

# Gas Permeation Through Water-Swollen Polysaccharide/Poly(vinyl alcohol) Membranes

Min-Joung Kim,<sup>1</sup> You-In Park,<sup>1</sup> Kyung-Ho Youm,<sup>2</sup> Kew-Ho Lee<sup>1</sup>

<sup>1</sup>Membranes and Separation Research Center, Korea Research Institute of Chemical Technology, P.O. Box 107, Yusong, Taejeon 305-606, Korea

<sup>2</sup>School of Chemical Engineering, Chungbuk National University, Cheongju 361-763, Chungbuk, Korea

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**ABSTRACT:** The effect of Na-alginate content on the gas permeation properties of water-swollen membranes prepared by varying Na-alginate and poly(vinyl alcohol) (PVA) content in membranes was investigated. The influences of water content and crystallinity of the membranes on the gas permeation performance of the water-swollen membranes were studied. The gas permeation rate and selectivity of Na-alginate/PVA water-swollen membranes were compared with those of the dry membranes. The permeation rates of nitrogen and carbon dioxide through water-swollen membranes were in the range of  $0.4\text{--}7.6 \times 10^{-7}$  to  $3.7\text{--}8.5 \times 10^{-6}$  cm<sup>3</sup> (STP)/cm<sup>2</sup> s<sup>-1</sup> cmHg<sup>-1</sup>, which were 10,000 times higher than those of dry-state membranes. The permeation rates of mixture gases through water-swollen Na-

alginate/PVA membranes were found to increase exponentially with the increase of Na-alginate content, whereas carbon dioxide concentration in permeates was decreased linearly. It was found that the gas permeance of the water-swollen membranes increased with increasing the Na-alginate content in the membrane. Gas permeation rates of the water-swollen Na-alginate/PVA membranes increased with increasing the water content in the membrane and decreasing the crystallinity of the membrane. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 3225–3232, 2004

**Key words:** polysaccharide; Na-alginate; poly(vinyl alcohol) (PVA); gas permeation; membranes

## INTRODUCTION

Environmental issues in recent years have resulted in increased efforts in the industrial world to develop clean and energy-efficient separation processes. Traditionally, well-developed technologies like cryogenic distillation, pressure-swing adsorption, and absorption have not always been economically attractive in industrial applications where capital cost, space, simplicity of operation, and power consumption are of concern. Membrane systems for the separation of gases are potentially attractive because they offer low capital and operating cost, along with low energy consumption.<sup>1</sup>

Recently separation and recovery of carbon dioxide have been investigated with respect to the global warming problem. For the separation of carbon dioxide using a membrane, the supported liquid membranes (SLMs) containing a mobile carrier in membrane pores have been studied because of their high selectivity toward carbon dioxide. Although remarkably high selectivity for SLMs have been reported, the disadvantage of the membrane degradation attributed

to the evaporation of the membrane solution and/or the “wash-out” of the carrier was well recognized.<sup>2,3</sup>

One of the methods for preventing this degradation is the use of a gel-type swollen membrane. The gel-type swollen membrane that is intermediate between liquid and solid phase can be made by swelling a polymer film in a solvent. If the solvent used to swell the polymer film is a good physical solvent for the gas of interest, the solvent-swollen membrane can be used as a membrane for gas separation. Yasuda et al.<sup>4</sup> reported the permeation of various gases through water-swollen membranes and proposed an interpretation of the process as dissolution of gas in the water phase, and diffusion of the dissolved gas through the phase. Ito et al.<sup>5</sup> studied the permeability of N<sub>2</sub>, CO<sub>2</sub>, and CH<sub>4</sub> through a wet chitosan membrane and found the influence of the membrane preparation conditions and operating conditions on the separation performance of the wet chitosan membrane. Matsuyama et al.<sup>6</sup> prepared a water-swollen cation-exchange membrane by grafting acrylic acid onto a microporous polypropylene membrane by a plasma-grafted polymerization technique. They studied the facilitated transport separation of CO<sub>2</sub> in a water-swollen cation-exchange membrane, containing ethylenediamine as a carrier. A water-swollen hydrogel membrane for gas separation was recently developed in our laboratory by using poly(vinyl alcohol) (PVA). This technique

Correspondence to: K.-H. Lee (khlee@pado.kriict.re.kr).

was successfully applied to the carbon dioxide separation over a long period of operation.<sup>7</sup>

