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Impact of characteristic membrane parameters on the transfer rate of ammonia in membrane contactor application

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ABSTRACT

Membrane contactors are a promising approach for ammonia removal from waste streams. The objective of this research was to evaluate the potential of common membrane parameters (bubble point, breakthrough pressure, airflow, pore size, contact angle and thickness) to predict the free ammonia mass transfer coefficient of hydrophobic membranes. 22 flat sheet membranes from different manufacturers and of various compositions were included in this study. Their main characteristics as well as the corresponding mass transfer coefficients were determined and statistically analyzed.

Mass transfer coefficients between 0.04×10^{-3} and 24.59×10^{-3} m/h were obtained. Using multilinear regression, the relative impact of each parameter on the mass transfer was identified. The developed statistical models successfully explain up to 63.2% of the observed variation. The parameters bubble point, airflow and pore size which are related to the void space of the membrane, as well as membrane thickness, determining the transport length, showed the strongest impact. In the simplest version a statistically significant model, explaining 55.3% of the variation, was obtained using only bubble point and thickness.

Obtained results can facilitate the design of suitable membranes with improved ammonia transfer and assist researchers or practitioners to choose appropriate membranes for screening tests.

1. Introduction

Membrane contactors are a technology that uses hydrophobic and microporous membranes to accomplish mass transfer between two phases without dispersion. The phases are either gas/liquid or liquid/liquid. The physical and/or chemical properties of the membrane and the permeating components allow the transport or separation of certain components. In the case of gas filled pores, in a liquid/liquid application with two polar phases, the underlying principles are firstly the mass transfer at the interface at the mouth of each membrane pore where the two phases get in contact and secondly diffusion processes inside the gas filled pore. Compared to conventional contactor methods, like for example scrubbers, there are several advantages when using membrane contactors. Besides preventing the dispersion, the following benefits are identified: no flooding at high flow rates, no unloading at low flow rates, no density difference between fluids required, high interfacial area [9]. In comparison to conventional scrubbers membrane contactors have a much larger specific surface area thus space requirements and capital costs could be reduced [14]. Membrane contactors also have disadvantages including among others the introduction of another resistance to the mass transfer, fouling or the finite life [9]. Nevertheless the mentioned advantages make this technology interesting for researchers and industries.

Studies investigating the mass transfer of volatile materials have been conducted for more than 30 years [12]. Since then different fields of applications have been explored. An extensive summary can be found in Gabelman and Hwang [9]. They report among others the use of membrane contactors for gas absorption and stripping, wastewater treatment, fermentation and enzymatic transformation or metal ion extraction. Mansourizadeh and Ismail [22] published a review on acid gas capture with hollow fibers membrane contactors in a gas/liquid operation mode.

Certain attention has also been given to the removal of ammonia from wastewaters due to the need for economical and efficient nitrogen removal solutions. Free ammonia occurring in wastewaters and liquid wastes from industry, municipalities and agriculture is a major environmental problem. It is toxic for most fish species, decreases dissolved oxygen in natural water bodies, causes corrosion and reduces disinfection efficiencies [3,10]. Several recent studies focus on modeling of operational parameters to increase the mass transfer [3,21]. Also specific applications in the field of anaerobic digestion have been investigated such as ammo-

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nia removal from anaerobic digester effluent with high content of suspended solids [28] or directly out of a digestion process treating slaughterhouse waste [18]. So far, mostly laboratory scale studies have been published and only few attempts to scale up the process have been reported [16,17,23]. Thus, despite encouraging results, the application of membrane contactors for ammonia removal is still in its beginning.

With a specific look on the performance of membrane contactors, only few studies have been published about the influence of membrane characteristics on mass transfer. Iversen et al. [14] attempted to correlate the SO_2 mass transfer coefficient with the porosity–tortuosity relationship of the porous structure in a gas/liquid application according to following formula:

$$k = \frac{D * \varepsilon}{t * T} \tag{1}$$

k, mass transfer coefficient; D, diffusion coefficient; ε, porosity; t, thickness; and T, tortuosity factor.

They claim that the mass transfer coefficient for SO_2 diffusion (Eq. (1)) can be predicted with reasonable accuracy from typical information provided by the membrane manufacturers. Nevertheless, they conclude that extra information on the pore geometry or the manufacturing method is required to estimate tortuosity.

