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# Gas Phase Membrane Bioreactors

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**Abstract**
This mini review aims to introduce the gas phase membrane bioreactors (GPMBR). These systems can be considered as a combination of membrane contactors and bioprocesses. Constructions, features, and operations of these structures are described, moreover examples are given for their application possibilities, comparison to conventional processes is presented, advantages, limitations and perspectives are discussed.

**Keywords**
waste gas, pollutants, integrated systems

## 1 Introduction

Membrane bioreactors (MBRs) can be considered as integrated systems where a membrane separation step and a bioprocess take place simultaneously [1, 2]. In the MBR systems designed for treatment of gas compounds, gaseous substrates are transferred through a membrane to the other side, where the biocatalysts are located [3–5]. Thus, the MBR combines the beneficial properties of the membrane techniques and the clean, mild biochemical reaction. Moreover, these equipments can be operated simply, contain no moving parts, easy to scale-up, gas and liquid flows can be varied independently, no flooding, no foaming problems [4]. There are some drawbacks, as well: the quite high construction cost and the shaky long-term stability.

## 2 About the membranes

The role of the membrane here is two-fold: to separate the gas and liquid phase on one hand, and to provide a suitable surface for the contact of the two phases, on the other hand. Therefore, this membrane technique is often called membrane contactors. They were first used in blood oxygenation and tested as artificial gills [4].

Membrane contactors [1, 6–9] can be divided into two groups: (i) where both phases are liquids and (ii) where one phase is liquid, the other is gas. Moreover, the membrane itself may have pores or have a non-porous character (film or dense membrane). For the treatment of gaseous compounds, gas-liquid contactors are usually applied in hollow fiber modules with high surface area (having high packing density). These membranes provide a suitable contact area for the process.

Regarding transport, the gas molecules may be driven in both directions: from the gas phase into the liquid and the other way around: from the liquid phase into the gas phase. Membrane contactors have been used for e.g. aeration (wastewater oxygenation), or for gas absorption (elimination of sulfur dioxide or ammonia from gas mixtures).

During the operation of the contactors using porous membranes one of the most important parameters is the wettability of the membrane. If the liquid used is an aqueous phase, and the membrane is hydrophilic, then the liquid phase will fill all the pores of the membrane. If the membrane is hydrophobic, on the other hand, the gas molecules will occupy the pores.

The mechanism of the transport of the gas molecules in the porous membrane contactors is diffusion [1, 4]. Usually, no transmembrane pressure is applied. The driving force here is only the concentration difference between the two phases.

When dense membranes are used, the transport takes place in two steps: firstly, the gas molecules are absorbed into the membrane material, then diffusion occurs (solution-diffusion mechanism). Thus, the hydrophilic or hydrophobic character of the membrane is still important, since it is strongly connected with the solubilities.

Regarding the membrane modules, flat and tubular membranes can be used mainly for testing

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purposes, preliminary laboratory measurements. Hollow fiber (or capillary) membrane modules are more appropriate for semi-pilot and pilot experiments, since their packing density (the ratio between the membrane surface area and the volume of the membrane module) is much higher than flat or tubular modules, thus smaller membrane area is enough for the treatment of larger volumes.

## 3 Bioprocesses

Biological treatment of gaseous pollutants generally means the degradation of these compounds by various microorganisms, mainly bacteria. The biomass may be present in the form of biofilm on the membrane surface or suspended in the liquid phase [4, 10].

In many cases carbon and oxygen sources are provided for microorganisms from the gas, but the other nutrients should be supplied additionally. The membrane itself serves not only as an interface between the phases, but (i) it should be non-permeable for the microbes and (ii) it must protect the microbes from the effects of certain hazardous components (e.g. heavy metals) present in the gas phase.

Most of the gaseous compounds – where MBRs were used for the treatment – belong to the group of volatile organic compounds (VOCs). VOCs mean volatile organic compounds comprising various chemicals, which are easily vaporized and released as gases from certain liquids or solids. These compounds have high volatility, mobility and are usually resistant to degradation.

Many VOCs are considered to be toxic, dangerous and harmful to health. They are formed during the production of, e.g., solvent-based paints, printing inks, many consumer products, organic solvents and petroleum products.

Beyond VOCs, other gaseous compounds can be treated with this technique, as well, e.g., $H_2S$ or $CO_2$ elimination from biogas, or $NO_x$ removal.

