6	7.6	17	44	[63]
XYLs	0.17–0.25	2	n.r.	Activated sludge	b	C, NP, PDMS	0.31	1000	26	4–15	3.0–8.0	0.19	0.20	96	[76]
BENZ	0.19	2	100	Activated sludge	b	HF, P, PP	0.50	34,890	518	760	4.3*	2.6	2.7	98	[78]
			n.r.	Activated sludge	b	C, NP, PDMS	0.012	n.r.	n.r.	1445	2.9	39	259	15	[20]
			40	Activated sludge	b	C, NP, NLR	0.006	368	6.2	570	1.4	65	81	80	[78]
TOL	0.22	2	90	<i>Pseudomonas putida</i> Tol1A	b	HF, P, PE	0.23	10,256	205	377	0.8–4.2*	1.6	1.6	97	[68]
			<1	<i>Pseudomonas</i> GJ40	s	F, P, PP	0.0040	500	250	75	1.6–9.6	2.8	8.1	35	[63]
			120	Activated sludge	b	HF, P, PP	0.29	20,000	120	754–3770	0.9–1.8*	3.0	8.6	35	[3]
			168	Activated sludge	b	C, P, PSf*	0.056	2622	n.r.	754–2261	16/32	3.9	4.7	84	[64]
			n.r.	n.r.	b	C, NP, PDMS	12	n.r.	60	30–4200	n.r.	16	84	20	[75]
			150	<i>Pseudomonas putida</i> A1	b	HF, PE	0.082	n.r.	n.r.	743–2231	0.5–1.3**	n.r.	n.r.	86	[71]
339	<i>Pseudomonas putida</i> TVA8	b	CM, PDMS/PVDF	0.004	500	250	4–3200	2–24	17.7	23	84	[23]			
	37	Activated sludge	b	T, NP, PDMS	0.0096	558	12	4650	1.0	144	720	20	[79]		
TCE	0.35	2	21	<i>Methylosinus</i> OB3b (METH)	s	HF, P, PP	0.72	5000	2913	141–191	96–300*	0.018	0.034	52	[66]
			130	Activated sludge (TOL)	b	C, P, PSf*	0.056	2622	n.r.	80–107	21/42	0.054	0.102	53	[65]
			13	Activated sludge (TOL)	b	HF, P, PP	0.29	20,000	120	43–228	3.6–7.2	0.060	0.17	36	[67]
PROP	8.6	3	81	<i>Xa. Py2, Mycobacterium</i> Py1	b	F, P, PP	0.0040	500	250	17–1735	0.5	1.3	6.4	20	[80]
			34	<i>Xanthobacter</i> Py2	b	F, P, PP	0.0040	500	250	430–5163	0.5	3.6	6.2	58	[6]
			170	<i>Xanthobacter</i> Py2	b	C, P, PP	0.10	1966	637	568–6000	7.4–80	4.2	16	26	[72]
NITR	19.8	4	12	<i>Methylobacter</i>	b	F, n.r., n.r.	0.85	1020	283	6.2	30	0.015	0.017	88	[81]
			164	Activated sludge	b	HF, P, PO	0.063	20,000	126	124	1.9*	0.15	0.20	74	[82]
DMS			80	<i>Hyphomicrobium</i> VS	b	CM, PDMS/Zfr	0.004	500	250	33–375	8–24	1.9	2.7	74	[2]
HEX	74	1	n.r.	n.r.	b	C, NP, PDMS	12	n.r.	60	30–2400	n.r.	9.6	48	20	[75]
Mixtures BTEX			20	<i>Pseudomonas putida</i> TX1 & BTE1	b	HF, P, PP	1.4	20,522	2180	7680–15,360	8.0–16*	4.0	6.6	61	[15]
			52	<i>Pseudomonas putida</i> TX1 & BTE1	b	C, NP, PDMS	0.21	3920	337	2258–9783	4.3–15	7.5	8.4	90	[83]
MeOH			n.r.	n.r.	b	C, NP, PDMS	12	n.r.	60	110	n.r.	0.26	2.2	12	[75]
TOL									121		1.2	2.4	55		
HEX									112		0.17	2.2	7		

