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Modeling Study on a Circulatory Hollow-Fiber Membrane Absorber for CO_2 Separation

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이산화탄소 분리를 위한 순환식 중공사 막흡수기에 관한 모델링 연구

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Abstract: For several years lots of attempts have been made to establish the liquid membrane-based techniques for separations of gas mixtures especially containing carbon dioxide. A more effective system to separate CO_2 from flue gases, a *circulatory hollow-fiber membrane absorber*(HFMA) consisting of absorption and desorption modules with vacuum mode, has been considered in this study. Gas-liquid mass transfer has been modeled on a membrane module with non-wetted hollow-fibers in the laminar flow regime. The influence of an absorbent flow rate on the separation performance of the circulatory HFMA can be predicted *quantitatively* by obtaining the CO_2 concentration profile in a tube side. The system of CO_2/N_2 binary gas mixture has been studied using pure water as an(inert) absorbent. As the absorbent flow rate is increased, the permeation flux(i.e., defined as permeation rate/membrane contact area) also increases. The *enhanced selectivity* compared to the previous results, on the other hand, shows the decreasing behavior. It has been found obviously that the permeation flux depends on the variations of pressure in gas phase of desorption module. From an accurate comparison with the results of conventional flat sheet membrane module, the advantageous permeability of this circulatory HFMA can be clearly ascertained as expected. Our efforts to the theoretical model will provide the basic analysis on the circulatory HFMA technique for a better design and process.

요 약 : 이산화탄소를 포함하는 기체혼합물의 분리를 위해 액체막을 기반으로 하여 관련된 많은 연구가 시도되어 왔다. 폐가스로부터 이산화탄소를 분리하기 위해 본 연구에서는, 흡수제(absorbent)가 흡수(absorption)모듈과 진공조작식 탈착(desorption)모듈간을 계속 순환하는, 보다 개선된 형식의 순환식 중공사 막흡수기(circulatory HFMA)를 새로이 구성하고 분리성능에 관한 기초적인 해석을 하였다. 기-액 물질전달 모델식에 수치해를 적용하여 중공사 막 내부에서의 액상에 대한 농도분포를 정량적으로 예측하였고, 순환하는 흡수제의 유속에 따른 투과플럭스와 선택도의 변화거동을 살펴 보았다. CO_2/N_2 원료혼합기체와 흡수제로서 반응이 없는 순수(pure water)에 대해 계산을 수행하였다. 결과로 흡수제의 유량이 증가함에 따라 투과플럭스는 증가하는 반면에 종전 방식에 비해 높은 값을 나타낸 선택도의 경우는 점차 감소

하였다. 한편 투과플럭스는 진공조작변수인 탈착모듈에서의 기상압력(p_d)에 크게 좌우됨을 보았다. 기존의 평판형 막모듈과의 비교로부터 예상했던 바와 같이 본 연구에서의 중공사 막모듈이 우수한 투과율을 나타냄을 확인할 수 있었다. Circulatory HFMA의 실제 설계를 위한 기초해석이 본 연구가 갖는 의의이다.

1. Introudction

Since the potential of immobilized liquid membrane(ILM) for gas separation was realized[1], the attractive features have stimulated significant research in the field of gas separation using liquid membranes[2]. Due to the principle of facilitated transport through liquid membranes, it is possible to achieve high permeability and selectivity. To overcome the problem of mechanical stability Sirkar et al.[3, 4] have developed the idea of the hollow-fiber contained liquid membrane(HFCLM) technique under four different operational modes. Although the stability of such a system is better than that of ILM, a mass transfer resistance through the liquid existing in the shell side between hollow-fibers is large because the overall diffusion thickness is too large and stationary. In this respect, it should be noted that a more developed type of liquid membrane, designated as a "flowing liquid membrane" was proposed by Teramoto et al.[5]. By circulating a liquid membrane solution in a thin channel between polymeric membranes the diffusional resistance decreased compared to a conventional liquid membrane system, and so higher permeability and stability could be obtained consequently.