We selected PVA as a membrane material to prepare a water-swollen membrane for carbon dioxide separation. PVA is a very well known material to have good mechanical property and hydrophilicity. The polyelectrolyte used in this study was Na-alginate, which is a linear polysaccharide of (1-4)linked  $\beta$ -D-mannuronate and  $\alpha$ -L-guluronate residues arranged in a nonregular blockwise pattern along the linear chain. This polysaccharide, abundantly supplied from marine algae, was reevaluated as an attractive natural resource, possessing high water swelling property. The highly water swollen membrane has an advantage, in that a high permeation rate is obtained because mobility of a gas through the membrane is high. In this study, water-swollen polysaccharide/PVA membranes were prepared to improve gas permeation rates of PVA hydrogel membranes. The separation performance of CO<sub>2</sub> over N<sub>2</sub> in water-swollen polysaccharide/PVA membranes with different Na-alginate contents was investigated. The effect of Na-alginate composition on the gas separation performance of the water-swollen membranes was discussed. The influences of the water content and the crystallinity of the membranes on the separation performance of the water-swollen membranes were investigated.

## EXPERIMENTAL

### Materials

To prepare water-swollen hydrogel membrane, Na-alginate and PVA were used as membrane materials. The Na-alginate [number-average molecular weight ( $M_n$ )  $\sim$  500,000] used in this research was purchased from Showa Co. (Tokyo, Japan) and PVA ( $M_n$   $\sim$  50,000) was purchased from Aldrich Chemical Co. (St. Louis, MO). HCl (35%), isopropylalcohol (IPA), and glutaraldehyde (GA, 25% in water) as a crosslinking agent were obtained from Junsei Chemical (Tokyo, Japan) and used without further purification. The deionized water was used as a solvent. UOP polysulfone (PSf) UF membranes [molecular weight cutoff (MWCO)  $\sim$  30,000 g/mol; Fluid System, San Diego, CA] were used as supports for the formation of Na-alginate/PVA composite membranes.

### Preparation of water-swollen membranes

Aqueous solutions of Na-alginate and PVA were prepared separately at a concentration of 2.5 wt %. The powder materials were dissolved in deionized water at room temperature ( $\sim$  25°C) for Na-alginate and at 90°C for PVA, with continuous stirring. The two polymer solutions were mixed in the desired proportions (Na-alginate : PVA = 0 : 100, 2.5 : 97.5, 5 : 95, 7.5 : 92.5,

10 : 90 wt %) at room temperature and the GA solution was added to mixture solutions with Na-alginate/PVA mixture solution : GA solution = 90 : 1 weight ratio and then stirred for 24 h. The resulting solutions were optically clear and showed neither visible separation into bilayers nor any precipitation on standing. Water-swollen membranes were prepared by dip-coating of PSf UF membrane, which was fixed onto the glass plates using tapes, with the Na-alginate/PVA/GA mixture solutions, followed by drying at room temperature for 24 h, and then the dry nascent membranes were crosslinked by immersion for 4 h at 40°C in crosslinking solution (IPA : water : HCl = 90 : 10 : 1 weight ratio). In all cases, after the crosslinking reaction, the membranes were removed from the reaction solution and washed several times with pure water. The crosslinked Na-alginate/PVA membrane, which was in the form of alginic acid, was subject to an alkaline treatment so that the acid groups ( $-\text{COOH}$ ) in alginic acid were converted into the alginate form ( $-\text{COONa}$ ). The alkaline solution was 0.1M sodium hydroxide in pure water. After alkaline treatment for 24 h, the membrane was thoroughly rinsed with deionized water. The thickness of the coating layers of the membranes was 0.2–1  $\mu\text{m}$ .

### Characterization of membranes

Swelling behavior was studied by the general gravimetric method. Dry crosslinked Na-alginate/PVA films were immersed in distilled water thermostat at 25°C for 5 days to allow the films to reach equilibrium sorption. The film dimension was 2  $\times$  2 cm. After measuring the swollen weight, film was dried for 48 h at room temperature under a vacuum and the dry weight was measured. Equilibrium water swelling ratio was calculated according to the following expression:

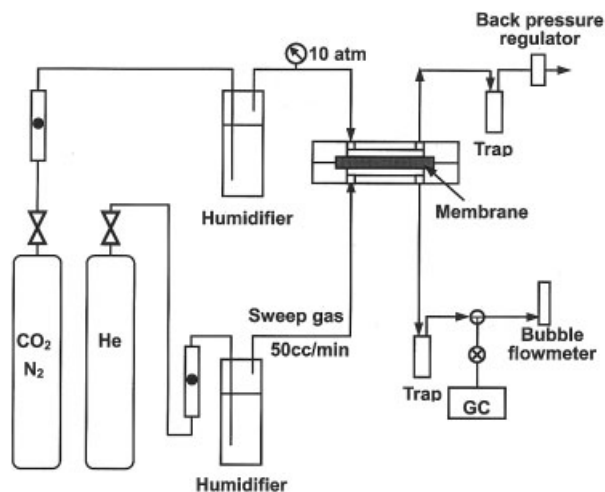
$$\text{Water swelling index (\%)} = \frac{W_s - W_D}{W_D} \times 100 \quad (1)$$

where  $W_s$  and  $W_D$  denote the weights of swollen and dry samples, respectively.

