

# Application of Polymeric Membrane in CO<sub>2</sub> Capture from Post Combustion

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Received April 27, 2012; revised May 28, 2012; accepted June 8, 2012

# ABSTRACT

 $CO_2$  capture from post combustion does not need significant alteration of the current power generation facilities and is therefore of more interests to the research and industrial circles. Polymeric membrane separations, which are based mainly on physical phenomena, are easy for operation and to scale up. The details and future research trends are covered in this most updated review, which serve as an excellent technique reference for the research circle and technology evaluation for the related industrial circle.

Keywords: Polymeric Membrane; CO<sub>2</sub> Capture; Hollow Fiber Membrane; Selectivity; Membrane Wetting

# **1. Introduction**

Since the industrial revolution took place in the middle 18th century, 321 billion tons of  $CO_2$  has been released to the atmosphere and half of the  $CO_2$  emission has taken place in the recent 30 years (1971 to 2002) [1,2]. Therefore, the increasingly intensive energy requirement, which comes mainly from the fossil fuel combustion, accounts for the significant atmospheric  $CO_2$  increase and thus potentially the global climate change concerns.

While renewable energy will reduce the  $CO_2$  emission in the long term, by switching the energy resource from the current fossil fuel based toward to emerging solar, biomass, wind and tides,  $CO_2$  capture and sequestration from the large stationary coal/natural fired power plants, cement plants, oil refineries and steel plants, will reduce  $CO_2$  emission in the intermediate time span. Therefore,  $CO_2$  capture from the post combustion facilities, which does not need significant modification of the current industrial infrastructures, has aroused more research interests and industrial attentions.

The easy operation, reliable performance (based mainly on physical phenomena) and easy scale up has made polymer membrane as favorite media to capture CO<sub>2</sub> from post combustion [3]. Historically, polymer membrane has been applied successfully in ammonia synthesis/purge, petrochemical/refinery and CO<sub>2</sub> separation on industrial scale in the recent 30 years. The latter mainly includes natural sweetening, CO<sub>2</sub> recovery from land fill gas (biogas) and Enhanced Oil Recovery (EOR/natural gas processing) [4]. Therefore, the 30-year's commercial success strongly suggests polymer membrane's potential for CO<sub>2</sub> capture from flue gas [5-7].

# 2. Systematic Optimizations

Polymer membrane applied in gas separation falls mainly into three categories: 1) physically selective membrane; 2) hollow fiber membrane; and 3) facilitated transport. They have some common advantages and disadvantages. Compared to packed/tray columns, membrane operations are more flexible, economic, linear scale up, predictable, compacted and higher mass transfer rate per volume [3,8]. However, compared to the liquid adsorbent scrubbing (amine for example), membrane separation consumes more energy, has the low driving force due to the low  $CO_2$  concentration in flue gas, smaller flow rate (laminar) and higher transport resistance; in addition, the high temperature and the potential fouling membrane of flue gas are also major concerns, which need to be addressed adequately for  $CO_2$  capture [3,9,10].

To improve membrane's performance, pressure, vacuum and dual/multi stages of membrane are exerted or simulated to reach the competitive performance and minimum cost of  $CO_2$  capture [7,9-17].

## 2.1. Pressure and Vacuum

Pressure and vacuum have been introduced to membrane for  $CO_2$  capture improvement. In a modeling followed with validated lab tests, the  $CO_2$  capture efficiency of 90% was achieved with gas pressure increase [17].

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However, pressure increase has some potential drawbacks. Pressure increase could cause significant decrease of permeability and selectivity [18] and significant power loss (for example, as much as 20% of the power plant output for 5 bar compression) due to the dilute concentration of  $CO_2$  in the flue gas [7]. In comparison, under lab vacuum permeate conditions, a cost of \$54/ton CO<sub>2</sub> captured was avoided, which is equivalent to 2/3 capture cost of a pressurized bed [12]. Although popular lab scale vacuum for CO<sub>2</sub> capture with membrane is cost effective, practically, vacuum as low as 0.2 baris only available for large scale CO<sub>2</sub> capture [7]. Therefore, an adequate combination of pressure and vacuum seems to improve the membrane's performance for CO<sub>2</sub> capture and hold a potentially competitive cost benefit. The lab experimental results of pressure (1.5 bar) and vacuum (0.05 bar) combination [13] and modeling [10] are in favor of this prediction.