## 4 Classifications, examples

Gas phase membrane bioreactors (GPMBRs) have been studied so far for the treatment of numerous volatile (or vapor) compounds including VOCs and other components. In this chapter these processes are classified according to the membrane module used: tube, flat sheet and hollow fiber (capillary).

### 4.1 Removal of VOCs

VOCs can be classified (arbitrarily) into four groups:

- chlorinated hydrocarbons (e.g., trichloroethene, trichlorobenzene, dichloromethane, etc.)
- aromatic hydrocarbons (e.g. BTEX: benzene, toluene, ethylbenzene, xylenes; …)
- "classical" organic solvents (e.g. *n*-hexane, *i*-octane, 1-butanol, methanol…)
- sulfur containing compounds (e.g. dimethylsulfide…).

Examples from these groups are given in Tables 1–3 sorting them based on the membrane module applied. They give some data on the inoculum used, moreover the removal efficiency ($\eta$) and the reference, as well.

Table 1 presents some details of the processes carried out in porous tube membrane modules. It seems that the first studies on application of MBRs for treatment of gaseous compounds were published in 1986 by German researchers [11]. They investigated the biodegradation of xylene, *n*-butanol and dichloromethane (Table 1) using

**Table 1** Porous tube membranes

| Compound | Inoculum | $\eta$ (%) | Ref |
|---|---|---|---|
| xylene, *n*-butanol, dichloromethane | activated sludge | 5-25 | [11] |
| *n*-hexane, toluene | activated sludge | 10-30 | [12] |
| toluene | activated sludge | 20 | [13] |
| benzene | activated sludge | 80 | [10] |

**Table 2** Flat sheet membranes

| Compound | Inoculum | $\eta$ (%) | Ref |
|---|---|---|---|
| toluene, dichloromethane | *Pseudomonas* | 8-35 | [14] |
| propene | *Xanthobacter* | 10-50 | [15] |
| toluene | *Burkholderia vietnamiensis* | 82 | [16] |
| trichloroethane | activated sludge | 13 | [17] |
| dimethyl-sulfide | *Hydro-microbium* | 74 | [18] |

**Table 3** Hollow fiber membrane modules

| Compound | Inoculum | $\eta$ (%) | Ref |
|---|---|---|---|
| trichlorobenzene | denitrifying sludge | 94 | [19] |
| toluene | *Pseudomonas putica* | 86 | [20] |
| toluene | activated sludge | 59 | [21] |
| benzene | activated sludge | 98 | [22] |
| xylene | activated sludge | 59-84 | [23] |
| *n*-hexane | activated sludge | 20 | [24] |
| *i*-octane | activated sludge | 85 | [24] |
| methanol | *C. boidini* | 90 | [25] |
| 1-butanol | activated sludge | 99 | [26] |

silicon tube membranes and activated sludge [10–13]. Later the investigations were extended to toluene, hexane and other VOCs, where microporous hydrophobic membrane materials were used.

Beyond tube modules, flat sheet membranes (Table 2) could be found, as well, in the first couple of studies on GPMBR (as early as 1992) [14–18]. It was used for toluene and dichloromethane degradation. Then the research was further expanded and developed: not only porous, but non-porous (composite) membranes have been applied using various microorganisms.

Hollow fiber modules can be considered as the most popular membranes recently in GPMBRs (Table 3) [19–26]. Both porous and non-porous membranes have been applied in the experiments, and the efficiency levels seem higher.

### 4.2 Other applications

$H_2S$ elimination from biogas is a large volume application studied for a long time. It can be completely removed by GPMBR [27] (Table 4). Biogas production from kitchen waste has been studied intensively recently [28], since it is a separately collected section of solid wastes. The odorous $H_2S$ formed during the process – similarly to the conventional method – should be removed.

Another interesting possibility is $NO_x$ removal, where oxidation or decomposition of gaseous $NO_x$ takes place by nitrifying bacteria (typically derived from activated sludge). NO removal was studied by Chinese researchers using activated sludge for the treatment. In both cases hollow fiber membrane modules were used [29, 30].

## 5 Comparison with other, non-membrane techniques

MBRs can be considered as an alternative for conventional bioreactors applied for treatment of gas compounds. Biofilters, trickle bed reactors and bioscrubbers belong to the most important traditional techniques [4, 31]. All the three reactors are actually packed bed bioreactors, where the gas is blown through.

