

CO₂ CAPTURE BY THE ABSORPTION PROCESS IN THE MEMBRANE CONTACTORS

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Abstract. Post combustion CO₂ capture is corresponding to the most widely applicable option in terms of industrial sectors and is compatible to a retrofit strategy. In addition to the conventional chemical absorption process, membrane gas absorption is considered as one of the promising alternatives to conventional techniques for the CO₂ separation from the flue gas of fossil fuels combustion. As a hybrid approach of chemical absorption and membrane separation, it may offer a number of important features, e.g., economical viability due to its larger interfacial area, no flooding at high flow rates, no foaming and channeling, linear scale-up with predictable performance. This paper is to describe and present the state-of-the-art of the R&D efforts on membrane contactors focused on the microporous hollow fiber structure. The operating principles, liquid absorbents selection, influence of membrane wetting phenomenon, membrane materials and module types have been intensively reviewed. Model parameters including economic performance has been evaluated with comparison of other technologies. Technical obstacles of applying membrane contactors in CO₂ capture process have also been discussed. The knowledge and application gaps have been examined and identified, thus providing a recommendation for the future studies.

Keywords: CO₂ capture and storage; carbon dioxide; hollow fiber membrane; membrane contactors; numerical modeling

1 Introduction

1.1 CO₂ capture and storage and main challenges

Carbon dioxide capture and storage (CCS) has been recognized as a technically viable option for mitigating atmospheric emissions of CO₂ due to human activities, especially from large fossil fuels combustion units such as those used for electrical power generation. There have been three commercial projects linking CO₂ capture and geological storage until mid-2005: the offshore Sleipner natural gas processing project in Norway, the Weyburn Enhanced Oil Recovery project in Canada and the In Salah natural gas project in Algeria, with 1-2 Mt CO₂ capture and storage capacity per year for each project [1]. Current CO₂ capture system mainly includes post-combustion system, oxy-fuel combustion system and pre-combustion system, which make it possible to capture CO₂ from large, centralized sources like power plants and large industrial processes [2].

The main challenge for CO₂ capture systems is the large amount of energy consumption which reduces net plant efficiency significantly. It was reported that the fuel consumption increase per kWh for the plants with 90% CO₂ capture using chemical absorption technology ranges from 24-40% for new supercritical PC plants, 11-22% for NGCC plants, and 14-25% for coal-based IGCC systems compared to conventional plants without CO₂ capture [3][4]. While for CO₂ storage, the legal and policy issues are particularly troublesome. For example, who is liable for the stored carbon? What if stored carbon seeps from one jurisdiction to another? How much storage capacity exists and can these sites be secured for potentially thousands of years into the future? What if one of

these sites “burps” [1]?

1.2 Post-combustion CO₂ capture and its current status

CO₂ capture from flue gases produced by fossil fuels combustion is referred to as post-combustion capture, which is the most challenging system since a diluted, low pressure, hot and wet mixture has to be treated. However, post combustion capture is corresponding to the most widely applicable option in terms of industrial sectors and is compatible to a retrofit strategy and makes it possible to install this type of module without making major changes in the structure around the existing flue gas duct.

At present, a wide variety of techniques have been proposed to capture CO₂ from the flue gas of power station, such as chemical and physical absorption, solid adsorption, cryogenic distillation and membrane techniques [5] [6] [7]. Among these, chemical absorption is the most well established technology, but it suffers from several disadvantages such as large space, high capital cost and a variety of operational problems, e.g. liquid channeling, flooding, entrainment and foaming. Therefore, many researchers have examined the possibilities of enhancing the efficiency of these processes to reduce the effect of their drawbacks.

1.3 Why membrane contactor

Membrane contactors are a new way to accomplish separation processes like gas absorption and liquid-liquid extraction, with dramatically improved performance over conventional dispersed-phase contactors. The membrane contactors are usually hollow-fiber modules offering fast-mass transfer because of their large surface area per volume.