To investigate the crystallinity of Na-alginate/PVA water-swollen membranes, a wide-angle X-ray diffractometer (WAXD, Diano XRD 8200) was used. WAXD patterns were recorded by the reflection method with nickel-filtered Cu-K $\alpha$  radiation operated at 50 kV, 180 mA in the 2 $\theta$  scanning mode between 5 and 50°. The degree of crystallinity was calculated according to the usual method.<sup>8</sup>

### Permeation measurement

The apparatus used in this study is shown in Figure 1. The apparatus, consisting of a water bubble chamber,



**Figure 1** Experimental setup for gas permeation rate and selectivity measurements.

was attached to a permeation cell, a gas vessel, a pressure gauge, an autoinjector, a gas chromatograph (GC), and a temperature-controlling oven as the main elements. The effective area of the permeation cell was  $9.6 \text{ cm}^2$ , and the cell was set in a water bath the temperature of which was controlled to within  $\pm 0.5^\circ\text{C}$ . Water vapor in the permeation side was collected in a cold trap. The permeation rates of pure gases and ideal separation factor ( $\alpha$ ) through the swollen membrane were measured by a soap film flow meter and calculated by using the following equations:

$$P = Q / (p_1 - p_2)At \quad (2)$$

$$\alpha_{\text{ideal}} = \frac{P_{\text{CO}_2}}{P_{\text{N}_2}} \quad (3)$$

where  $Q$  is the permeation volume of gas [ $\text{cm}^3$  (STP)];  $t$  is the permeation time (s);  $A$  is the effective membrane area ( $\text{cm}^2$ ) for gas permeation; and  $p_1$  and  $p_2$  are feed side and permeate side pressures (cmHg), respectively. In addition,  $P_{\text{CO}_2}$  and  $P_{\text{N}_2}$  are the permeation rates of carbon dioxide and nitrogen, respectively. The permeation experiments were carried out at  $25^\circ\text{C}$  and 10 atm until steady state was reached. The gas permeation data of the dry membrane were obtained using a high-vacuum method, as described in previous articles.<sup>9,10</sup> The mixed gas permeation experiments involved a startup similar to that of the single gas permeation experiments. The total permeation rates of mixture gas and compositions of the permeates were determined using a GC (DS6200; Donam Development Co.). The measurements were carried out with a mixture ( $\text{CO}_2/\text{N}_2 = 20/80$ ) of  $\text{CO}_2$  and  $\text{N}_2$  gas at  $25^\circ\text{C}$  while pressure on the feed side was 10 atm and the

sweep side pressure of the helium stream was atmospheric. After steady state was reached, the  $\text{CO}_2$  selectivity was calculated according to the ratio of gas composition at the feed side and the permeate side. The sweep gas was helium and the flow rate was  $50 \text{ cm}^3/\text{min}$ . The outlet sweep gas was analyzed by a gas chromatograph equipped with a thermal conductivity detector.

## RESULTS AND DISCUSSION

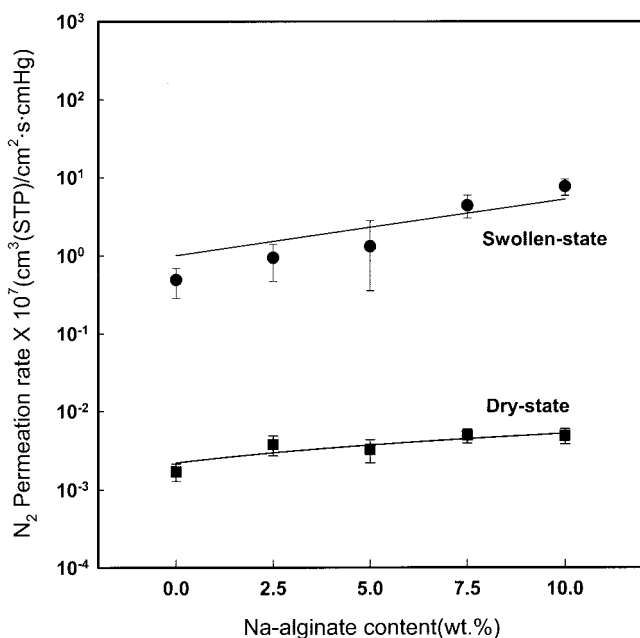
### Characterization of membranes

For the study on the relative crosslinking degree of the membranes, swelling indices of the crosslinked Na-alginate/PVA membranes with different Na-alginate contents were obtained, as listed in Table I. The swelling index was calculated by using eq. (1). The PVA membrane showed a swelling index of 23.8% after crosslinking for 4 h. The water swelling indices of Na-alginate/PVA membranes increased from 23.8 to 54.3% with increasing Na-alginate content.

