Preparation and Simulation of Polycarbonate Hollow-Fiber Membrane for Gas Separation

Mahmoud Reza Sohrabi[1], Azam Marjani[1,*], Sadegh Moradi[2], Mehran Davallo[3] and Saeed Shirazian[4]

[1]Department of Applied Chemistry, Faculty of Science, Islamic Azad University Tehran-North Branch, Tehran, Iran
[2]Department of Chemical Engineering, Arak University, Arak, Iran
[3]Department of Polymer, Faculty of Science, Islamic Azad University, Tehran-North Branch, Tehran, Iran
[4]Research Lab for Advanced Separation Processes, Department of Chemical Engineering, Iran University of Science and Technology, Narmak, Tehran 16846, Iran

*Corresponding author: E-mail: a-marjani@iau-arak.ac.ir

In present studies, the gas separation in hollow-fiber membranes was investigated theoretically and experimentally. Hollow fibers were prepared by melt-extrusion method at speed of 110 m/min. Simulations were performed for gas absorption in liquid absorbents. Simulations were performed for absorption of CO_2 in aqueous solution if K_2CO_3. Moreover, the simulation results were validated with experimental data obtained from literature for physical absorption of CO_2. The simulation results were in good agreement with experimental data. Simulations showed percentage removal of 75 % for CO_2 removal. The results indicated that hollow-fiber membranes have a good performance in the area of gas absorption.

Key Words: Polycarbonate membrane, Melt extrusion, Aqueous solution, Fiber, Simulation.

INTRODUCTION

Hollow-fiber membrane contactors are devices that allow two phases to come into direct contact with each other, for mass transfer without dispersion of one phase into the other. In gas-liquid hollow-fiber membrane contactors, the gas mixture flows in one side of a hydrophobic membrane while the liquid absorbent flows in the other side. A gas-liquid interface will be formed at the adjacent to liquid when the membrane are not wetted with liquid absorbent but filled with gas phase. In hollow-fiber membrane contactors, gases diffuse from gas phase to gas-liquid interface and are absorbed in the solvent. Recently, hollow-fiber membrane contactors have attracted many researchers' attention as a new type of gas absorption[1-3]. In comparison to the conventional tower absorbers, hollow-fiber membrane contactors have some advantages such as larger interfacial area per unit volume, no flooding, foaming, entraining. In addition, energy consumption is lower compared with other gas-liquid contactors. The main purpose of the present study is to prepare and simulate polycarbonate hollow-fiber membrane for absorption of CO_2 in K_2CO_3 aqueous solution.

EXPERIMENTAL

The hollow fibers were made from polycarbonate (Grade PC-175) supplied by CHIMEI-ASAHI Corporation (Taiwan).

Hollow fibers preparation: The hollow fibers were extruded at high pressure through a die (a fine capillary) which was designed and fabricated for this work. The polymer was extruded into cooling water that both cools the fiber and exerts a drag force on the fiber. The air was used as lumen fluid at 25 ºC. At some distance below the spinneret (60 cm), a mechanical roll provides the force that drives the process. Variables in the spinning process were: Extrusion rate: 110 m/min; output rate: 60 g/min; melt temperature: 210 ºC; cooling conditions: water at 20 ºC. Stretching of hollow fibers was carried out at room temperature. The dimensions of obtained hollow fibers are shown in Table-1.

TABLE-1
DIMENSIONS OF HOLLOW FIBER
MADE FROM POLYCARBONATE

Parameter	Value
Fiber inner diameter (μm)	400
Fiber outer diameter (μm)	600
Fiber length (cm)	20

Simulation of gas absorption in hollow-fiber membranes
Mass transfer equations for hollow-fiber membrane: A 2D mass transfer model was used for a hollow fiber. The gas mixture flows with a fully developed laminar parabolic velocity profile. The fiber is surrounded by a laminar liquid flow in an opposite direction. Therefore, the hollow-fiber

membrane contactor consists of three sections: tube side, membrane and shell side. The steady state two-dimensional mass balances were carried out for all three sections.

