



Water membrane for carbon dioxide separation

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ABSTRACT

Carbon dioxide separation has drawn much interest because of the concern over global warming. A simple and yet effective way of removing CO₂ is highly desirable for the separation involving combustion effluents, consisting mainly of CO₂ and N₂, hydrogen production involving CO₂ and H₂ mixtures, and natural gas sweetening involving mainly CO₂ and CH₄. Here, we present water membrane for CO₂ separation. The water membrane is simply a water layer formed on a hydrophobic support membrane to prevent the water from permeating through the support layer. The membrane exploits the fact that the solubility of CO₂ in water is almost two orders of magnitude higher than the solubility of the gases of N₂, H₂, and CH₄. Both the permeance and the selectivity increases with decreasing water layer thickness, contrasting the traditional tradeoff between permeance and selectivity. The carbon dioxide selectivity with respect to nitrogen, methane, and hydrogen is 86, 66, and 74, respectively. The permeance of 1.1 GPU reached with 5 mm thick water layer should improve in due time with further work on reducing the thickness.

1. Introduction

Technologies for removing carbon dioxide from effluent gases have drawn much interest because of global warming. Many processes based on diffusivity, solubility, and reactivity have been developed to separate CO₂ from the effluent gases [1,2]. In the case of separation based on diffusivity and reactivity in membrane, indiscriminate permeation of gas due to defects in the specimen often occurs, making it difficult to fabricate the specimen thinner and larger [3]. Therefore, the gas separation technique based on the difference in solubility of gases in a specific solvent is widely used industrially because it is easy to produce a continuous and defect-free separation layer.

Common industrial methods for CO₂ separation use a specific solvent as an absorption material. This mechanism utilizes the principle that gases have different solubility in the identical solvent. In spite of high selective transport properties to target gas molecule, low energy efficiency and liquid loss in desorption are regarded as disadvantages of this mechanism [2,4].

In this context, facilitated transport of a specific gas through polymeric membranes or liquid membranes containing mobile carriers can be employed to overcome the limits of the conventional absorption process, as in ionic liquid membranes. This approach has the advantages of having high selectivity, diffusivity, low loss of solvent and carrier, and low performance degradation [5–7]. But it requires a

polymeric membrane that possesses a small effective thickness and also demands a specific liquid that is compatible with the polymer material [8]. Water mixed with a surfactant and a stabilizer was used in the form of ‘dynamic liquid membrane’ [9] recently for separation performance of CO₂/CH₄ mixture. An air flow was used to facilitate the evolution of CO₂ dissolved in the water film. While the process would be useful for purification of methane gas, mixing the separated CO₂ with air defeats the very purpose of capturing and sequestering CO₂ pertinent in CO₂ separation.

In this work, we explore water membrane for gas separation, mainly for carbon dioxide separation. The separation cell is a dead-end system consisting of a water layer supported by a hydrophobic solid support membrane with a relatively large pore size. As schematically illustrated in Fig. 1(a), a pressurized gas or gas mixture is fed into the space above the water layer in the separation cell. The pressurized gas dissolves into the water layer and then evolves from the bottom side of the water layer that is in contact with the solid support membrane. The bottom of the solid membrane is exposed to atmospheric pressure. As shown in Fig. 1(b), the contact angle of water on the solid membrane we used, which is polytetrafluorethylene (PTFE), is larger than 140°. The PTFE membrane is known to have many favorable qualities such as excellent chemical resistance, good thermal stability and low surface energy [10]. The hydrophobic nature of the solid membrane prevents water from permeating into the pores of the membrane, provided the pressure

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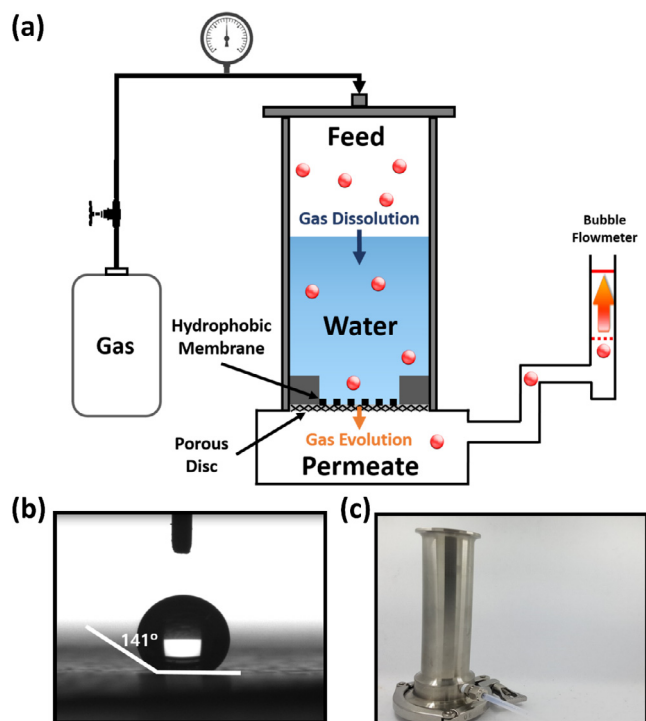


Fig. 1. Gas separation with water membrane. (a) Schematic illustration of gas separation with water membrane. (b) Contact angle of water on polytetrafluoroethylene (PTFE) that was used for hydrophobic support layer. (c) Digital camera image of stainless steel holder for experiment.

applied does not exceed a certain level. Under the conditions, the solid membrane is devoid of water. The actual separation cell we used is shown in Fig. 1(c), consisting mainly of stainless steel membrane holder, water, and commercial PTFE for porous support membrane. No mobile carriers are involved.