In this study, because the crosslinking of the membranes was carried out in the isopropanol–water mixture containing HCl as a catalyst, the crosslinking reaction started with diffusion of proton ( $\text{H}^+$ ) with water molecules into the membranes. Therefore, the two reactions, crosslinking and swelling, were occurring at the same time, as described in previous work.<sup>11</sup> The swelling reaction induced the increase of intermolecular distances of the membrane material. In the case of Na-alginate/PVA membranes, because the water sorption amount of the Na-alginate is so much higher than that of PVA, the intermolecular distances of the membrane increase with increasing Na-alginate content. The longer intermolecular distance in the swollen state was disadvantageous for the crosslinking reaction. As a result, the swelling indices of the membranes crosslinked for the same GA concentration were increased with Na-alginate content in the membranes. The swelling indices of the prepared membranes increased with increasing Na-alginate content, indicating that the relative crosslinking degree decreased. This result confirms that the water swelling

**TABLE I**  
The Physical Properties of Water-swollen Na-Alginate/PVA Membranes

Na-alginate content in membrane (wt %)	Water swelling ratio (%)	Crystallinity (% swollen state)
0.0	23.80	11.38
2.5	30.00	7.47
5.0	43.60	6.54
7.5	50.73	5.08
10.0	54.33	3.50



**Figure 2** N<sub>2</sub> permeation rates of water-swollen Na-alginate/PVA membranes as a function of Na-alginate content in membrane ( $T = 25^{\circ}\text{C}$ ,  $P = 10$  atm).

indices of membranes are affected by Na-alginate content in the swollen membranes.

The crystallinity of swollen Na-alginate/PVA membranes was evaluated from wide-angle X-ray diffraction measurements (WAXS) and the results are presented in Table I. The crystallinity of the swollen membranes decreased from 11.38 to 3.5% with increasing Na-alginate content from 0 to 10 wt %. The crystallinity of the membrane is associated with the Na-alginate content because Na-alginate used in this study is an amorphous polymer. The crystalline structures of Na-alginate/PVA membranes become amorphous with increasing amorphous Na-alginate in the membranes. Also our crosslinking procedure involved a swelling step that led to partial disruption of the crystalline order existing in the semicrystalline PVA region. From this result, it was seen that the overall crystallinity of the membranes decreased with increasing Na-alginate amount.

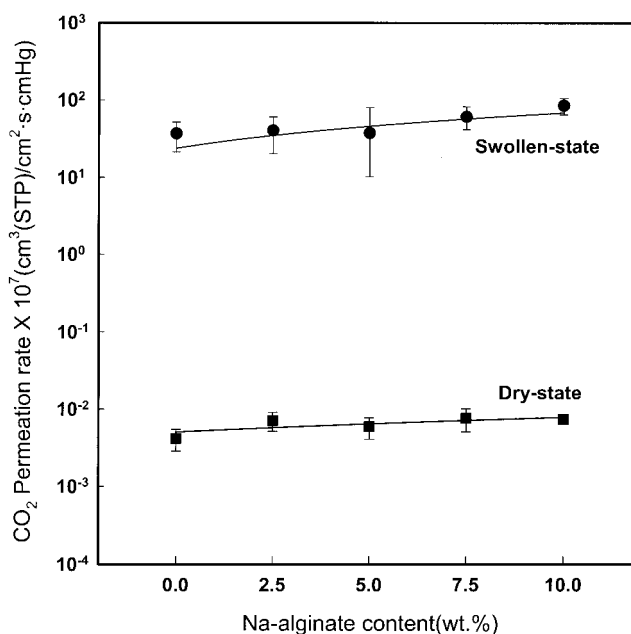
### Single gas permeation

For the separation of carbon dioxide from a carbon dioxide/nitrogen mixture, water-swollen Na-alginate/PVA composite membranes were prepared by a dip-coating method with UOP UF membranes (MWCO = 30,000 g/mol) as a support as explained in the experimental section.