Recently, Shelekhin and Beckman[6] considered the possibility of combining absorption and membrane separation processes in one integrated system called a "circulatory membrane absorber". They proposed a configuration that performs the gas transport between absorption and desorption modules by the circulation of an absorbent. Their investigation has predicted efficiently the performance of flat sheet membrane, however, there are restrictions for describing a convective flowing effect because the plug flow recognized in their work is not a general case. The increased selectivity was also observed in experiments on a CO_2/CH_4 gas mixture separation, using a laboratory membrane absorber.

In practice, the use of hollow-fiber module in the membrane absorber is of great benefit than that of flat sheet membrane. It is specifically advantageous to reduce the diffusion resistance of liquid layer using hollow-fiber membranes as it is relatively easy to obtain the hollow-fibers with an inner diameter of about several tens micrometers. At the same time, the hollow-fiber module is the most effective one for a membrane absorber design because it provides the highest values of surface area to equipment volume ratio[7]. These points have led investigators to explore many kinds of different applications. The studies on hollow-fiber gas-liquid contactor were carried out in many systems of gas mixtures, where the hollow-fiber acted as a boundary between the gas and liquid phases. Matsumoto et al.[8] studied the gas-liquid mass transfer in an artificial gill, which allowed simultaneous absorption and stripping of CO_2 and O_2. The liquid phase mass transfer coefficient inside the lumen was found to be in accordance with Sieder-Tate equation. As an application of hollow-fiber based membrane absorber, Matsumoto et al.[9] have also conducted a series of experiments for CO_2 removal from the flue gas of a thermal power plant, although they employed desorption operations with the high temperature steam. It has been obtained that microporous hydrophobic hollow-fibers are suitable for their CO_2 separation system.

In the present study, we first discuss the construction of hollow-fiber based membrane absorber consisting of absorption and desorption modules; we name it circulatory hollow-fiber membrane absorber(HFMA). A laminar flow mass transfer in non-wetted hollow-fibers is analyzed to obtain the accurate concentration profile. The separation performance for CO_2/N_2 gas mixture on this new type of circulatory HFMA is investigated carrying out

the theoretical prediction. Our predicted results will be also compared with the previously proposed flat sheet membrane modules. The present analysis attempts to provide a basic understand on a membrane absorber as well as its promising aspects on the separations of gas mixtures containing acid gases such as carbon dioxide.

2. Model Development

2. 1. Circulatory Hollow-Fiber Membrane Absorber

By applying the structural type of membrane absorber proposed by Shelekhin and Beckman[6], a schematic description of circulatory HFMA can be shown in Fig. 1. The circulatory HFMA is a composed unit having an absorption module on the high pressure side and an in situ desorption module on the low pressure side. In a desorption module, a regeneration of the absorbent passed through the hollow-fibers of absorption module is occurred by stripping at a reduced pressure, and subsequently the permeating gases desorb from the absorbent. For both the separation and the recovery of CO_2 from gas mixture vacuum mode of operation in the permeate side is applied here, and more detailed descriptions regarding this vacuum operation may be found elsewhere[4].

The membrane absorber can have a number of

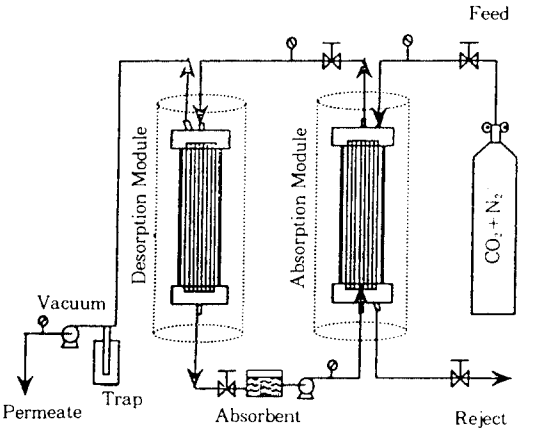

Fig. 1. Schematics of circulatory hollow-membrane absorber.