Also Drioli et al. [7] describe that transport phenomena in membrane contactors are related to the same structural membrane properties and that these properties influence performance parameters like transmembrane flux and permeate or product characteristics. However, none of these studies considers membrane contactors in liquid/liquid applications as investigated in our study. Similar findings are reported for membrane distillation, a process relying on thermally driven difference in vapor pressure, with similar transfer mechanisms as in membrane contactor applications. Recent publications mention parameters influencing transfer performance in membrane distillation, these are among others: liquid entry pressure, membrane thickness, membrane porosity, mean pore size and membrane morphology [1,15].

The selection of good membranes, by choosing the membrane with suitable characteristics, helps to maximize the ammonia mass transfer. Performance improvement reduces the required membrane surface, module size and hence the investment costs. Therefore the development of high throughput membranes can significantly contribute to the establishment of membrane contactor processes for ammonia removal in the large scale.

This study tries to gain a better understanding of different membrane characteristics and their impacts on free ammonia mass transfer in membrane contactor applications. Moreover models are provided that help to compare different membranes and estimate their free ammonia mass transfer by knowing their most common parameters.

2. Materials and methods

2.1. Membranes

Table 1 lists 19 hydrophobic microporous membranes, one non porous membrane and two hydrophilic membranes. These membranes were selected for this study because they are available on the market. The hydrophilic membranes and the non porous membrane were included as references in this study.

2.2. Methods for membrane characterization

Different procedures were applied to characterize the most common membrane parameters namely bubble point, breakthrough pressure, thickness, airflow, contact angle and mass transfer.

Table 1
Selected membranes: trade name, manufacturer, material and pore size as given by the manufacturer.

Trade name	Manufacturer	Material	Pore size (μm)
Pallflex UTF series cast	Pall	PTFE	0.00
Emflon PTFE membrane	Pall	PTFE	0.02
Emflon PTFE membrane	Pall	PTFE	1.00
PTFE membrane filter	Sartorius	PTFE	0.45
PTFE membrane filter	Sartorius	PTFE	1.20
PTFE membrane filter	Sartorius	PTFE	5.00
Durapore	Millipore	PVDF	0.10
Durapore	Millipore	PVDF	0.22
Durapore	Millipore	PVDF	0.45
Polypropylene membrane	SterliTech	PP	0.45
Polypropylene membrane	SterliTech	PP	1.20
BTS 65	Pall	PSU	0.10
BTS 55	Pall	PSU	0.20
Versapor	Pall	AcrylicCopolymer	0.20
Versapor	Pall	AcrylicCopolymer	0.45
Versapor	Pall	AcrylicCopolymer	1.20
Tracketch	Sabeu	PET	0.40
BioTrace	Pall	PVDF	0.45
Pallflex	Pall	Glass fiber	Not available
Supor	Pall	PES	0.20
MFK	Koch membrane	PES	0.10
MFK	Koch membrane	PES	1.00

2.2.1. Bubble point

The bubble point is the minimum air pressure necessary to displace a fluid out of the largest pores of a liquid-filled membrane [7]. The determination was done according to DIN 58355-2 [6]. Deionized water was used as wetting fluid and compressed air (oil free) as test gas. Pre-wetting of the membrane to flood the pores was made with QuantachromePorofil (surface tension 16 dynes/cm). Previously, the contact angle (see also Section 2.2.5) of QuantachromePorofil was measured for each membrane to guarantee the applicability of the bubble point determination.

First, the membrane was placed into a filter holder (Fig. 1) and then wetted with the pre-wetting fluid by applying a water suction pump. After a thorough wetting with QuantachromePorofil (approximately 5 mL of liquid were pumped through the membrane), the wetting fluid was exchanged with water (another 5 mL of liquid were pumped through the membrane). The final water level above the membrane was 2 cm. Subsequently air pressure was stepwise increased onto the membrane until the first air bubble was observed. This pressure was considered as the bubble point. The experiments were done in triplicate.

2.2.2. Water breakthrough pressure

The wettability of hydrophobic membranes can be characterized by the water breakthrough pressure [20]. In liquid/liquid membrane contactor operations with two polar phases neither the feed nor the absorption solution must enter the pores. Only as long as pores of hydrophobic membranes are not wetted, the gaseous mass transfer and the separation of the liquids are ensured.

To determine the water breakthrough pressure, the filter holder for the determination of the bubble point was supplemented with an external water pressure vessel. The dry membrane was placed into the holder. The filter holder and the detector unit were filled with water. Then, the pressure in the external vessel, thus the water pressure onto the membrane, was increased until the water level in the detector unit started to rise.