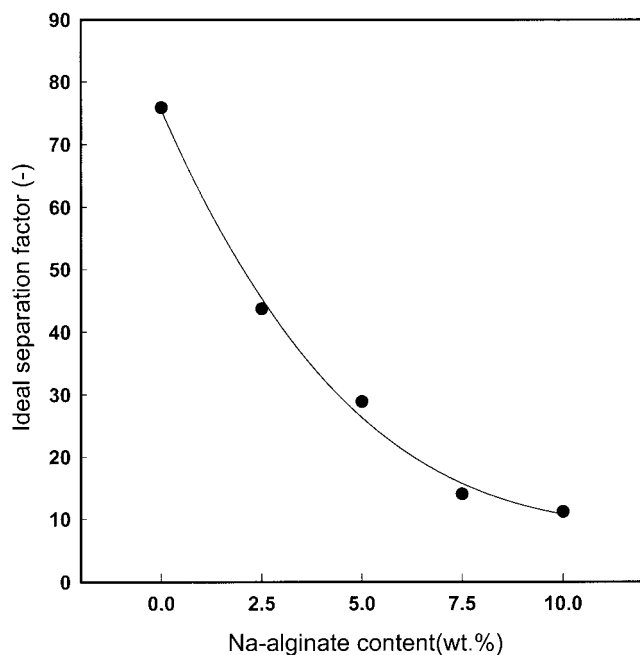
The gas permeation rates of the dry-state Na-alginate/PVA composite membranes were measured using a high-vacuum method. The difference in gas per-

meation rates between the “dry-state” Na-alginate/PVA membranes and the “swollen-state” Na-alginate/PVA membranes was investigated, and compared in Figures 2 and 3. The nitrogen permeation rates of the dry-state Na-alginate/PVA composite membranes were in the range of  $1.7\text{--}4.9 \times 10^{-10}$  cm<sup>3</sup>(STP)/cm<sup>2</sup> s<sup>-1</sup> cmHg<sup>-1</sup>, compared with those of swollen Na-alginate/PVA membranes. The carbon dioxide permeation rates through the dry-state composite membranes were  $4.1\text{--}7.6 \times 10^{-10}$  cm<sup>3</sup>(STP)/cm<sup>2</sup> s<sup>-1</sup> cmHg<sup>-1</sup>. The dry-state Na-alginate/PVA membranes show very low gas permeation property, similar to that of other hydrophilic polymers.<sup>13–15</sup> In hydrophilic polymers such as Na-alginate and PVA, the intermolecular hydrogen bonds are formed mainly between the ring ether oxygen and hydroxyl groups (OH) in PVA, whereas other bonds are also formed with secondary OH at either C-2 or C-3 position and the OH of the PVA component. These hydrogen bonds led to a physical crosslink between polymer chains; the resulting physical crosslink restricted polymer chain mobility, thus decreasing the gas permeation rate. The low level of gas permeation rate is probably attributable to the high level of hydrogen bonding in the dry-state Na-alginate/PVA membranes.

There are dramatic changes in transport properties between the “dry” and the “water-swollen” Na-alginate/PVA membranes. The nitrogen permeation rates of water-swollen membranes were in the order of  $0.4\text{--}7.6 \times 10^{-7}$  cm<sup>3</sup>(STP)/cm<sup>2</sup> s<sup>-1</sup> cmHg<sup>-1</sup> and the carbon dioxide permeation rates were  $3.7\text{--}8.5 \times 10^{-6}$  cm<sup>3</sup>(STP)/cm<sup>2</sup> s<sup>-1</sup> cmHg<sup>-1</sup>, which were 10,000 times



**Figure 3** CO<sub>2</sub> permeation rates of water-swollen Na-alginate/PVA membranes as a function of Na-alginate content in membrane ( $T = 25^{\circ}\text{C}$ ,  $P = 10$  atm).



**Figure 4** Ideal separation factor of water-swollen Na-alginate/PVA membrane as a function of Na-alginate content in membrane ( $T = 25^{\circ}\text{C}$ ,  $P = 10$  atm).