configurations, however, the countercurrent absorbent liquid flowing through the lumen of the hollow-fibers in modules is considered for the present circulatory HFMA as shown in Fig. 1. Depending on the interaction of the hollow-fiber material as well as the pressure conditions on each side of the membrane, either wetted or non-wetted cases may be realized[10, 11]. Many studies were published on the application of hydrophobic microporous membranes for separations or gas absorptions. Sirkar et al.[3, 4] used Celgard polypropylene hollow-fibers to make the hollow-fiber contained liquid membrane permeators. As mentioned in the earlier section, Shelekhin and Beckman[6] obtained successfully a high selectivity on the CO_2 separation using their flat sheet membrane absorber made with polyvinyltrimethylsilane (PVTMS). A stagnant layer of gas is present in the pores of a non-wetted membrane mode. In this case, the pressure condition of each phase has to be $\Delta P_c > P_{liquid} > P_{gas}$, where ΔP_c means a critical pressure estimated from a surface tension and a contact angle for the used membrane. If this condition is vice versa, the membrane pores become wetted with an absorbent liquid(i.e., liquid-filled pores), and as a result a stagnant liquid layer is present.

2. 2. Mass Transfer Analysis

It is reasonable to solve for the concentration profile in a single hollow-fiber of liquid phase to predict the separation performance. Firstly, in order to develop a theoretical model the following assumptions have been made[12]:

i) no axial diffusion
ii) fully developed laminar flow in the tube side
iii) ideal gas behavior
iv) applicability of Henry's law.

The physical situation involved in the present analysis is depicted in Fig.2. Hypothetic dashed lines of shell side mean the free surface radius based on an outer radius of hollow-fiber and a packing fraction in a module[11]. The concentration profile of liquid phase in a tube side $C_{l,t}$ can be calculated

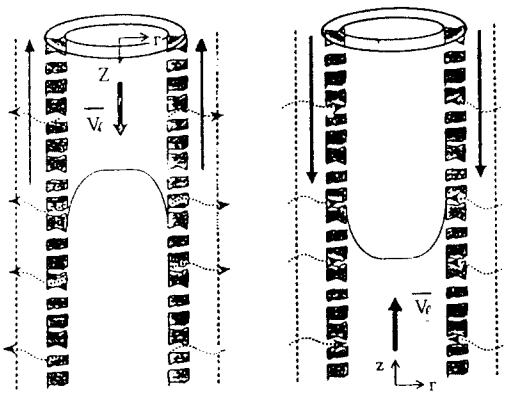

Fig. 2. Mass transfer in a gas/membrane/liquid system of absorption and desorption modules, with non-wetted hollow membranes.

from the differential mass balance in cylindrical coordinates that describes entirely both the diffusion and the convection in a medium

$$V_{z,l}\frac{\partial C_{l,t}}{\partial z}=D_l\left[\frac{\partial^2 C_{l,t}}{\partial r^2}+\frac{1}{r}\frac{\partial C_{l,t}}{\partial r}\right]+R_t \qquad (1)$$

Absorption processes accompanied by chemical reactions usually are very attractive because the mass transfer can be enhanced by the reaction. However, in this work the aspects of physical absorption is considered confinely, therefore, the reaction term in Eq.1 will be left out. Basically, the radial velocity profile of absorbent is given by

$$V_{z,l}(r)=2\bar{V}_l[1-(r/r_t)^2] \qquad (2)$$

At the centerline of the tube, the concentration profile is symmetrical and therefore satisfies

$$\frac{\partial C_{l,t}}{\partial r}=0 \qquad \text{at } r=0 \qquad (3)$$

With partition coefficient, namely, dimensionless Henry's law constant H, the following condition between gas and liquid phases is valid at the membrane-liquid interface($r=r_t$):

$$C_{g,m}=C_{l,t}/H \qquad (4)$$

Flux continuity at this boundary is also valid as

$$D_m\frac{\partial C_{g,m}}{\partial r}=D_t\frac{\partial C_{l,t}}{\partial r} \qquad (5)$$