The motivation for exploring the water membrane stems from the fact that the solubility of carbon dioxide in water or equivalently, Henry's constant, is much larger than that of other gases of interest such as N_2 , CH_4 , H_2 and O_2 . Therefore, the large difference in the solubility can be exploited for CO_2 separation. Furthermore, water is not only cheaper but safer than other chemical solvents due to its abundance and non-toxicity. We first examine the hypotheses made for the water membrane. We then explore in detail CO_2/N_2 pair for the permeance and selectivity attainable with the water membrane because of its relevance to carbon dioxide removal from flue effluents, which accounts for 40% of all CO_2 emissions [11], followed by investigation into the other gases.

2. Experimental section

2.1. Materials

Hydrophobic polytetrafluoroethylene (PTFE) disc membranes (47 mm diameter, 0.45 μm and 0.22 μm opening) were purchased from Millipore (FHLPO4700 and FGLPO4700, USA) and used as received. A PTFE membrane with small opening size (0.22 μm) was used for the investigation on the effect of pressure. All carbon dioxide, methane, nitrogen, oxygen and hydrogen with a purity of 99% or more were procured from Daehan Special Gas CO. Membrane cells for gas permeation experiments were custom-made using stainless steel (sus-316). A bubble flow meter (10 mL volume, Standard Version) was purchased from Sigma-Aldrich. Sodium chloride (NaCl, molecular weight of 58.44 g/mol) for making salt water buffer solution (pH 4, 10, respectively) for acidic and basic water tests were purchased from

Daejungchem.

2.2. Gas transportation measurement

The experimental setup for measuring gas permeance is shown in Fig. S1. For the dead-end experiments, a PTFE filter membrane, 47 mm in diameter (Fig. S1(a)), is loaded into the customized cell that is supported by stainless steel porous disc and then the cell is filled with water. Water does not permeate through the PTFE filter because of its hydrophobicity and thus a water layer forms on the filter membrane upon filling the cell with water. As explained later, a thinner water layer yields a higher permeance. However, the minimum amount of water was 4 mL on the 47 mm diameter filter membrane that can sustain a water layer with a continuous water surface without forming a glob (Fig. S2). In this experimental setup, each gas bomb with its own valve is connected to a line into the membrane cell and the gauge pressure of the gas entering the cell was fixed using a flow meter (Fig. S1(b)). The gas that permeates through the membrane cell enters the bubble flow meter through the pipeline, and the bubble moves upward. The permeance is calculated from the distance of the bubble covers in a given time. Due to the bubble travels very slowly, we measure the displacement of bubble per 20 min to estimate the gas permeance [12]. We use GPU as the unit of permeance ($1 \text{ GPU} = 1 \times 10^{-6} \text{ cm}^3/\text{cm}^2/\text{s}/\text{cmHg}$ at STP) [13]. The selectivity is simply the ratio of the permeances of two gases.

2.3. Effects of water variables on permeation

Experiments were conducted mainly with pure water. For the investigation of pH effects on the permeation, acidic and basic buffers were used. A buffer solution of pH 4 and a buffer solution of pH 10 were tested. For salinity effects, sodium chloride powders were added to deionized (DI) water to have a salt content of 3.5 wt%, which is equivalent to the salinity of seawater. Temperature effects were also investigated using a low temperature circulation bath (AND Korea, AD-RC08).

3. Results and discussion

3.1. Hypotheses validation

One major hypothesis regarding the water membrane is that the water in the cell would not permeate through the PTFE filter membrane when a pressure is applied on the water layer. The water pressure resistance of a porous medium is expressed by the following Young-Laplace equation:

$$h = \frac{2\gamma \cos\theta}{\rho g R} \quad (1)$$

where γ , θ , ρ , g , R and h are the surface tension, the contact angle, the density of water, the gravitational acceleration, the pore radius, and the water head, respectively [14]. With the properties of water, the water head h for the effective PTFE pore radius of 0.45 μm was calculated to be 51.27 m. Thus, the PTFE filter can hold roughly 4.95 bar gauge pressure since a water head of 10.35 m corresponds to 1 bar. The pore size of filter, however, is an average value and therefore, a gauge pressure of 1 bar is applied to most experiments in this work to guard against the presence of larger pores. To further confirm the theoretical result, the weight of a beaker placed at the permeate end of the cell was measured for its change with time at 1 bar gauge pressure. The test condition of the experiment is exactly the same as that of gas permeation experiments. As shown in Fig. S3(a), no change in the weight was observed at all over a period of 20 h. This result confirms that there was no permeation of water. Meanwhile, the contact angle influenced by the surface of PTFE filter can be changed over time due to the hydrophobicity loss of the filter. We checked that the filter maintains its

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