Configurations: HF: hollow fibre (i.d. < 0.5 mm); C: capillary (0.5 mm < i.d. < 10 mm); T: tubular (i.d. > 10 mm); SW: spiral-wound; F: flat membrane. Membrane type: P: porous; NP: nonporous; CM: composite membrane; \*gas residence time in lumen; \*\*gas residence time in shell and lumen. Membrane polymer: PP: polypropylene; PSf: polysulfone; PE: polyethylene; PDMS: polydimethylsiloxane; NLR: natural latex rubber; PO: polyolefin; \*pores are water-filled; PVDF: polyvinylidene fluoride; Zfr: zircon; n.r.: not reported or not sufficient data to calculate. References: 1 [73], 2 [29], 3[6], 4 [74]. Compounds: MeOH: methanol; BuOH: 1-butanol; NH<sub>3</sub>: ammonia; BENZ: benzene; TCE: trichloroethylene; TOL: toluene; PROP: propylene; NO: nitric oxide; HEX: hexane; DMS: dimethylsulfide; BTEX: mixture of benzene, toluene, ethylbenzene and xylenes; DMS: dimethylsulfide; DCM: dichloromethane; DCE: dichloroethane.

compounds that require cometabolism like TCE has also been observed [65–67].

Operating results in terms of EC are comparable to other conventional biological techniques, with a wide range of values reported. The ECs of the VOC undergoing treatment depend on many factors related to the design and operation of the MBRWG, as well as the properties of the pollutants. In particular, the water solubility and pollutants Henry coefficient are important. For easily biodegradable VOCs such as toluene, ECs of up to  $397 \text{ g m}^{-3} \text{ h}^{-1}$  can be obtained [23]. Hydrophobic VOCs are usually removed slower because of mass transfer limitations. In addition, EC can also be limited by biological reaction rate, that is, in the case of poorly biodegradable and/or toxic pollutants. Interestingly, some poorly biodegradable VOCs such as DCE, require a long start-up phase (months rather than days) before significant removal is observed, but once the reactor reaches steady state, the EC is comparable to that of more easily biodegradable pollutants [76,77].

Given that mass transfer limits most such systems, the flux limits EC. Reported VOC fluxes are roughly  $100 \text{ g m}^{-3} \text{ h}^{-1}$  in systems using single membranes, so much higher ECs can be achieved with systems using commercial designs with high specific areas (membrane area per reactor volume). In addition to good ECs at gas residence times of seconds, membrane biofilters are clearly able to operate under high pollutant loads and high pollutants concentrations. At high mass loading (short residence times), MBRWG become mass transfer-limited rather than biologically limited, as is observed at low mass loading [4,23].

As with biofilters the kinetic limitation may be due to either the electron acceptor or donor. Both enter the active biofilm from the membrane side, so the relative ratios of diffusion coefficient and degradation stoichiometry determine limitation, as is seen even in trickling filters. Van Langenhove et al. [4] reported that ammonia, provided as a nutrient from the liquid side, might decrease EC, perhaps because nitrifiers compete for oxygen with heterotrophs degrading organic contaminants.

A variety of membrane materials have been used in MBRWG, such as PDMS, PP and PE. Membrane materials are selected to provide high specific surface area and selected separation. Some membranes provide satisfactory support for the bacterial growth and this consideration is generally not a problem.

Depending on the inlet concentration and EBRT, removal efficiencies of individual compounds in MBRWG can be near 100%.

One point of concern is that VOC concentrations are too low to sustain an active, population degrading the VOCs. This may be of particular importance in MBRWG.