Then the following boundary condition of the third kind(i.e., Robin type) can be imposed at the membrane-liquid interface[13]

$$D_t\frac{\partial C_{l,t}}{\partial r}=\left[(C_{g,m})_{r=r_o}-\frac{(C_{l,t})_{r=r_t}}{H}\right] \quad \text{at } r=r_t \qquad (6)$$

where $k_m\left(=\frac{D_m}{r_o-r_t}\right)$ is the mass transfer coefficient of gas phase in membranes, and $\left(\frac{r_o-r_t}{\ln(r_o/r_t)r_t}\right)$ is the shape factor ($=S$) based on the inside radius of tube r_t. In Eq. 6, $(C_{g,m})_{r=r_o}$ corresponds to a gas phase concentration, which equals to the product of solubility coefficient (σ) and the gas phase pressure in a shell side of hollow-fiber(e.g., in case of absorption module, it is expressed by p_a). Initial condition at the tube entrance($z=0$) is

$$C_{l,t}=C_{inlet} \qquad \text{at } z=0,\ r=r \qquad (7)$$

We introduce the following dimensionless variables

$$C^*=\frac{C_{l,t}}{C_{inlet}},\ z^*=\frac{zD_t}{v_l r_t^2}=\frac{z}{r_t}\frac{1}{Pe},\ r^*=\frac{r}{r_t}$$
$$\beta=\frac{\sigma p_a}{P_{inlet}},\ Sh=\frac{r_t k_m S}{D_t}=\frac{D_m}{\ln(r_o/r_t)D_t} \qquad (8)$$

The dimensionless axial distance down the hollow-fiber z^* is the inverse of the Graetz number. Then the Eq.1 and the above boundary conditions of Eqs. 3 and 6 can be written as

$$2(1-r^{*2})\frac{\partial C^*}{\partial z^*}=\left[\frac{\partial^2 C^*}{\partial r^{*2}}+\frac{1}{r^*}\frac{\partial C^*}{\partial r^*}\right] \qquad (9)$$

$$\text{IC.}: \quad C^*=1 \qquad \text{at } z^*=0,\ r^*=r^* \qquad (10)$$

$$\text{BC. 1}: \frac{\partial C^*}{\partial r^*}=0 \qquad \text{at } r^*=0 \qquad (11)$$

$$\text{BC. 2}: \frac{\partial C^*}{\partial r^*}=Sh\left[\beta-\frac{C^*}{H}\right] \quad \text{at } r^*=1 \qquad (12)$$

The above Eqs.9~12 are solved using a finite difference scheme of Crank-Nicholson method[14, 15]. The discritizations of diffusion and convection terms give a formation of tridiagonal matrix, and this set is solved simultaneously. In most cases, a

grid of 20×600 is chosen for the computations with sufficiently high accuracy. Depending on the state of BC. 2 imposed at the membrane-liquid interface, the above Eq.9 will describe the concentration profiles in the absorption and/or desorption modules, accordingly.

2. 3. Computational Procedures

Since the average concentration(i.e., mixing cup concentration) is defined as averaging the local concentration weighted with the velocity profile over the cross section of the hollow-fiber, it can be calculated by summations with the index running over the radial grid points. Then the transmembrane flow rates of i component, entering or leaving the absorption and desorption modules, can be determined in accordance with, respectively

