Study of Mass Transfer Rates for CO_2 Absorption in Hollow Fiber Membrane contactors

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1. INTRODUCTION

Many countries in the world recently agreed to reduce emissions of greenhouse gases into the atmosphere or at least to keep them at the current level at the Kyoto Conference. Carbon dioxide has been proven to be 80% of a greenhouse gas, contributing to the increase of the earth's surface temperature. And it is reported that half of the CO_2 emissions are produced by industry and power plants using fossil fuels [1].

The amount of interfacial area in the hollow fiber modules is about $10000 m^2/m^3$, while $1000\ m^2/m^3$ is reported in that of the conventional absorbers [2]. These advantages of the hollow fiber membrane contactor have been proved through many researches. Karoor and Sirkar [3] studied the absorption of CO_2 and SO_2 from CO_2/N_2 and SO_2/air mixtures, respectively into water using a parallel flow module employing microporous polypropylene fibers.

The mass transfer rate in the membrane contactor module is limited by the mass transfer resistance in the gas, liquid phase and additional resistance introduced by the membrane itself. Although the interfacial area of the membrane is much more than conventional absorbers, the increase of its additional resistance can cause to decrease the mass transfer capacity of the membrane due to the resistance of the membrane itself, a resistance which is increased if the liquid wets the membrane. Therefore, we need to control the non-wetted condition of pores of membrane to keep the good performance of the membrane for gas absorption.

In the present study, the gas absorption accompanied by chemical reaction using hollow fiber membrane absorbers was investigated in the theoretical and experimental aspects. Numerical model for mass transfer in gas absorption was developed and the CO_2 absorption rate was simulated according to gas, liquid velocity and external mass transfer coefficient including mass transfer in gas and membrane. Using PTFE and PVDF membrane module, CO_2 removal efficiency and flux with liquid and gas velocity were experimentally investigated and overall mass transfer coefficients were calculated. Through the comparison between the experimental results and the numerical model, we predicted the external and liquid resistances in CO_2 absorption by PTFE and PVDF hollow fiber membrane.

2. THEORY

Mass transfer in chemical reaction of liquid phase

In the hollow fibers used in this study, the liquid of absorbent flow laminarly through the lumen side and the gas phase flow at the shell side in the module(Fig[1]). Firstly, the radial velocity profile v_r which is formed by forced convection under assuming a fully developed laminar flow in tube is described by Eq.(1)

$$v_r = 2\bar{v}\left[1 - (\frac{r}{R})^2\right]$$ (1)

Next, we can obtain Eq.(2) and Eq.(3) by establishing the differential mass balance of CO_2 and MEA in the condition of chemical reaction. Conveniently, the CO_2 and MEA are denoted as component A and B, respectively.

Component A (CO_2): $v_r \dfrac{\partial C_A}{\partial z} = D_A(\dfrac{1}{r}\dfrac{\partial}{\partial r}(r\dfrac{\partial C_A}{\partial r})) - k_1 C_A C_B$ (2)

Component B (MEA): $v_r \dfrac{\partial C_B}{\partial z} = D_B(\dfrac{1}{r}\dfrac{\partial}{\partial r}(r\dfrac{\partial C_B}{\partial r})) - v_B k_1 C_A C_B$ (3)

The following boundary conditions for three regions are imposed on the membrane system.
At the membrane-liquid interface

Component A : $D_A(\dfrac{\partial C_A}{\partial r}) = k_{ex}(C_{A,g} - C_{A,g,i})$ (4a)

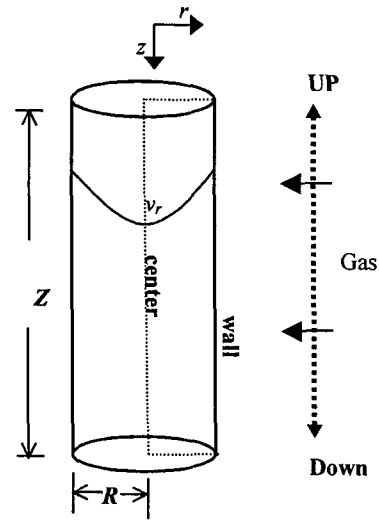where $k_{ex} = \dfrac{1}{\dfrac{1}{k_g} + \dfrac{1}{k_m}}$ (4b)

Component B : $(\dfrac{\partial C_B}{\partial r})_{r=R} = 0$ (4c)

At the centerline of the tube

Component A: $(\dfrac{\partial C_A}{\partial z})_{r=0} = 0$ (4d)

Component B: $(\dfrac{\partial C_B}{\partial z})_{r=0} = 0$ (4e)

At z=0 : $C_A = 0$ and $C_B = C_{B0}$ (4f)

The concentration profiles of component A and B are obtained numerically by using Crank-Nicholson method.

3. EXPERIMENTAL

The experimental sep-up for CO_2 removal and recovery was shown in reference[6], in which it constituted hybrid process of membrane contactor and thermal stripping column. In the case of absorber, the gas containing 25% of CO_2 (balance N_2) was passed upstream in the tube side of the membrane module and the absorbent of MEA (2-monoethanalamine) 5wt% was supplied downstream in the shell side. The membrane contactors used as CO_2 absorber in this study are the PTFE (Polytetrafluoruethylene, Sumitomo Co. Japan) and PVDF (Polyvinylidinefluoride, Krict Korea) hollow fibers. Table 1 shows the properties for each module.