In biofilters the bed contains is usually compost [32], where the microbes are present to degrade instantly the dangerous organic pollutants. The biomass should be provided with some water (prehumidification) during the process.

**Table 4** Other applications of GPMBR

| Compound | Inoculum | $\eta$ (%) | Ref |
|---|---|---|---|
| $H_2S$ | activated sludge | 99 | [27] |
| NO | activated sludge | 73 | [29] |
| $NO_x$ | denitrifying bacteria | 80-95 | [30] |

In trickle-bed reactors and bioscrubbers inert bed is used, and it is always wet since it is continuously humidified (by spraying water). Microorganisms are placed on the bed in case of trickle-bed reactors, while in bioscrubber reactors [33] they are located in a separate tank. Thus, the gaseous compounds should diffuse through a water phase to reach the degradation step. If the water solubility of the gaseous compounds is low (as it is the case in most VOCs), this diffusion step means a substantial barrier regarding mass transfer.

In MBRs, however, the gas compound should pass through only the membrane, liquid (water) phase does not form a resistance. It means that the pressure drop is much lower, as well, than in the case of e.g. biofilters.

Applying hollow fiber membrane modules for the process, a large gas-liquid interface can be provided (even 10 000 $m^2/m^3$), ensuring high mass transfer rate, which is again a beneficial character of MBRs compared to the traditional techniques.

## 6 Operation of GPMBR

Regarding operation of these GPMBR, recirculating and non-recirculating modes of operation are possible to use. Most of the systems work by recirculating the gas containing the pollutants, which is regarded as a batch reactor. In non-recirculating mode, it is possible to form a continuous operation, that is more advantageous from the higher volume, industrial application points of view [34].

The removal efficiency is sometimes difficult to compare since the initial conditions (e.g., concentration of the pollutant, volume of the treated gas…) are different. Perhaps the elimination capacity (g pollutants removed during one day related to the membrane surface area, g $m^{-2}$ $d^{-1}$) is a better unit for comparison [11]. The other environmental factors, like gas residence time, mass loading rate, etc., however, have significant impact on the process efficiency, as well.

The treated gases are usually waste gases or air containing pollutants (waste air). Both are delivered to the GPMBR at atmospheric pressure; thus, no transmembrane pressure can be applied (or it is extremely difficult and costly). These sources of gases may contain not only one but several pollutants, which are – as expected – more difficult [11] to degrade in one step, and various synergistic effects may occur.

## 7 Perspectives

GPMBRs are considered as novel alternative systems for effective treatment, removal of volatile compounds from gaseous media. The development of the technique is still in progress, new methods and practices are described in the literature. Ozonisation, for instance, is an interesting way to improve the treatment of xylene [23]. It was found that addition of $O_3$ has significantly enhanced the removal efficiency of xylene in GPMBR, since it helped the first oxidation step of xylene biodegradation to proceed.

Volckaert and colleagues have applied silicon oil to improve mass transfer in the reactor [35]. Water-silicon oil emulsion inoculated with activated sludge was used as a recirculation mixture to enhance removal efficiency.

Another exciting design strategy is when both aerobic and anaerobic regions are constructed within the biofilm of the MBR [4], as was presented for trichloroethene treatment. In this way dechlorination takes place in the anaerobic zone, while further degradation occurs in the aerobic zone takes place.

Regarding novel bioprocesses involving gas compounds, that may be realized in GPMBR with higher efficiency, nitrogen fixation could be a promising candidate [36]. The process itself is actually a novel method to produce a nitrogen bioinoculant by enriching a soil microbial community in bioreactors supplying $N_2$ by air pumping. The air might be delivered into the bioreactor through a hollow fiber membrane module resulting in a higher contact surface area.

In GPMBRs mainly bacteria have been used for the biodegradation process so far. Some fungi and algae, however, may find utilization [11] in the near future in special cases. For example, recently in the frame of a project at University of Boras (Sweden) the aim was to develop an optimized biological process to purify biogas: i.e. to eliminate $H_2S$ and remove $CO_2$. A MBR and microalgae are applied as an integrated system (Fig. 1) resulting in three product streams during the process: a stream rich in oxygen, a stream rich in methane (purified biogas), and algae biomass. This scheme, however – in my opinion – is not suitable for the establishment of a successful system, it should complete with a gas separation unit to separate the methane from the pollutants of the biogas, as a first step of the process. In this way the $H_2S$ can be eliminated in the MBR (containing biofilm) and then the $CO_2$ rich stream can be flown into the algae bioreactor to consume it. The advantage of this kind of operation is that no need of a stirrer, the bubbles of the gas stream will provide an appropriate mixing in the reactor.