$$F^i_{abs} = \bar{v}_i A_T (C_{inlet,\ abs} - C_{out,\ abs}) \quad (13a)$$

$$F^i_{des} = \bar{v}_i A_T (C_{inlet,\ des} - C_{out,\ des}) \quad (13b)$$

where AT means an overall cross-sectional area of hollow-fibers packed in the membrane module. Here, $C_{inlet,\ abs}$ and $C_{inlet,\ des}$ are correspond to the $C_{outlet,\ des}$ and $C_{outlet,\ abs}$, respectively. Since an absorbent is continuously pumping in the circulatory HFMA, the magnitudes of both the transmembrane flow rates in absorption and desorption modules should be equal. Computations are repeated until this condition for convergence is satisfied as shown in Fig. 3. The converged transmembrane flow rate gives the permeation rate F^i and the permeation flux J^i(i.e., permeation rate/membrane contact area between gas and liquid) at the desorber outlet. Next, the selectivity or ideal separation factor of the membrane to any two gases i and j can be estimated as

$$\alpha_{ij} = (J^i/\Delta p^i_m)/(J^j/\Delta p^j_m) \quad (14)$$

where Δp^i_m is the partial pressure difference of i component across the membrane. This definition is applicable for near vacuum conditions on the low pressure side and in the absence of interactions between the diffusing species, as well as between the species and the membrane. Selectivity of the membrane may reasonably be assumed constant in engineering practice, and furthermore it is identical with the actual selectivity in case of the vacuum operation. Fig. 3 shows an overall computation algorithm employed in this study.

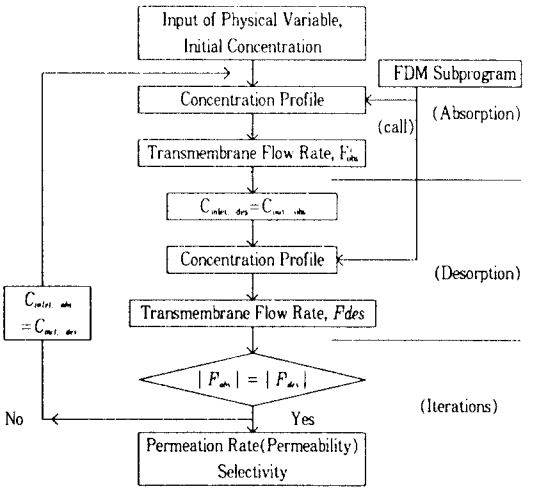

Fig. 3. Flow chart of computation program for circulatory HFMA.

3. Results and Discussion

For the computations we have taken the parameters given in Table 1. The Celgard X-20 hydrophobic membrane is chosen, which has 0.045 and 0.04cm as outer and inner diameters, respectively. The circulatory HFMA is composed of two membrane modules with an equal membrane surface area, in which 50 hollow-fibers with 20cm length

Table 1. Physical Parameters of Gases through Pure Water for Computations

	*$D_i \times 10^5$ (cm^2/sec)	**$D_m \times 10^2$ (cm^2/sec)	***$\sigma \times 10^4$ scc(g)/cc(w) · cmHg
CO_2 (25℃)	1.92	1.1	109.3
N_2 (25℃)	1.88	1.6	1.86
CH_4 (25℃)	1.49	1.4	3.97

* From Refs. 2, 3, and 12

** Porosity/Tortuosity = 0.4/2.5 for Celgard X-20

*** From Refs. 3 and 16

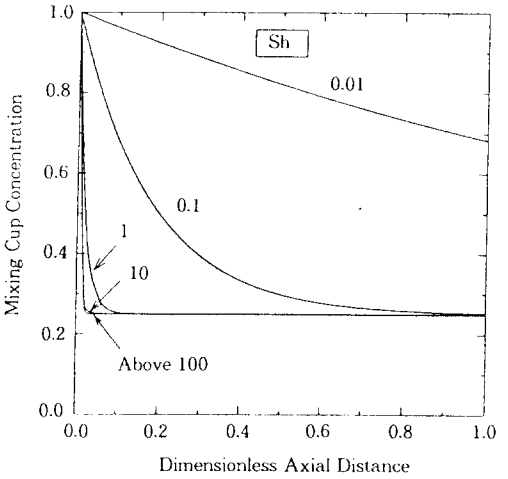

Fig. 4. Dimensionless mixing cup concentration of CO_2 as a function of the dimensionless axial distance and the Sherwood number for the case of desorption(p_d=30cmHg).