Biofilms in MBRWG do show some aging [43]. For example, clogging was reported when the liquid phase was on the tube side of HFMBR [5]. However, consistent removal has been reported for such systems in operation for at least 1 year. Another temporal issue is the aging of the membrane material. A decrease in the permeability of dense phase silicone rubber used intermittently over 2 years, and apparent intrusion of organisms into microporous membranes have been reported in a number of studies [15,78].

Little study has explicitly been made of the response of the membrane bioreactors to transient loads. Three important time-dependent conditions exist for bioreactors: startup response, response to varying loads, and long-term performance. Startup generally is accomplished by inoculation with an acclimated suspension, followed by a rapid development of activity, with apparent steady-state performance after 1–2 weeks [3]. During this startup period, an initial high removal at 1–2 days is apparently followed by a decline in performance, attributed to either starvation in the liquid phase as the forming biofilm inhibits mass transfer, or to changes in the membrane due to swelling.

MBRWG appear to respond well to diurnal loading based on a 40-h week [70,78]. A number of investigators have shown that MBRWG respond well to changing loads, with new steady states established in a few days, but have not reported results during this transition period.

Finally, comparison of the studies in Table 2 is very difficult because of different reactor configuration or operation and the rates of removal are highly pollutant/substrate dependent. Caution is needed in interpreting the results because the varying methodologies used in the respective studies raise difficulties for making comparisons.

## 7. Challenges for membrane technology integration in industrial processes

Membrane-based biological waste gas treatment is scientifically recognized as a suitable treatment technology. Membrane bioreactors will be at some point used because of no gas-phase clogging, high removal of poorly soluble contaminants, minimal water requirements, and competitive elimination capacities. However, in practice its use is limited, so far no full scale installation. The reasons proposed or possibly limiting the adoption of this technology are mainly membrane cost and robustness. From our viewpoint, the real bottlenecks can be summarized as follows:

- (i) Cost of the membranes as compared to conventional biofilter packing, illustrated by the analysis of De Bo [84]. With microporous membranes replaced every 3 years, both capital and operating costs are as much as tenfold greater than for any other common waste gas treatment methods.
- (ii) The robustness of the technology in terms of dealing with fluctuating pollutants load, wide range of temperature and humidity.
- (iii) Excessive biofilm growth is one of the major drawbacks of membrane biofilters. The accumulation of biomass can lead to membrane fouling, resulting in mass transfer limitation of substrates (VOC and oxygen) leading to a decline of biomass activity and finally to the breakdown of the reactor.
- (iv) Lack of demonstrated multiyear performance.

## 8. Conclusions and future directions

Membrane bioreactors have opened the possibility to treat low concentrations of volatile and/or poorly water-soluble

pollutants from waste gas. Different membrane bioreactor configurations have been used, i.e. hollow fibre, capillary, and flat sheet. Selection of membrane material mainly depends upon the mass transfer properties of gaseous pollutant within MBRWG. For the successful application, the membrane material should strike a balance between reasonable mechanical strength, high permeability, selectivity and a support for the microbial population. So far, PDMS membrane has been reported as a suitable material for the biological removal of waste gas.

However, all the studies presented in this review are lab scale studies, and little is known about the interference of this technology by the presence of other volatiles in the waste gas. The effects of biofilm materials on the membrane surfaces in the long run have not been sufficiently tested. In addition to the durability of the membrane material, the stability of the biomass is essential as well.

Future research must focus on removal of gaseous mixtures, effect of temperature, and humidity to demonstrate and evaluate MBRWG performance, both under controlled conditions in lab-scale and pilot-scale MBRWG placed on industrial sites. As biofilm morphology is of special importance in the operation of biofilm reactors, research should also focus on biofilm material (thickness, location, diffusion through biofilm, quantification of microbial population). For the financial implications and technology developments, the research should also focus on process design, taking several aspects in to consideration such as costs, ease to control biomass, and membrane density.

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