Equations of tube side: The mass transfer equations are the convection-diffusion equations[4]:

$$\frac{\partial C_i}{\partial t} + \nabla(-D_i \nabla C_i + C_i V) - R_i = 0 \tag{1}$$

where C_i, D_i, R_i, V and t are the concentration, diffusion coefficient of species i, reaction rate of species i, velocity and time, respectively. Velocity distribution in the tube is assumed to follow Newtonian laminar flow[4]:

$$V_{z-tube} = 2u\left[1 - \left(\frac{r}{r_1}\right)^2\right] \tag{2}$$

where u is average velocity in the tube side and r_1 is inner radius of fiber.

Membrane equations: Mass transfer of gas inside the membrane is considered to be due to diffusion alone (V = 0). Boundary conditions are given as:

at $r = r_1$, $C_{G-membrane} = C_{G-tube}$ (3)
at $r = r_2$, $C_{G-membrane} = C_{G-shell/m}$ (4)

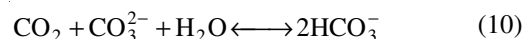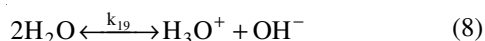where m = physical solubility of gas (CO_2) in solvent.

Equations of shell side: In the shell side, we must solve mass transfer equation with chemical reaction for gas and liquid solvent (potassium carbonate) where gas is absorbed and reacts with solvent. Happel's free surface model can be used to characterize the out fibers velocity profile[5]. The laminar parabolic velocity profile in the outside of fibers is[5]:

$$V_{z-shell} = 2u\left[1 - \left(\frac{r_2}{r_3}\right)^2\right]$$
$$\times \frac{(r/r_3)^2 - (r_2/r_3)^2 + 2\ln(r_2/r)}{3 + (r_2/r_3)^4 - 4(r_2/r_3)^2 + 4\ln(r_2/r_3)} \tag{5}$$

where u, r_3, r_2 are the average velocity, radius of free surface and fiber outer radius, respectively.

Reaction rate for CO_2 absorption in aqueous solution OF K_2CO_3: Reaction rate for carbon dioxide with the aqueous carbonate solutions has been studied by many researchers[6,7]. Their work has been carried out using high bicarbonate to the carbonate ratios. In these cases the carbon dioxide into the aqueous carbonate buffer solutions can be treated as absorption accompanied by an irreversible pseudo-first order reaction. The reaction mechanism under the conditions of low or negligible bicarbonate to carbonate ratio is more complex and contains two-step reversible reactions[6,7].

When the potassium carbonate is dissolved in water it is ionized to K^+ and CO_3^{2-}. The HCO_3^- and OH^- are then generated by the inverse of reaction 7 in following reactions. Various reactions taking place during the absorption of carbon dioxide into the aqueous potassium carbonate solution are given below[6,7].

$$CO_2 + OH^- \xleftarrow{k_{17}} HCO_3^- \tag{6}$$

$$HCO_3^- + OH^- \xleftarrow{k_{18}} CO_3^{2-} + H_2O \tag{7}$$

$$2H_2O \xleftarrow{k_{19}} H_3O^+ + OH^- \tag{8}$$

$$CO_2 + 2H_2O \xleftarrow{k_{20}} HCO_3^- + H_3O^+ \tag{9}$$

$$CO_2 + CO_3^{2-} + H_2O \longleftrightarrow 2HCO_3^- \tag{10}$$

RESULTS AND DISCUSSION

Effect of liquid flow rate on the absorption of CO_2: Fig. 1 shows the percentage removal of CO_2 as a function of liquid flow rate for a constant gas inlet flow rate (100 mL/min). The effect of the concentration of carbonate solution is also shown in the figure. As the concentration of the carbonate solution is decreased, the concentration of the absorbed carbon dioxide is also increased. This indicates that the increase in the concentration of carbonate ions results into the decrease in carbon dioxide absorption.
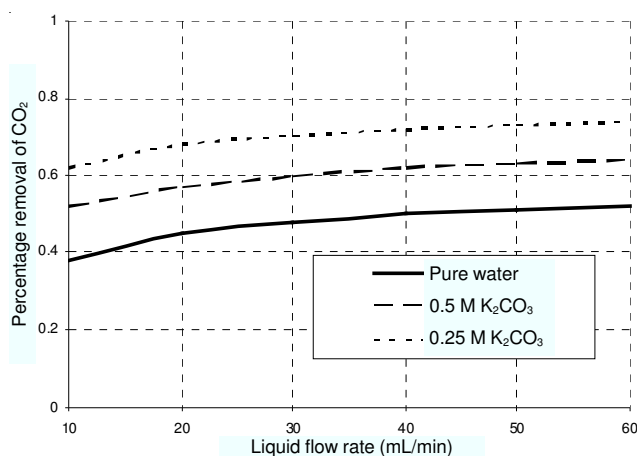