Zhang and Cussler [8] were the pioneers who proposed the idea of CO₂ absorption by sodium hydroxide in a hollow fiber membrane contactor. Instead of depending on the membrane selectivity, the liquid flowing in a hollow fiber contactor provides the selectivity and the unselective microporous membrane only acts as the physical interface between the liquid and gas phases without dispersing one phase into another. CO₂ is absorbed in the membrane contactor when the gas stream contacts with the liquid phase flowing on the opposite side of the membrane, while the regeneration of the absorbent is carried out by stripper using steam. The liquid phase pressure should be slightly higher than that of the gas phase to prevent dispersion of gas bubbles into the liquid.

The advantages of membrane gas-liquid contactors make them very attractive in comparison with conventional absorption equipments such as packed towers or bubble columns for offshore applications:

- Compact structure, high specific surface area and less voluminous is less energy-consuming. The amount of interfacial area in the hollow fiber modules is about 10000m²/m³, while 1000m²/m³ is reported in that of conventional absorbers [9]. Rangwala [10] has also noted a three-to-nine-fold increase in the overall mass transfer rate when using hollow fiber device over a conventional packed column. It was reported that hollow fiber contactors are about 30 times more efficient for gas absorption than conventional equipment and may reduce the size of the absorber and stripper units by 65% [11].
- The modularity of membrane modules makes the design simple and easy to be scaled up linearly with predictable performances. This is particularly valuable for application in the field of environmental technology, as it enables an efficient and economical treatment of small scale emission sources. A membrane gas absorption pilot plant with a capacity of 100m³/h has been designed and built in collaboration between TNO and industrial partners [12], which has been tested successfully for two production seasons each six months long at potato starch production plant. During the experiments no fouling of the membranes or decline in mass transfer have been observed.
- The gas and fluid phase can be manipulated independently avoiding the problems such as flooding, foaming, channeling and entrainment which are commonly encountered in conventional absorption devices.

Even though the mass-transfer coefficient of the membrane contactor is inferior to the conventional devices for

the flow of gas and liquid are normally laminar where the turbulent flow is power-consuming, the membrane gas absorption, due to its large interfacial area, is still considered as one promising alternative to conventional and potential large scale application technology for CO₂ recovery and removal [13].

Recently, gas absorption in a hollow fiber membrane contactor was identified as a viable option by the International Energy Agency's (IEA) working group on CO₂ capture [14], a hybrid process that combines the advantages of chemical absorption (high selectivity) and membranes (modularity, small size). Kaerner Oil & Gas and W.L. Gore & Associates GmbH have been developing a membrane gas absorption process for the removal of acid gases from natural gas and exhaust of the offshore gas turbines [15]. Feasibility study has also demonstrated that the CO₂ can be produced economically from flue gas on a large scale [16]. A development project is currently underway in which a pilot plant producing around 150 kg CO₂/h will be built. Successful completion of the development will then pave the way towards a demonstration on a large scale (10 ton CO₂/h) for the greenhouse [12].

2. R&D on membrane gas absorption technology

2.1. Membrane materials and module types

Different polymeric materials such as polytetrafluoroethylene (PTFE), polyethylene (PE), polypropylene (PP) and polyvinylidene fluoride (PVDF) have been used for CO₂ absorption in hollow fiber membrane contactor [17] [18] [19] [20], in which PTFE is more resistant to wetting by aqueous solutions due to its higher hydrophobicity. However, the application of PTFE is limited for its high production cost and lack of commercial availabilities. Compared with other fibers, PP fibers are less expensive and commercially available in a wider size range. Wang et al. [21] performed a theoretical simulation to study CO₂ absorption using three typical alkanolamine solutions of AMP, DEA and MDEA in PP hollow fiber membrane contactor. The effects of operating parameters, membrane configuration, module structures and different solvents on absorption flux and removal efficiency were investigated.

The membrane module should be well designed to achieve good mass transfer and can be divided into two types according to the relative flow directions of the gas and liquid phases: normally used longitudinal flow module and cross-flow module patented by TNO of the Netherlands [22]. Matsumiya et al. [23] also developed a novel facilitated transport membrane system where the feed gas and aqueous diethanolamine solutions were supplied to the lumen side of the hollow fiber ultrafiltration membrane module with an upward flow. In addition, they evaluated the energy consumption and compared with conventional separation processes. Liu et al. [24] examined the mass transfer performances using coiled hollow fiber membrane modules. Their results showed that coiled hollow fiber modules can remarkably enhance the mass transfer compared with conventional straight module.