#### 2.2. Dual/Multistage Membrane

Dual stage membrane has been introduced to enhance driving force and increase purity of captured  $CO_2$ . Two staged pressure membrane (with 4 bar pressure at the first stage) was modeled and a cost of \$57/ton  $CO_2$  captured was avoided with final  $CO_2$  concentration of 90% [18,19]. Two staged vacuum membrane was simulated to achieve 90% capture efficiency at a cost of \$39/ton  $CO_2$ captured [7]. However, since multi staged membrane consumes more energy and its facility cost is more than those of a single stage one [15], a compromise between the stage number of membranes and the capture efficiency has to be taken into consideration.

#### 2.3. Membrane Length and Module Number

Similar to vacuum, pressure and multi stage factors, optimization of membrane length and number of membrane module can also contribute to high selectivity. Like the scenario in distillation tower, making membrane longer can only improve  $CO_2$  capture when the membrane module is shorter than the maximum effective module length  $L_{eff}$  [20]. Therefore, there is a balance between number of parallel modules (more of the membrane areas with higher driving force than the according length increase however with more cost of module manufacture) and the according cost [21].

#### 2.4. Scale up and Capture Cost

Polymer membrane for  $CO_2$  capture is still in lab scale, since rare pilot tests have been reported and evaluated. Pilot scale of membrane operation for capturing  $SO_2$ from flue gas has been reported to run smoothly six months with no problems from  $CO_2$ ,  $NO_x$  and dust [22]. A screening of suitable polymer materials for pilot scale set up and no further progress is followed yet [23]. Although energy cost for CO<sub>2</sub> capture of polymer membrane is estimated to be \$23/ton CO<sub>2</sub> with 90% capture efficiency from modeling [7], the other researchers have insisted that membrane is much more expensive than chemical absorption, therefore, it still has a long way to go for commercialization, despite its promising future for CO<sub>2</sub> capture [3,9,19].

## 3. Physically Selective Membranes

Ideally, a highly selective  $CO_2/N_2$  and permeable membrane works well for post combustion  $CO_2$  capture; however, there is always a trade-off between selectivity and permeability [10,15]. The highly selective membrane is comparable to a filter and it only allows filtrate to go through, which can be enhanced by a sweeping media as a "vacuum".

There are still some arguments about the appropriate selectivity of  $CO_2/N_2$  in flue gas capture. An earlier argument stated that a  $CO_2/N_2$  selectivity should be more than 200 [6] or higher than 100 with  $CO_2\% > 20\%$  [24]; therefore, the current level of 50 is too low for the post  $CO_2$  capture from flue gas (about 13%  $CO_2$ ) [9,11,24,25]. Other researchers mentioned that with the  $CO_2/N_2 = 50$ , capture efficiency of 90% and cost of \$23/ton  $CO_2$  can be achieved with combined membrane technologies [7]. Because the above statements are all based on modeling and parametric study, more experimental verification, including lab and pilot scales, will be crucial to evaluate  $CO_2/N_2$  selectivity from flue gas for the most promising polymeric membranes.

#### 4. Hollow Fiber Membranes

Hollow fiber membrane is highly gas permeable but not gas selective by itself. The gas mixtures on one side of membrane permeate into the lumen of the membrane, and the solvent selectively dissolve or reacts with and remove  $CO_2$  from the flue gas [9,11,26]. Therefore, the selectivity role in this membrane separation is realized through the solvent. The illustration and SEM picture of Hollow fibre membrane are illustrated below, as in **Figures 1** and **2** [27,28].