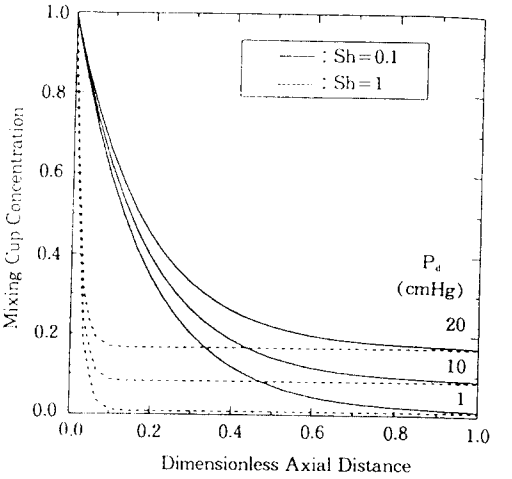

Fig. 5. Dimensionless mixing cup concentration of CO_2 as a function of the dimensionless axial distance z* and the pressure in gas phase for the case of desorption.

are regularly packed. The mixing cup concentration is a function of both the Graetz number and the Sherwood number(Sh) for mass transfer. As given in Eq.8, the Sh can be considered as the ratio of mass transfer resistance in the tube side to that in the membrane. Fig. 4 shows the variation of the dimensionless mixing cup concentration of CO_2 with the dimensionless axial distance as a function of the Sh. With increasing the Sh, the variation of the mixing cup concentration approaches the case of the infinite Sherwood number. It is noteworthy that for Sh is infinite the membrane resistance is negligible and the problem reduces to the classical Graetz problem [12, 13]. While the Sh is sufficiently less than 1, the membrane resistance is dominant and the problem has a constant flux boundary condition. Effects of the gas pressure at desorption module on the mixing cup concentration, at a given value of Sh, are depicted in Fig. 5. Correspondingly, this feature of mixing cup concentration with the variations of Sh can also be obtained in the case of absorption.

Let us consider the feed gas as a binary gas mixture CO_2/N_2 with composition 20/80 for here. Numerical results predict the CO_2 concentrations at desorption module as illustrated in Fig. 6. The CO_2 inlet concentration at desorption module ($C_{inlet,\ des}$), which equals to the CO_2 outlet concentration at absorption module ($C_{out,\ abs}$), is decreased with an increase of circulating absorbent liquid velocity. On the contrary, the CO_2 outlet concentration at desorption module is increased, and a difference between these two concentrations determines the permeation rate F' majorly related with the performance of the circulatory HFMA. As shown in Fig. 7, with an increase of absorbent velocity the permeation rate increases. We can also observe a decrease of permeation rate with an increase of the pressure in gas phase of desorption module. This pressure is an operational condition controlled by the vacuum process. Of course, it will be seen that continuously increasing absorbent velocity causes eventually the decrease of permeation rate. The behavior of permeation flux J' can be also observed as shown in Fig. 8.

Another important factor of separation performance to be observed is a selectivity, which can be estimated from the Eq.14. As shown in Fig. 9, the se-

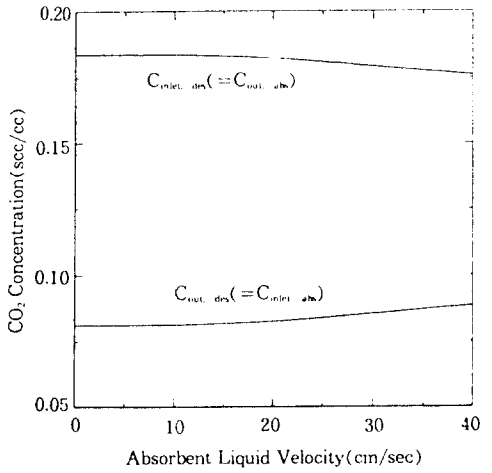

Fig. 6. CO_2 concentration changes with an increase of the absorbent liquid velocity.

