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Theoretical and experimental studies of CO_2 absorption by the amine solvent system in parallel-plate membrane contactors

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A B S T R A C T

The absorption efficiency of CO_2 in ethanolamine (MEA) solvent system of a parallel-plate membrane contactor with both concurrent- and countercurrent-flow operations was investigated theoretically and experimentally. A two-dimensional modeling equation for predicting the concentration distribution and total absorption rate was developed, and the analytical solution for the resultant partial differential equations is obtained using the separated variables method with an orthogonal expansion technique. The theoretical predictions of the absorption efficiency, total absorption rate, average Sherwood number and concentration distributions were presented graphically with the mass-transfer Graetz number, inlet CO_2 concentration, and both gas feed and absorbent flow rates as parameters.

1. Introduction

Membrane technology has been extensively applied to liquid/liquid and gas/liquid systems and widely used in many separation processes [1,2] such as gas absorption and metal ions removal due to the advantages of low energy consumption, a large mass transfer area, continuous operations, and the flexibility to scale up. The application of membrane contactor to gas absorption process is aiming to allow the soluble gas mixture components being selectively absorbed in the solvent on membrane surface of liquid phase [3,4]. When applied in CO_2 absorption, previous studies has proved some materials such as PMSQ aerogel, Al_2O_3/SiO_2-FAS used for membrane contactor for CO_2 absorption are durable and reusable [5,6]. The hybrid silica aerogel and highly porous PVDF/siloxane nanofibrous membranes were proved to enhance the CO_2 absorption flux significantly [7,8]. The separation efficiency of membrane gas absorption process depends on the distribution coefficient of gas solute in the two-phase system in which a gradient exists in the compositions of the gas and liquid flowing streams.

Mathematical treatments and experimental work were carried out to study the influences of mass transfer efficiency of CO_2 absorption based on physical absorption [9,10]. A more efficient absorption process is chemical absorption by amine liquid solutions as chemical absorbents with the use of hydrophobic microporous membrane to allow CO_2 being selectively absorbed on the membrane surface of liquid phase. The two-dimensional mathematical formulation of such a coupled boundary value problem in the membrane contactor is referred to as the conjugated Graetz problem associated mutual boundary conditions [11,12], and the analytical solution of such a conjugated Graetz problem was obtained by using the orthogonal technique and separated variables method [13,14]. The purpose of this study is to develop the mathematical formulation for a parallel-plate membrane contactor gas absorption in MEA solvent system and to obtain the solutions analytically then to verify with experimental data. The CO_2 concentration in MEA absorbent stream, absorption rate and absorption efficiency was investigated theoretically and experimentally with the MEA absorbent flow rate, gas mass flow rate and inlet CO_2 concentration in the gas stream as parameters under both concurrent- and countercurrent-flow operations.

2. Theory

A parallel-plate two subchannels flow separated by a membrane gas-liquid contactor with length L, width B, distances between membrane and upper or lower plate are W_a and W_b, respectively, as shown in Fig. 1. The thickness of the hydrophobic microporous membrane is δ and gas feed and absorbent flow were passed through different subchannels. The overall mass transfer process includes three steps. First, the solute gas transfers into the membrane surface from the bulk gas phase. The solute then

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Nomenclature

B	conduit width, m
C	concentration in the stream, mole/m^3
D_a	ordinary diffusion coefficient of CO_2 in N_2, m^2/s
D_b	ordinary diffusion coefficient of CO_2 in MEA solution, m^2/s
d_{mn}	coefficient in the eigenfunction $F_{a,m}$
E	the accuracy of the experimental results
e_{mn}	coefficient in the eigenfunction $F_{b,m}$
F_m	eigenfunction associated with eigenvalue λ_m
Gz	mass-transfer Graetz number
H	dimensionless Henry's law constant
I_M	absorption efficiency
J	average absorption flux, mol/s · cm^2
K	overall mass transfer coefficient defined
k	mass transfer coefficient
k_ξ	local mass transfer coefficient of CO_2
L	conduit length, m
N_{exp}	the number of experimental measurements
N_{rep}	the number of experimental measurements repeated
N_{sam}	the number of analytical and experimental data compared samples
Q	volumetric flow rate of conduit, m^3/s
S_m	expansion coefficient associated with eigenvalue λ_m
Sh_ξ	local Sherwood number
$\overline{Sh}$	average Sherwood number
v	velocity distribution of fluid, m/s
$\bar{v}$	average velocity of fluid, m/s
W	distance between two parallel plates, m
x	transversal coordinate, m
z	longitudinal coordinate, m