2.2. Selection of liquid absorbents

A proper choice of the solvent is a critical and determining step in developing membrane gas absorption processes, thus reducing the capital and energy costs for flue gas treatment to separate and capture CO₂. The chemical that is to be used as a commercial absorbent for removal of CO₂ will require both a high net cyclic capacity and high solubility rate of CO₂, as well as high chemical stability, low vapor pressure and low corrosiveness. The most important requirement of solvent in membrane gas absorption is the long-term compatibility with the membrane, without damaging the membrane fibers physically or chemically.

Various liquid absorbents including pure water and aqueous solutions of NaOH, KOH, monoethanolamine (MEA), diethanolamine (DEA), 2-Amino-2-methyl- 1-propanol (AMP), N-methyldiethanolamine (MDEA), CORAL and the potassium salt of glycine and taurine were used as absorption liquids in polyethylene (PE) or polypropylene (PP) or polytetrafluoroethylene (Teflon) microporous hydrophobic hollow fiber membrane

contactors, in which the MDEA and MEA aqueous solutions in PP hollow fiber membrane contactor are the most widely used for CO₂ absorption [25] [26] [27]. Lu et al. [28] studied the effects of activators 2-Amino-2-methyl-1-propanol (AMP) and piperazine (PZ) on mass-transfer enhancement using MDEA alkanolamine solutions in a hollow fiber contactor and concluded that the mass transfer fluxes of the activated MDEA solutions are significantly higher than that of MDEA solution and effects of operation conditions on mass-transfer enhancement are limited. Theoretical simulations and corresponding experiments were carried out to describe the CO₂ absorption by distilled water and aqueous diethanolamine (DEA) solutions for better understanding of CO₂ physical and chemical absorption in a hollow fiber membrane contactor [29].

2.3. Membrane wetting

2.3.1 Influence of membrane wetting phenomenon

The membrane micropores can be theoretically filled with either gas for the hydrophobic membrane or liquid for the hydrophilic membrane. Correspondingly, there are two types of operation modes: overall-wetting mode and non-wetting mode. For membrane gas absorption system, it is essential to avoid a strong increase in mass transfer resistance in a liquid filled membrane pore compared to a gas filled pore. This means that the absorption liquid is not allowed to enter the membrane pores over prolonged period of operation time. However, in practical application, the aqueous solutions with organic absorbents can penetrate into partial pores of the hydrophobic membrane and the membrane pores are gradually wetted over long-period operation time, thus the hollow fiber membrane contactor is operated under partial-wetting mode. Wetting phenomenon of the membrane leads to the increase of overall mass transfer resistance and deterioration of membrane performance. A theoretical model was developed based on CO₂ absorption simulation by water and DEA under the wetted and the non-wetted operation modes to study the influence of membrane wetting on CO₂ capture in microporous hollow fiber membrane contactors [30]. Experimental study of CO₂ absorption in water revealed that the membrane wetting could reduce the gas absorption rate considerably [31]. Wang et al. [32] reported that the reduction of overall mass transfer coefficient may reach 20% even if the membrane pores are 5% wetted. Long-term performance tests carried out by Supakorn Atchariyawut [33] using pure water, NaOH and MEA as the absorbents revealed that the CO₂ flux continuously decreased about 43% of the initial flux during 15 days of operation due to membrane wetting. The simulation results of Zhang [34] also revealed that for the physical absorption of CO₂ by water, the proportion of membrane phase resistance in the overall mass transfer resistance increased from less than 5 to about 90% when the operation mode was shifted from non-wetted mode to wetted mode. The theoretical calculation and experimental data of Lu [35] also concluded that the wetting of membrane pores significantly affected the mass transfer coefficients of the membrane module, and resulted in the resistance of membrane phase increasing sharply and operation performance declining rapidly. Therefore, the membrane wetting has to be addressed first in order to ensure efficient absorption process and long-time stable operation.