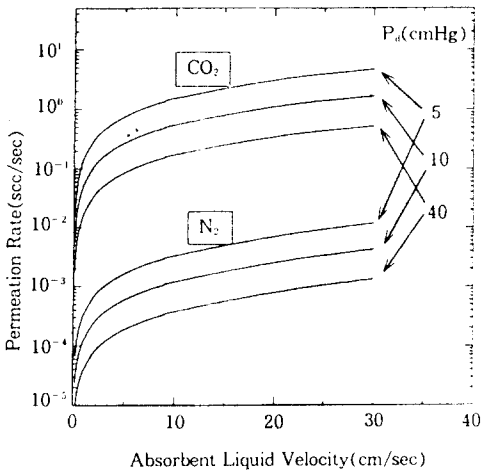

Fig. 7. Permeation rates of CO_2 and N_2 in the circulatory HFMA versus absorbent liquid velocity.

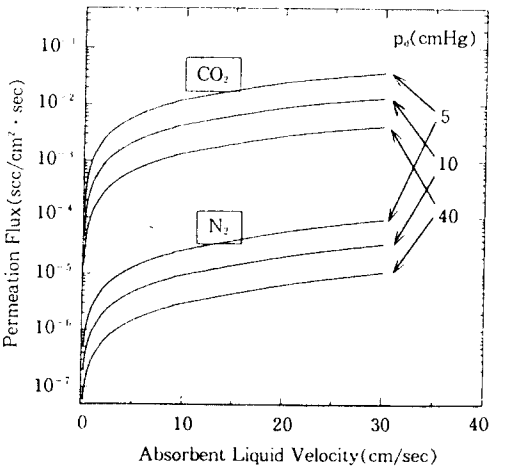

Fig. 8. Permeation fluxes of CO_2 and N_2 in the circulatory HFMA versus absorbent liquid velocity.

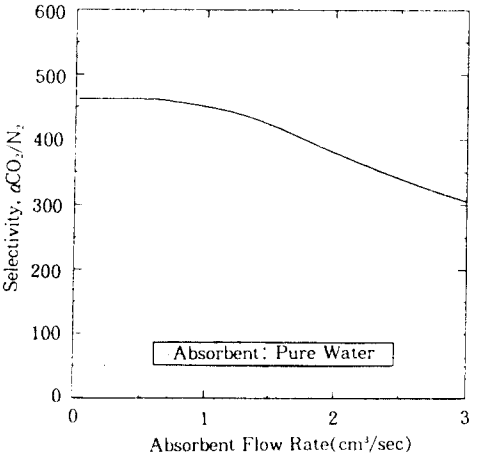

Fig. 9. Selectivity for CO_2/N_2 in the circulatory HFMA versus absorbent flow rate.

lectivity of CO_2/N_2 system is decreased gradually with an increase of absorbent flow rate. The maximum selectivity achieved at an infinite lower flow rate is higher than 450. This value is more than 7 times higher than the selectivity simply based on the solubility alone, that is about 59(=109.3/1.86). Our predicted result is for the case of pure water as an absorbent, nonetheless, this value is higher than the results of Guha et al.[2]. They reported the selectivity for CO_2/N_2 through the ILM containing 20% (wt.) diethanolamine absorbent as the range about 350~400. This fact obviously allows us that the circulatory HFMA is a more efficient technique than the previous ILM or HFCLM. It is noted that at high flow rates the influence of solubility coefficients on the permeation rate is not significant. The selectivity in this case is determined mainly by the nature of the membrane itself [6].