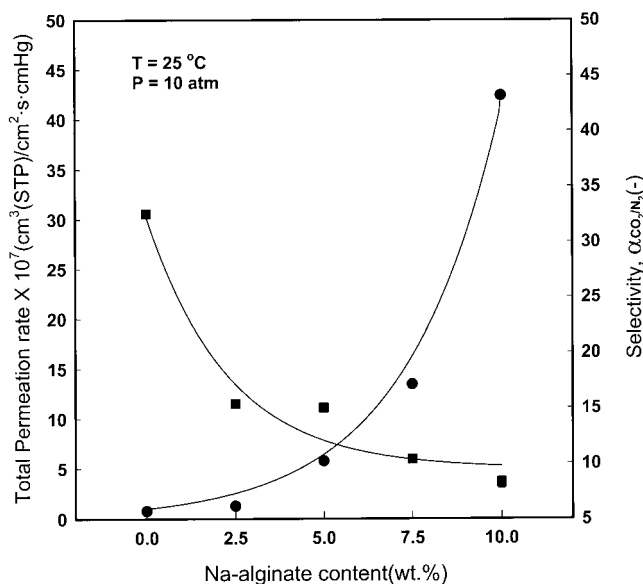
higher than those of dry-state membranes. For water-swollen Na-alginate/PVA membranes, water molecules that induced the increase of intermolecular distances of the membrane material caused the great change of the gas permeation rates of such hydrophilic membranes. The result indicated that water in the swollen membrane played an important role in the transport process. Given that a water molecule in swollen membrane is a good swelling agent and a moving carrier for gases, the permeation rates of gases mainly depend both on the sorption and desorption rates and the diffusivity of the gases in the water that exists in the membrane. The permeation rates of  $\text{CO}_2$  and  $\text{N}_2$  gases through the water-swollen membranes increased with increasing the Na-alginate content from 0 to 10 wt %. However, the ideal separation factor ( $\text{PCO}_2/\text{PN}_2$ ) decreased with the Na-alginate increase, as shown in Figure 4, because the increase of  $\text{N}_2$  diffusivity is much higher than that of  $\text{CO}_2$ .

#### Na-alginate on mixture gas permeation

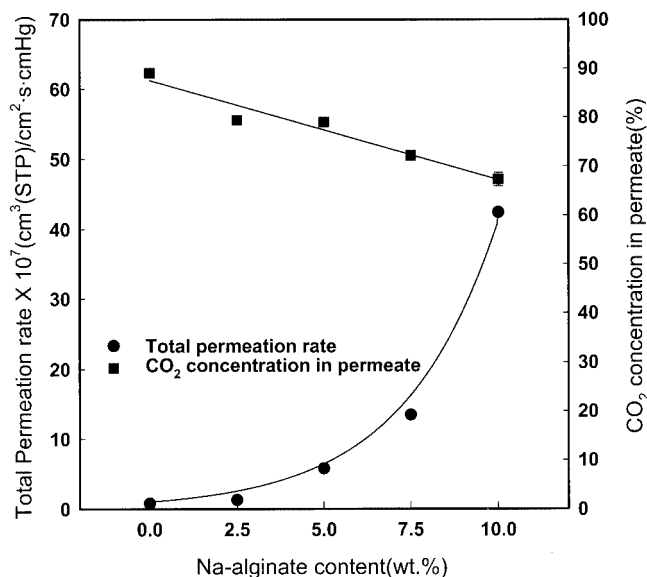
The measurements were carried out with a mixture ( $\text{CO}_2/\text{N}_2 = 20/80$ ) of  $\text{CO}_2$  and  $\text{N}_2$  gas at  $25^{\circ}\text{C}$  while the pressure on feed side was 10 atm and the sweep side pressure of the helium stream was atmospheric. The total permeation rates of mixture gas and compositions of the permeates were determined using a GC apparatus. The mixture gas permeation results on water-swollen Na-alginate/PVA membranes are shown in Figure 5. The water-swollen PVA membrane

showed a low gas permeation rate. The Na-alginate/PVA membranes showed significantly improved gas permeation rates by incorporating Na-alginate in the membrane. When a hydrophilic polymer membrane is exposed to a water vapor, the water molecules may dissolve in the matrix and change the microstructure of a membrane. This dissolution process could lead to an increase in the fractional free volume of a membrane. The Na-alginate, which possessed high water sorption property, had a substantial effect in promoting gas permeation properties of water-swollen membranes.

Figure 5 shows the carbon dioxide selectivity for the mixed gas system. The carbon dioxide selectivity decreased from 32.5 to 8.3, in response to the increase in the molar content of Na-alginate component from 0 to 10%. There are differences between the selectivity (real separation factor) and the ideal separation factor. Often the selectivity is not equal to the ideal separation factor because of "coupling effects" of the dissolved gases on the membrane.<sup>16</sup> The change of permeation rate of each gas component within the membrane is caused not only by the interaction between individual gas components and the membrane matrix, but also by the mutual interaction between gas components within the membrane, which is called the coupling effect. For a water-swollen Na-alginate/PVA membrane, the mutual interaction exists between water molecules and the carbon dioxide, increasing solubility of carbon dioxide in the membrane. The increased solubility can cause a longer residence time of carbon dioxide in the swollen Na-alginate/PVA polymer matrix. Even though the solubility of nitrogen decreases



**Figure 5** Total permeation rates and the selectivity of water-swollen Na-alginate/PVA membrane as a function of Na-alginate content in membrane ( $\text{CO}_2/\text{N}_2 = 20 : 80$  v/v%).