2.3.2 Wetting mechanism and prevention

The long-term compatibility of the membranes with absorbents or the membrane wetting caused by the absorbents is a big concern for the practical application of membrane contactors. For a given liquid absorbent, the breakthrough pressure to be applied on the liquid phase to enter the membrane pores can be estimated by the Laplace equation:

$$\Delta p = \frac{4\sigma_L \cos \theta}{d_{\max}} \quad (1)$$

Where σ_L is the surface tension of the liquid; θ is the contact angle between the liquid phase and membrane; d_{\max} is the maximum membrane pore diameter. However, the membrane surface and pore morphology could be

changed in the presence of a liquid. The non-wetting intrusion of the water meniscus into some pores can result in the enlargement of the pore entrances of polypropylene membranes, which lead to a lower breakthrough pressure than the value predicted by the Laplace equation [36]. Wang et al. [37] used polypropylene microporous hollow fiber membranes that had been immersed in the DEA aqueous solutions for different days in order to investigate the effects of liquids on the membrane surface morphology and surface tension. In our previous study [38], we also examine the surface morphological changes and relevant parameters of PP membranes after being immersed in 15% and 30% MEA, MDEA with and without CO₂ loading.

Because of the significant effect of wetting on the membrane performance, the prevention of membrane wetting is very critical in maintaining the high performance of CO₂ absorption in the membrane contactor. Australia Cooperative Research Center for Greenhouse Gas Technologies [39] chemically modified the surface properties of the polypropylene to make it as water-repellent as Teflon, thus providing a cheaper alternative to higher hydrophobic PTFE with high production cost and lack of commercial availabilities. It was also found that the durability and performance of PE membrane have been greatly improved after being treated on the surface with fluorocarbonic material [17]. D. C. Nymeijer [40] applied stabilization layer at the liquid side of the membrane to prevent the membrane wetting. In addition, efforts have to be made beforehand in selecting liquids with suitable surface tension so as to reduce the tendency to seep into the pores of a hydrophobic membrane..

3. Mass transfer and numerical models

The mass transfer process consists of three consecutive steps in the three phases shown schematically in Fig. 1: (a) diffusion of the gaseous component i ($i=CO_2$) from the gas phase bulk to the membrane wall; (b) diffusion through the membrane pores to the gas-liquid interface; and (c) dissolution into the liquid absorbent accompanying by chemical or physical reaction.

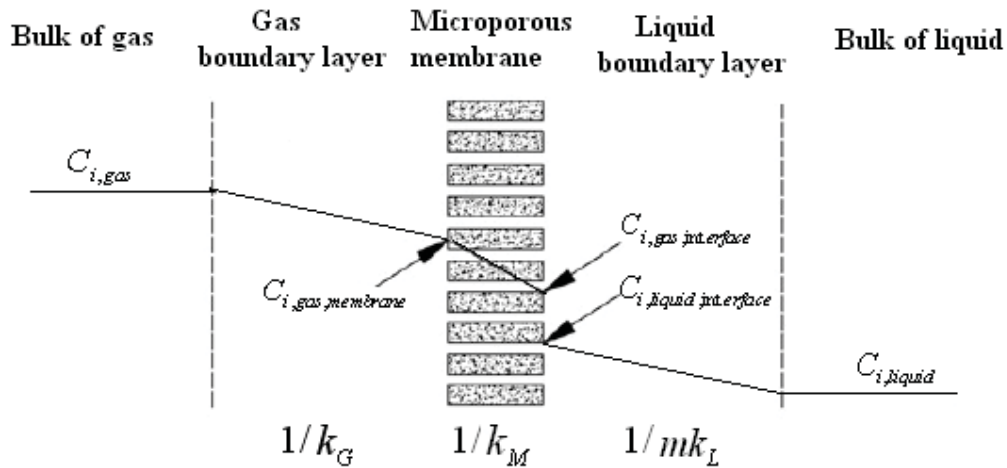


Fig.1. Mass transfer regions and dominant resistances in the membrane contactor.

Correspondingly, the overall mass transfer resistance ($1/K$) is the function of the gaseous phase boundary layer resistance ($1/k_G$), the membrane resistance ($1/k_M$) and liquid-phase boundary layer resistance ($1/mk_L$), which can be expressed as Eq. (2)

$$\frac{1}{K} = \frac{1}{k_G} + \frac{1}{k_M} + \frac{1}{mk_L} \quad (2)$$

Where K is the overall mass transfer coefficient; the k_G , k_M and k_L are the mass transfer coefficient of gas

phase, the membrane and the liquid phase, respectively; m is the distribution coefficient.