Moreover, bioprocesses using enzymes as biocatalysts can be applied in GPMBR, as well, for conversion of certain substrates. For instance, lipase enzymes are able to convert volatile alcohols and acids in vapor phase into esters [37]. This kind of enzymatic reaction could be implemented in GPMBR. As a summary it can be declared that GPMBRs have demonstrated their usefulness in many areas in the

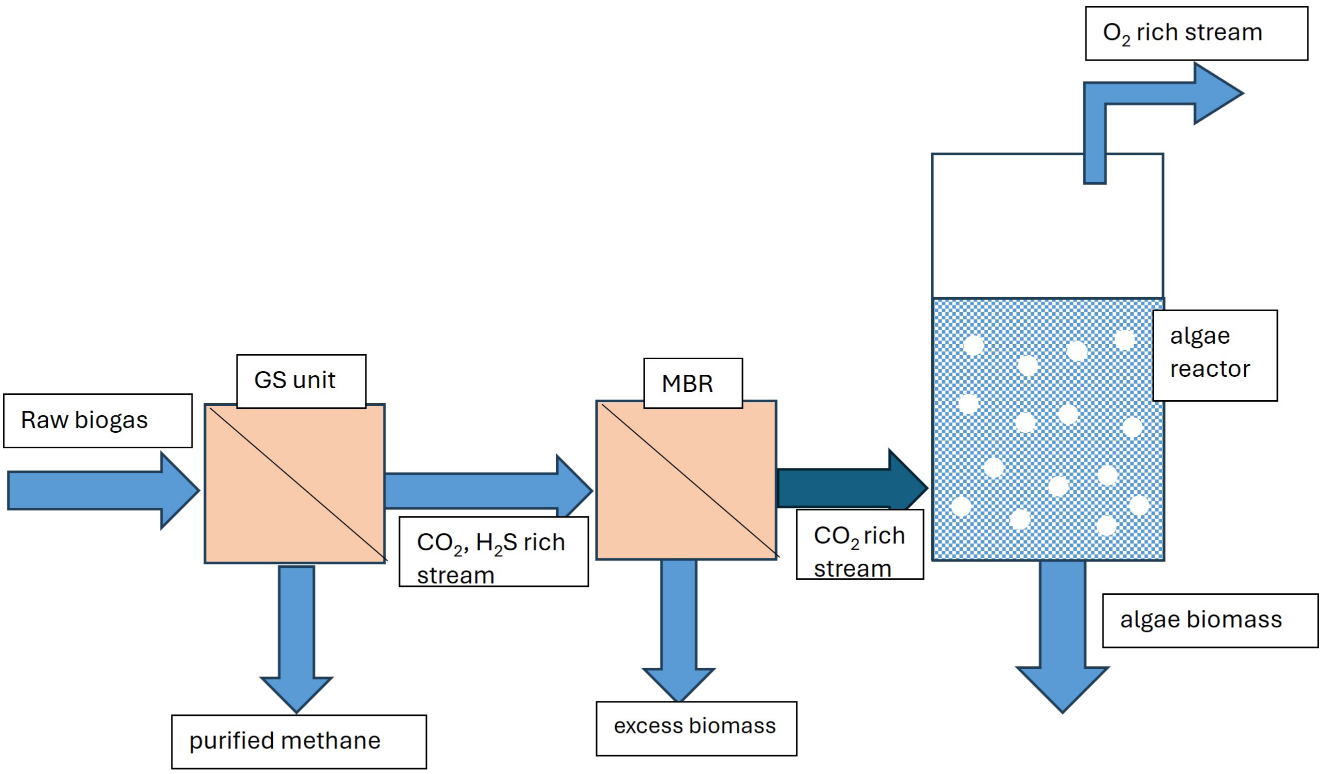


**Fig. 1** Scheme of the operation of a gas separation membrane unit, a membrane bioreactor + algae system for integrated biogas purification (elimination of $H_2S$ and $CO_2$)

treatment of gas phase compounds and seem to have a promising future of application in novel bioprocesses.

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