Fig. 1.   Effect of liquid flow rate on the percentage removal of CO_2 for the absorption of CO_2 in K_2CO_3. Gas pressure = 120 kPa; temperature = 297 K; gas flow rate = 100 mL/min

Model validation: In order to validate the mass transfer model developed here, literature data were employed to be compared with the simulation results. As shown in Fig. 2, the simulation results are in good agreement with the literature data[8]. The parameters used in the simulations are the same as reported by Costello *et al.*[8]. Simulation results show an average difference of 8 % with the experimental data.
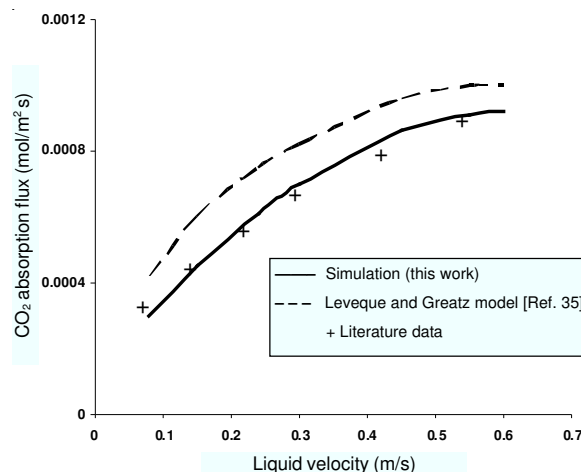

Fig. 2.   Comparison of simulation results with literature data. Fiber inner diameter = 600 μm, fiber length = 38 cm. Gas pressure = 107 kPa, temperature = 295 K

Conclusion

A 2D mathematical model was used to describe absorption of CO_2 in the membrane contactors. The model was based on solving the conservation equations for three sections of contactor. Gas absorption in carbonate aqueous solution of K_2CO_3 was simulated in this work. The simulation results for absorption of CO_2 in liquid solvents indicated that the removal of gas increased with increasing liquid velocity in the shell side. The polypropylene hollow fiber membrane contactor was found to be most efficient in purification of gas streams.

Nomenclature

C   : Concentration (mol/m^3)
C_i : Concentration of any species in the contactor (mol/m^3)
C_i : Concentration of any species (mol/m^3)
C_{inlet} : Inlet concentration of gas in the tube (mol/m^3)
C_{outlet} : Outlet concentration of gas in the tube (mol/m^3)
D_i : Diffusion coefficient of species i (m^2/s)
k   : Reaction rate coefficient of gas with absorbent (m^3/mol s)
m   : Physical solubility (-)
P   : Pressure (Pa)
r_1 : Tube inner radius (m)
r_2 : Tube outer radius (m)
r_3 : Inner shell radius (m)
r   : Radial coordinate (m)
R_i : Overall reaction rate of any species (mol/m^3 s)
T   : Temperature (K)
u   : Average velocity (m/s)
$V_{z\text{-}shell}$: z-Velocity in the shell (m/s)
$V_{z\text{-}tube}$: z-Velocity in the tube (m/s)
z   : Axial coordinate (m)

REFERENCES

1. H. Herzog, B. Eliasson and O. Kaarstad, *Sci. Am.*, **182**, 72 (2000).
2. U. Desideri and A. Paolucci, *Energy Convers. Manag.*, **40**, 1899 (1999).
3. A. Gabelman and S.T. Hwang, *J. Membr. Sci.*, **159**, 61 (1999).
4. R.B. Bird, W.E. Stewart and E.N. Lightfoot, Transport Phenomena, John Wiley & Sons (1960).
5. J. Happel, *AIChE J.*, **5**, 174 (1959).
6. H. Hikita, S. Asai and T. Takatsuka, *Chem. Eng. J.*, **11**, 131 (1976).
7. D. Roberts and P.V. Danckwerts, *Chem. Eng. Sci.*, **17**, 961 (1967).
8. M.J. Costello, A.G. Fane, P.A. Hogan and R.W. Schofield, *J. Membr. Sci.*, **80**, 1 (1993).