Generally, for CO₂ absorption in the hollow fiber contactor, the gaseous phase resistance can be neglected compared to the membrane resistance and liquid-phase boundary resistance [41]. Thus, the Eq. (2) can be rewritten as

$$\frac{1}{K} = \frac{1}{k_M} + \frac{1}{mk_L} \quad (3)$$

For non-wetting mode, according to the Fick law, the membrane resistance can be described as:

$$\frac{1}{k_M} = \frac{\tau\delta}{D_{i,G}\varepsilon} \quad (4)$$

While for partial wetting mode, Lu [35] developed a mathematical model of MGA by means of correlating the resistance-in-series equation, the Laplace equation and the pore size distribution function of membrane [42] based on the wetting mechanism, the membrane resistance is expressed as:

$$\frac{1}{k_M} = \frac{\eta\tau\delta}{D_{i,L}\varepsilon} + \frac{(1-\eta)\tau\delta}{D_{i,G}\varepsilon} \quad (5)$$

where τ , δ , ε , η , $D_{i,L}$ and $D_{i,G}$ denote the tortuosity of the membrane pores, thickness of the membrane wall, porosity of the hollow fiber, pore wetness, the diffusion coefficient of gas component i in the liquid and gas phase, respectively.

For chemical absorption, $k_L = Ek'_L$; E is the enhancement factor due to the chemical reaction, and k'_L is the liquid phase mass transfer coefficient for physical absorption..

At the gas-liquid interface, the mass transfer flux of component i (J_i) is given as Eq. (6):

$$\begin{aligned} J_i &= k_G(C_{i,gas} - C_{i,gas,mem}) = k_M(C_{i,gas,membrane} - C_{i,gas,interface}) \\ &= k_L(C_{i,liquid,interface} - C_{i,liquid}) = K\Delta C_i \end{aligned} \quad (6)$$

Where $C_{i,gas}$, $C_{i,gas,membrane}$, $C_{i,gas,interface}$, $C_{i,liquid,interface}$, $C_{i,liquid}$ and ΔC_i are the CO₂ concentration in the bulk of gas, at the gas and membrane interface, at the liquid and membrane interface, at the gas and liquid interface, in the bulk of liquid and logarithmic mean value, respectively.

4. Research gaps

Membrane gas absorption process is an exciting new alternative to conventional capture technologies due to its numerous advantages and has been extensively studied in recent years. However, there still is a long way to go before the technology is commercially available. To achieve further commercial applications, challenges needed to be addressed include as follows:

- (1) The membrane wetting might be a main drawback for industrial applications with the available membranes and solvents. It is necessary to improve the compatibility of the absorbents with membrane materials, so that the membrane contactors can be operated without serious performance deterioration for the long-term industrial operation;
- (2) Most papers focused on a pure CO₂ or CO₂/N₂ mixture system as the model gas streams. The effects of impurities on absorption, such as flash ash, O₂, H₂S, SO₂, and NO_x are never considered. In reality, even a small quantity of impurities may adversely impact the cost and suitability of absorption process. The solvents are

oxidized by O₂ forming organic acid. Flash ash may deposit on process equipment, block flow, and damage pumps and seals. SO₂ and NO₂ are prone to degrading the solvent, forming heat stable salts, and depleting the absorption capacity of the solvent. Therefore, it is important to investigate the effect of impurities on the contactor performance for the industrial applications;

(3) Time-dependent parameters such as diameter of membrane pores have never been introduced in the models at present. Furthermore, most of the previous reports assumed that the membrane pores are uniformly distributed or a lognormal function was always used to describe the pore size distribution. However, significant changes have been observed on the morphology of the membrane surface with the evolution of absorption due to solvent intrusion. It is necessary to develop a time-dependent mathematical model to simulate the capture process more accurately.

Anyway, there is no doubt that the gas-liquid contactors will lead to more compact, efficient and easily scaled-up units for CO₂ removal.

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