**Figure 6** Na-alginate effect on mixture gas permeation property: the total permeation rates of mixture gas versus carbon dioxide concentration in permeates ( $\text{CO}_2/\text{N}_2 = 20$ : 80 v/v%,  $T = 25^\circ\text{C}$ ,  $P = 10$  atm).

in the presence of water molecules and carbon dioxide, it appears that nitrogen is displaced from the fixed-volume sites by water molecules, causing it to diffuse faster through the polymer matrix. Therefore, diffusion selectivity of the mixture gas through the membrane can be decreased with increasing Na-alginate content in the membrane. The decrease of carbon dioxide selectivity is probably attributable to the increase of  $\text{N}_2$  diffusion rates through water-swollen membranes.

The permeation rates of mixture gas through water-swollen Na-alginate/PVA membranes were found to increase exponentially with the Na-alginate content, whereas the carbon dioxide concentration in permeates was decreased linearly as shown in Figure 6.

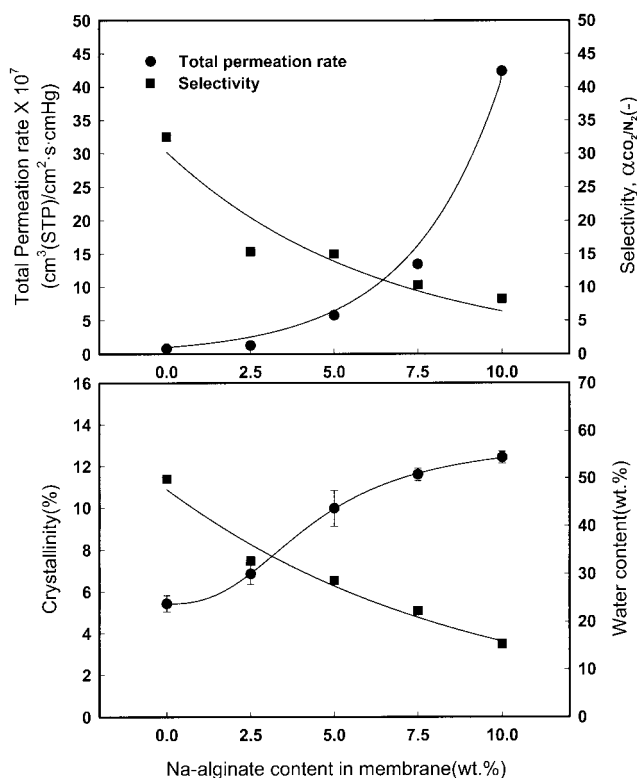
#### Effects of water content and crystallinity

The relationship between the water content of the swollen membrane and the permeation rates of the gases was investigated. It appeared that total permeation rates of the gases were strongly affected by the amount of the water content, as shown in Figure 7, and there was a significant difference between the single gas permeation rate and the mixture gas permeation rate. The effect of Na-alginate component on the gas permeation rate of water-swollen membranes showed that the permeation rate increased with increasing the Na-alginate content. The behavior of the gas permeation of the water-swollen Na-alginate/PVA membrane was proportional to the amount of the water content and the swelling be-

havior of the crosslinked Na-alginate/PVA membranes.

The swelling indices of the prepared membranes increased with increasing Na-alginate content, indicating that with increasing Na-alginate content, the relative crosslinking degree decreased. It is well known that a membrane with less crosslinking density has more polymeric chain mobility, leading to higher free volume in the membrane. Therefore, both the solubility and diffusivity of a permeating gas through the membrane can be increased in the gas permeation process to increase the permeation rate of the gas. The increase of water content in a membrane makes the membrane more flexible, thereby decreasing the energy required for diffusive transport through the membrane and increasing the diffusivity of the gas.

The diffusion transport of gases in a water-swollen membrane occurs through fluctuating water-filled spaces of the network by successive jumps through "holes" (free volume) that are larger than the gas. The effective free volume for gas diffusion corresponds to the free volume of water in the membrane. The permeation of gases through water-swollen membranes is therefore dependent on the degree of membrane hydration. Therefore, carbon dioxide permeation rates of the water-swollen membranes increased with increas-