Greek letters

δ	thickness of the membrane, m
ε	porosity of the membrane
η	dimensionless transversal coordinate, x/W
λ_m	eigenvalue
ξ	longitudinal coordinate
ω	absorption rate, mol/s
ψ	dimensionless concentration

Subscripts

a	in the gas feed flow channel
b	in the liquid absorbent flow channel

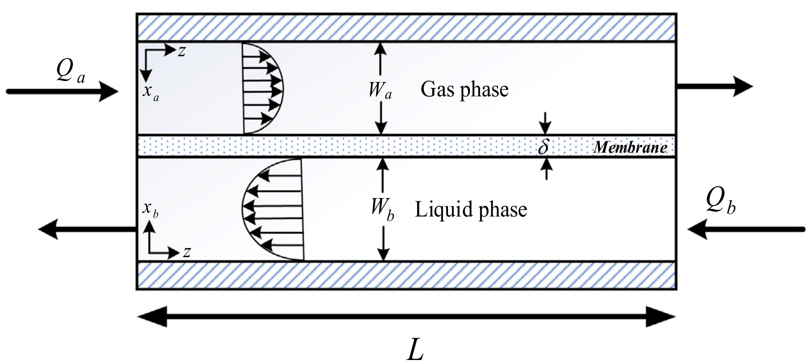

Fig. 1. Parallel-plate membrane gas-liquid contactor.

diffuses through the membrane pores. Finally, it transfers into the bulk liquid via the membrane–liquid interface. The velocity distributions and the conservation equations of mass may be described once the following assumptions were made: (a) steady state and fully developed flow in each subchannel; (b) negligible axial diffusion and conduction; (c) isothermal operation and constant physical properties; (d) the applicability of Henry's law; (e) the chemical reaction is very fast and the equilibrium state is reached.

2.1. Countercurrent-flow operation

The velocity distributions and the dimensionless equations of mass transfer for each subchannel may be written in terms of the dimensionless variables as

$$v_a(\eta_a) = \bar{v}_a(6\eta_a - 6\eta_a^2) \tag{1}$$

$$v_b(\eta_b) = -\bar{v}_b(6\eta_b - 6\eta_b^2) \tag{2}$$

and

$$\frac{\partial^2\psi_a(\eta_a,\xi)}{\partial\eta_a^2} = \left(\frac{W_a^2 v_a(\eta_a)}{LD_a}\right)\frac{\partial\psi_a(\eta_a,\xi)}{\partial\xi} \tag{3}$$

$$\frac{\partial^2\psi_b(\eta_b,\xi)}{\partial\eta_b^2} - K'\psi_b(\eta_b,\xi) = \left(\frac{W_b^2 v_b}{LD_b}\right)\frac{\partial\psi_b(\eta_b,\xi)}{\partial\xi} \tag{4}$$

where

$$\bar{v}_a = \frac{Q_a}{W_aB},\quad \bar{v}_b = \frac{Q_b}{W_bB},\quad \eta_a = \frac{x_a}{W_a},\quad \eta_b = \frac{x_b}{W_b},\quad \xi = \frac{z}{L},$$
$$\Delta = \frac{W_a}{W},\quad W_b = (1-\Delta)W,\quad W = W_a + W_b,$$
$$Gz_a = \frac{Q_aW}{D_aBL},\quad Gz_b = \frac{Q_bW}{D_bBL} \tag{5}$$

The boundary conditions required for solving Eqs. (3) and (4) are

$$\frac{\partial\psi_a(0,\xi)}{\partial\eta_a} = 0 \tag{6}$$

$$\frac{\partial\psi_b(0,\xi)}{\partial\eta_b} = 0 \tag{7}$$

$$-\frac{\partial\psi_a(1,\xi)}{\partial\eta_a} = \left(\frac{W_a\varepsilon}{\delta}\right)\left[\psi_a(1,\xi) - \frac{K'_{ex}}{H}\psi_b(1,\xi)\right] \tag{8}$$