#### 4.1. Absorbents

Ideal absorbents for hollow fiber membrane should preferably have 1) high reactivity with  $CO_2$  (reduction of  $CO_2$  transport resistance in the solvents); 2) are highly hydrophobic and high surface tension (minimization of the liquid penetration into the fiber lumen significantly reduces the mass transfer in liquid solvent); 3) low vapor pressure (reduction of solvent evaporation into the flue



Flue gas inlet

Figure 1. Illustration of Hollow fibre membrane (Yan *et al.* 2007).

gas mixtures and thus increase in the gas transport efficiency); 4) chemical compatibility with membrane; and 5) easy regeneration (low energy consumption) [3].

Modeling result has indicated that aqueous diethanolamine (DEA) is faster than water to dissolve  $CO_2$  since the mass transfer of  $CO_2$  in the aqueous phase is dominating the transfer resistance, is thus the overall control step [20]. The application of amino salts as solvents of hollow fiber membrane for  $CO_2$  capture proved to be successful in both ways: 1) enhancing the hydrophobic property and thus resulting in high mass transfer efficiency; and 2) reduction of amine corrosion concerns [29-31].

Furthermore, the interaction hydrophobic property, the non wetting of the membrane material with the aqueous solution, can minimize the transport resistance of  $CO_2$  in the lumen of hollow fiber [3,10].

## 4.2. Wettability

Membrane wetting has significantly increased the mass transfer resistance, as can be seen in Equation (1). Modeling and experimental results indicate that wetting of



Figure 2. SEM picture of Hollow fibre membrane (Mansourizadeh and Ismail 2010).

membrane pores significantly affects the mass transfer coefficients of the membrane module [32,33]. Simulation indicates that  $CO_2$  absorption rate six time slower when membrane operated in wetting than unwetting mode; even 5% wetting of the inner membrane leads to 20% reduction of the overall mass transfer coefficient [17]. This was validated by the following experiments that overall mass transfer is reduced to 20% of the control (no wetting) because of the wetting [17].

$$\Delta P = -\frac{4\sigma_L \cos\theta}{d_{\max}} \tag{1}$$

 $\Delta P$  minimum permeance pressure kPa;

 $\sigma_{\rm L}$  surface tension mN/m;

 $\theta$  contact angel between liquid surface and membrane; d<sub>max</sub> the maximum porosity diameter m.

Membrane wetting can result from the membrane materials, the liquid absorbents and possibly its interaction between materials and liquid absorbents. The membrane materials should be hydrophobic, have small pores and possibly thick membrane wall difficult for absorbents to penetrate through [3]. A queous water (maybe with inorganic salts) easily penetrates into many materials, thus leading to the dominant organic adsorbents to react with  $CO_2$  as first priority. The optimization of polymer concentration (polyetherimide) can lead to smaller pore size and effective porosity increased the  $CO_2$  adsorption and less wetting [8]. Low molecules additives can also lead to small pore size, high surface porosity and thus high wetting resistance [34].

However, the interaction between the absorbents and membrane becomes complicated, which needs to be carefully addressed so as to minimize the membrane wetting. Three months' immersion of PP in ionized water, MEA and MDA showed that diffusion of the absorbents molecules cause PP membrane swelling and thus a strong hydrophobicity of the inner membrane wall is recommended [35]. Other researchers have also attributed membrane wetting either to the chemical reaction between the absorbents or membrane materials [17]. A degradation of low density polyethylene with 8 - 45 days of immersion in MEA solution has been identified, which was assumed to be the result of oxidative degradation [36]. In parallel, plasticizing effect of CO<sub>2</sub>'s interaction with the polymer materials was also suggested for the degradation [23]. Screening of four polymer materials (PP, PVDF, PTFE and Nylon based) for pilot scale membrane operation has been based on the criteria of chemical aging with MEA [37].