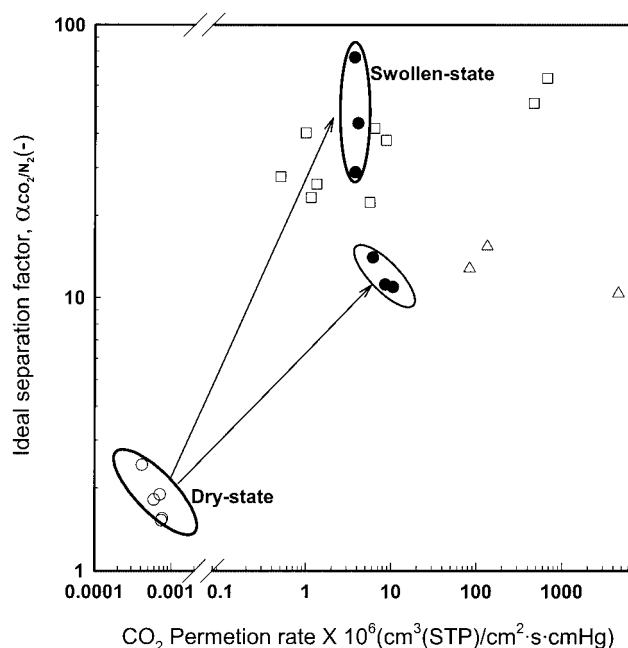
**Figure 7** Effects of water content and crystallinity on gas permselective properties of Na-alginate/PVA membranes as a function of Na-alginate content in membrane.

ing water content in the membrane, whereas carbon dioxide selectivity of the membrane decreased with increasing water content in the membrane.

The effect of crystallinity of the membranes on the separation performance of the water-swollen membranes was investigated. As shown in Figure 7, total permeation rates of mixture gas increased with decreasing crystallinity of the membranes. For synthetic materials, permeability of gas depends on several factors such as the ratio between the crystalline and amorphous zones, polymeric chain mobility, and specific interactions between the functional groups of the polymers and the gases in the amorphous zones. According to Donhowe and Fenema,<sup>17</sup> permeability increases with decreasing crystalline/amorphous ratio because permeation occurs through the amorphous zones of the film. It has been established that changes in the crystallinity degree affect many properties of polymer material, particularly permeation properties; for instance, in the presence of a larger number of crystallites, the diffusion path of a gas molecule in the membrane increases because of the greater tortuosity of the path. As a result, the gas permeation rate of the membrane, which has a larger number of crystallites, decreases with high crystallinity. Also the hydrophilicity of the membrane is an important factor controlling the permeation rate of water-swollen membrane. Hodge et al.<sup>18</sup> reported that water exists only in the amorphous regions of the PVA, but also destroys crystallinity, most probably by attacking crystallites at the crystalline/amorphous interface. At this interfacial region in semicrystalline PVA, the chain configuration is in transition between tightly packed planar zig-zag, where access to the hydroxyl group is restricted. In the swollen amorphous region more free volume is available and the water molecules encounter less resistance to diffusion. Because the water molecules penetrate the transitional region at the interface between crystallite and amorphous regions, the structure is forced open by water molecules disrupting intermolecular hydrogen bonds. Therefore, fresh areas of the crystallite are exposed to attack from the water molecules, and the crystalline regions are progressively dissolved, increasing the gas permeation rates of water-swollen Na-alginate/PVA membranes.

#### Comparison with other membranes

For the purpose of membrane performance comparison of Na-alginate/PVA membranes with other membranes, Figure 8 was compiled, presenting the carbon dioxide permeability from literature data for a number of membrane materials. As shown in Figure 8, incorporation of Na-alginate into PVA membrane has a substantial effect in promoting gas permeation prop-



**Figure 8** Comparison of swollen Na-alginate/PVA membranes with other polymeric membranes: ○, dry-state Na-alginate/PVA membranes; ●, swollen-state Na-alginate/PVA membranes; □, glassy membranes<sup>19,20</sup>; △, rubbery membranes.<sup>19</sup>

erties of water-swollen PVA membranes. Comparison with other membrane materials shows that the Na-alginate/PVA is found to exhibit relatively good permeation rates. However, lower separation factors are observed for the gas pairs CO<sub>2</sub>/N<sub>2</sub>. The additives or carriers that can react with carbon dioxide would be added to improve carbon dioxide selectivity of the Na-alginate/PVA membranes for future studies.

#### CONCLUSIONS

The PVA membranes and the Na-alginate/PVA membranes for gas separation can be prepared by a dip-coating method with UOP UF membranes with a MWCO of 30,000 g/mol as a support. The crosslinking of the membranes containing glutaraldehyde as a crosslinking agent can be achieved by immersion for 4 h at 40°C in crosslinking solution (IPA : water : HCl = 90 : 10 : 1 weight ratio). Incorporation of even a small amount of polysaccharide into PVA membrane has a substantial effect in promoting gas permeation properties of water-swollen polysaccharide/PVA membranes. The mixture gas permeation rates through water-swollen Na-alginate/PVA membranes were found to increase exponentially with the Na-alginate content. The gas permeation rates through membranes are dependent on the amount of water in the membrane as well as crystallinity degrees of membranes.

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