4. RESULT AND DISCUSSION

The calculated concentration profile of CO_2 absorbed in liquid is presented in Fig. 2. In the direction of the diameter of one fiber, this figure shows the largest concentration gradient at the wall side of the fiber. This means that the gas-liquid mass transfer at membrane interface is conducted mostly at the wall side of the fiber in which the numerous pores exists. In the direction of the fiber length, the absorbed CO_2 concentration is steeply increased at up side of fiber and then slightly increased along down side direction in the fiber's wall. This initial sudden

		PVDF	PTFE
Fiber	I.D. (μm)	1000	830
	O.D. (μm)	1913	1070
	Pore size (μm)	1	0.03
	Porosity (%)	70	-
	Packing density	0.64	0.4
	Number of fiber	70	139

behavior results from the active mass transfer between the fresh liquid and gas phase coming into the fiber's entrance.

It is expected that the PTFE membrane possessing relatively large pore size decrease the CO2 absorption flux by increasing the membrane resistance. It could be proven by observing the phenomena that the absorbent easily penetrates into gas phase in the absorption test using PTFE membrane contactor. Hence, we have to control not only the hydrophobic state of membrane but also membrane pore size in order to form the stable interface between gas and liquid phase

Fig. 3 showed the comparison of numerical and experimental fluxes in PVDF and PTFE membrane as a function of liquid velocity. When the numerical external mass transfer coefficients are 0.001m/s in PVDF and 0.0005 m/s in PTFE, respectively, the results of numerical and experimental fluxes are well agreed. As previously stated, in the numerical result the PTFE membrane had the lower mass
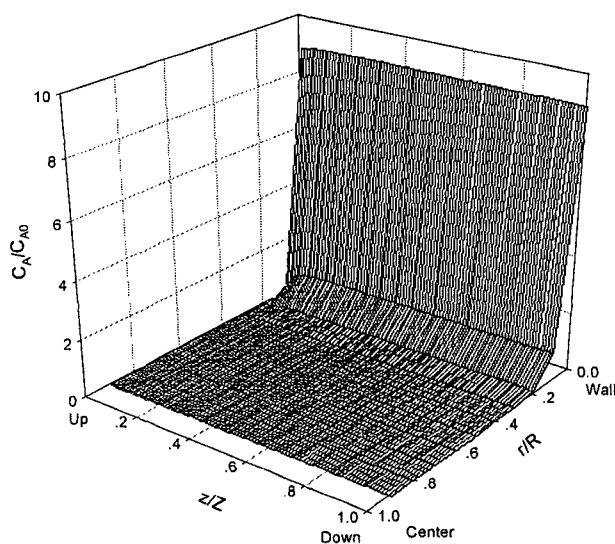
transfer coefficient than the PVDF membrane. Finally, If we define the sum of membrane and gas phase resistance as external resistance, $1/k_{ex}$, the liquid phase resistances can be obtained through the numerical external resistance and experimental overall mass transfer resistance on these two membranes.
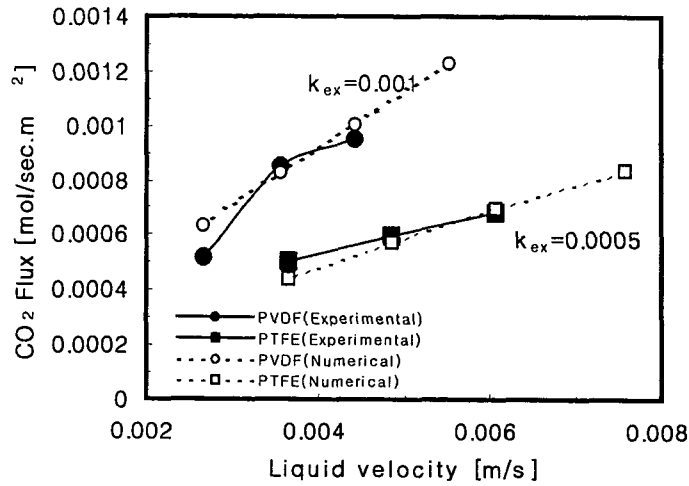
Fig 3. The flux comparison of PTFE and PVDF membranes
as a function of liquid velocity.

5.REFERENCES

1. Desideri U., Paolucci A., *Energy Conversion & Management*, 40 ,1899-1915,(1999)

2. Deshmukh S.P., Li K., *J. Membrane Sci.*, 150, 75-85, (1998)

3. Karoor S. Sirkar K.K., *Ind. Eng. Chem. Res.*, 32, 674-684,(1993)

4. Qi Z, Cussler E.L., *J. Membrane Sci.*, 23, 321-332,(1985)

5. Qi Z. Cussler E.L., *J. Membrane Sci.*, 23, 333-345. (1985)

6. S. H. Yeon, B. K.Sea, K.S. Lee, Y. I. Park, K.H. Lee, *Theories and Applications of Chem. Eng.*, Vol.7, No.2 (2001)