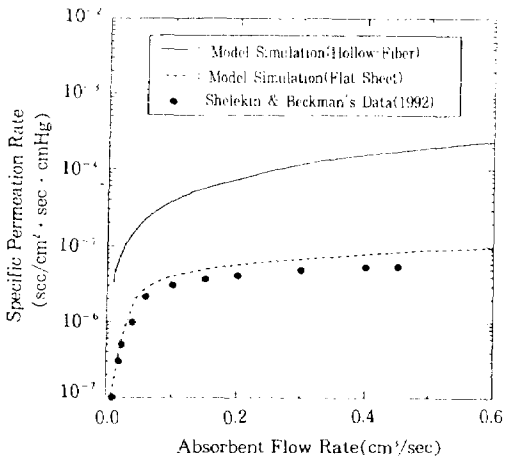

Fig. 10. Comparison of the specific permeation rate of CO_2 between hollow-filber and flat sheet membrane modules with the variation of absorbent flow rate.

We pointed out the advantage of hollow-fiber modules in introduction, therefore, it is necessary that the performance of our circulatory HFMA is compared with the flat sheet membrane previously considered by Shelekhen and Beckman[6]. Geometric dimensions of hollow-fiber membrane module having the same volume can be specified, in which the inner diameter of hollow-fiber is equivalent to the liquid layer thickness of flat sheet membrane. The concentration profile across the flat sheet membrane with parabolic velocity profile can be obtained by solving an one-dimensional problem satisfying the relevant boundary conditions at both sides of membrane-liquid interfaces. As shown in Fig. 10, it is found that the specific permeation rate(i.e., permeability/thickness) of hollow-fiber module is clearly higher than that of the flat sheet membrane module.

4. Concluding Remarks

In this paper, both the possibility and applicability of a new type of circulatory HFMA were considered by the theoretical model approach, with a view to developing a separation and recovery techniques of CO_2 from flue gases. The influence of an absorbent flow rate on the separation performance of circulatory HFMA was investigated using pure water. The permeation flux increases with an increase of absorbent flow rate. Contrary to permeation flux, it is found that the selectivity for CO_2/N_2 shows a decreasing behavior with the absorbent flow rate. The theoretically obtained selectivity at an infinite lower flow rate($\sim$460) is indeed higher than that obtained for the previous techniques. It has been ascertained that in comparing with the conventional flat sheet membrane the circulatory HFMA examined in this work has advantageous separation performance.

However, in order to confirm this technique an experimental verification should be carried out. Moreover, a proper selection of efficient CO_2 absorbent instead of the water is necessary to achieve higher enhanced selectivity. These are concerned with our present investigation.

Nomenclatures

A_T : overall cross-sectional area of hollow-fibers[cm^2]
C : concentration[scc(g)/cc(w) = cm^3(STP)/cm^3]
D : diffusion coefficient[cm^2/sec]
F : permeation rate[scc/sec]
H : dimensionless Henry's law constant[$-$]
J : permeation flux[scc/cm^2 · sec]
k_m : membrane mass transfer coefficient[cm/sec]
P_e : Péclet number[$-$]
p : gas pressure in the membrane module [cmHg]
ΔP_c : critical pressure[cmHg]
Δp_m : partial pressure difference across the membrane[cmHg]
R : reaction term[scc(g)/cc(w) · sec]
r : radial coordinate position[cm]
r_o : outer radius of hollow-fiber[cm]

r_i : inner radius of hollow-fiber[cm]
S : shape factor[−]
Sh : Sherwood number[−]
v : absorbent velocity[cm/sec]
$\bar{v}_l$: mean velocity of absorbent liquid[cm/sec]
z : axial coordinate position[cm]

Greeks

α : selectivity[−]
β : dimensionless variable[−]
σ : solubility coefficient[scc(g)/cc(w) · cmHg]

Subscripts

a, abs : absorption
d, des : desorption
g : gas phase
l : liquid phase
m : membrane side
t : tube side
z : axial direction

Superscripts

i, j : components of gas mixtures
* : dimensionless

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