Hydrodynamics and flow conditions could also possibly result in membrane wetting. While high flow pressure could be one reason for membrane wetting from theoretical calculations of Laplace equation [3], modeling and experiments results from complete, partial and non wetting of PP with MEA, water and NaOH solution illustrated that high flow rate could easily lead to membrane wetting [38].

## 5. Cross Linking (Facilitated Transport) Membrane

From the comparison that DEA can speed up  $CO_2$  capture through chemical absorption than purified water in hollow fiber membrane [20], it is expected that coupling of  $CO_2$  affinity coatings/cross linked materials (for example, organo amines) at the inner wall of membrane, which is compared to the catalyst in the chemical reaction, will improve the  $CO_2$  selectivity in membrane and thus speed the subsequent  $CO_2$  mass transport.

The facilitated transport membrane has been successfully patented for its promising application in CO<sub>2</sub> capture [39]. Based on this process, the membrane has a support coated and cross linked polyvinilamine, which serves as a fixed carrier of CO<sub>2</sub> and a "catalyst" (in the form of  $HCO_3^-$  with combination of moisture), helps quickly remove and transport CO<sub>2</sub> through the membrane lumen. It is reported through the improvement of this process, CO<sub>2</sub>/N<sub>2</sub> selectivity has approached 174 and 200 by the same research group [40,41].

Other researchers have also come up with the similar approaches of cross linking. Examples of the chemical structure of carriers and the related reactions are as shown in **Figure 3** and Equations (2)-(4). Amines in cross linked poly (vinyl alcohol) of polymeric membrane reached  $CO_2/N_2$  selectivity of 450 and the membrane has a descent performance even at the temperature range of  $100^{\circ}C$  -  $170^{\circ}C$ , much higher than the current prevalent working temperature of polymer membrane [18]. Highly hydrophilic compounds containing quaternary ammonium moieties attached to the reactive trimethoxysilane have  $CO_2/N_2$  selectivity up to 1500 and the permeability



Figure 3. Chemical structure of crossing linking media in facilitated transported membrane for CO<sub>2</sub> capture [18].

increased by 35 fold in moisture than that of dry mode without  $CO_2/N_2$  loss [42].

$$R-NH_2 + CO_2 + H_2O = R-NH_3 + HCO_3^-$$
 (2)

$$2R-NH_2 + CO_2 = R-NH-COO^- + R-NH_3^+$$
(3)

$$CO_3^{2-} + CO_2 + H_2O = 2HCO_3^{-}$$
 (4)

Modeling results indicate that facilitated transport membranes could capture  $CO_2$  efficiently even in the concentration of as low as 10% in flue gas, with 90% efficiency, 90%  $CO_2$  purity with competitive cost to that of aqueous amine scrubbing [41].

## 6. Conclusions

Polymer membrane separation process is simple, easy to scale up; therefore, it has a huge potential in the postcombustion capture of carbon dioxide applications. Tree typical polymer membrane, physical-selective membrane, hollow fiber membrane and facilitated transport membrane, as well as the operation optimization of pressure, vacuum and multi stag, have been comprehensively discussed in depth in this paper. The major conclusions and suggestions can be summarized below:

1) Pressure and vacuum combination will accelerate the mass transfer rate, reasonable cost, and thus more suitable for the polymer membrane separation of carbon dioxide;

2) Increase in membrane length and parallel membrane devices will increase the separation efficiency, but to balance thus increasing the cost of equipment;

3) Physical selective membrane due to the low selectivity and separation efficiency to reach the burning need for separation of carbon dioxide;

4) The selectivity of the hollow fiber membrane greatly increased, but its chemical adsorbent will increase the secondary pollution and increase the separation costs, a careful analysis of the factors to be studied later;

5) Auxiliary coating/cross-connecting material film should be in the future vigorously research, high selectivity and high mass transfer rates and may withstand  $170^{\circ}$ C high temperature, can adapt to the needs of the flue gas of large-scale separation of carbon dioxide.

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