

Analysis of CO₂ separation and simulation of a partially wetted hollow fiber membrane contactor

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Received 23 April 2007; received in revised form 30 July 2007; accepted 31 July 2007

Available online 7 August 2007

Abstract

A steady state model was developed for a microporous hollow fiber membrane contactor operated under partially wetted conditions accompanied by chemical reactions, to analyze CO₂ absorption into the aqueous solution of diethanolamine (DEA). The proposed diffusion-reaction model contains reversible chemical reactions in the liquid bulk as well as wetted parts of the membrane pores. A numerical scheme was employed to solve the simultaneous nonlinear mathematical expressions, and the results were validated with experimental data in the literature. The gas phase concentration and velocity profile in axial direction inside the shell, liquid concentration profile in axial and radial directions inside the fibers, and also those within the wetted parts of the pores were predicted by using the model. The results of the model and proposed numerical scheme show that membrane wetting, even in very low fractions, can decrease the absorption flux significantly. The wetting fraction of membrane was predicted both with and without consideration of chemical reactions inside the wetted pores. The results indicate that the chemical reactions inside the wetted pores, which have been disregarded in the literature, have considerable effects on the prediction of membrane wetting fraction.

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Keywords: Membrane contactor; Hollow fiber; Gas absorption; Partial wet

1. Introduction

Carbon dioxide is one of the major contributors to the greenhouse effect. There is an inexorable trend toward limiting anthropogenic emissions of carbon dioxide and other gases suspected of causing global climate change. Furthermore, the present interest in energy conservation and pollution control has led to search for more efficient and economical methods of CO₂ removal.

The application of microporous hollow fiber membrane contactor for gas absorption and stripping has gained considerable attention recently and still is a relatively new concept. In comparison with most membrane separation processes which use a dense selective layer on one side of the fibers, the microporous membrane used in membrane contactor is not selective. Instead of the selective layer, a liquid is flowed in one side of the fiber, which can absorb gas components physically or chemically. It leads to a higher mass transfer rate due to lower

membrane resistance. Additionally, because there is no selective layer, a very small pressure drop across the membrane is required for mass exchange. Compared to the traditional columns like packed and tray towers, bubble columns, venture scrubbers and spray towers, membrane contactor has several advantages. These include higher mass transfer rates, independent control of gas and liquid rates, known and constant interfacial area, easy scale up and no operational problems such as foaming, flooding and entrainment [1,2]. By forming the membranes as hollow fibers, a very compact unit can be made with a specific area 1600–6500 (m² m⁻³), which is higher than 30–330 (m² m⁻³) offered by packed/tray towers and 160–500 (m² m⁻³) for mechanically agitated columns [1]. Although the membrane wall introduces an additional resistance, which does not exist in conventional towers, the higher surface area in this type of modules offers much higher mass transfer rates.

These advantages have led to a number of investigations on the use of membrane contactors for gas absorption and stripping. Absorption of various gases by a number of solvents in a hydrophobic membrane were first carried out by Zhang and Cussler [3,4]. Kreulen et al. [5,6] studied the CO₂ absorption

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Nomenclature

A_s	shell side cross-section area available for gas (m^2)
A_w	gas–liquid mass transfer area (m^2)
C	concentration ($mol\ m^{-3}$)
C_g	total gas concentration ($mol\ m^{-3}$)
C_{ig}	component gas concentration ($mol\ m^{-3}$)
C_T	total amine concentration ($mol\ m^{-3}$)
d_h	hydraulic diameter of shell side (m)
d_i	inner diameter of fiber (m)
d_o	outer diameter of fiber (m)
D	diffusivity ($m^2\ s^{-1}$)
D_{ik}	Knudsen diffusivity of species i ($m^2\ s^{-1}$)
H	Henry's constant
k_{-1}	reverse first order rate constant (s^{-1})
k_2	second order reaction rate constant ($m^3\ mol^{-1}\ s^{-1}$)
k_b	second order reaction rate constant for base b ($m^3\ mol^{-1}\ s^{-1}$)
K_{eq}	reaction (3) equilibrium constant ($mol\ m^{-3}$)
K_{eq1}	reaction (7) equilibrium constant ($mol\ m^{-3}$)
K_{eq2}	reaction (8) equilibrium constant
K_{ex}	external mass transfer coefficient ($m\ s^{-1}$)
K_g	gas phase mass transfer coefficient ($m\ s^{-1}$)
K_m	membrane mass transfer coefficient ($m\ s^{-1}$)
L	module length (m)
M	molecular weight ($kg\ kmol^{-1}$)
P	pressure (Pa)
Q	volumetric flow rate ($m^3\ s^{-1}$)
r	radial direction or radius (m)
r_i	inner radius of fiber (m)
R	universal gas constant ($J\ mol^{-1}\ K^{-1}$)
R_i	chemical reaction rate ($mol^{-1}\ m^3\ s^{-1}$)
Re	Reynolds number
Sc	Schmit number
Sh	Sherwood number
T	temperature (K)
U	velocity ($m\ s^{-1}$)
x	wetting fraction
Z	axial coordinate (m)

Greek symbols

δ	membrane thickness
ε	membrane porosity
τ	tortuosity

Subscripts

A	carbon dioxide
B	amine
g	gas
i	any diffusing species
in	input
lm	log-mean average
L	liquid
p	pore
w	gas–liquid interface in the liquid filled pores

by water/glycol mixture as absorbent. Their results showed a better performance of membrane contactor compared to bubble column especially for higher viscous liquids. Karoor and Sirkar [7] studied absorption of CO_2 and SO_2 by water in a comprehensive experimental investigation and found that mass transfer rate is about ten times higher than those typically obtained in packed towers. Some other experimental and theoretical studies have been carried out focusing on CO_2 absorption from flue gases using various absorbents such as sodium hydroxide, potassium carbonate and alkanolamines [8–11]. The advantages of chemical absorption of CO_2 by alkanolamine or mixed alkanolamine solutions are quite obvious, since alkanolamines are weak basic compounds and their chemical bonds with CO_2 are easily broken at high temperatures, leading to efficient regeneration of the absorbents and as a result, can be recovered efficiently [12–16].

In most of these researches, hydrophobic membranes have been employed with gas filled membrane pores. In a hydrophilic fiber, pores are filled with a stagnant liquid film and mass transfer rate is much lower due to lower diffusivity of CO_2 in liquid. Therefore, the liquid phase pressure in a hydrophobic membrane should not exceed breakthrough pressure of the membrane to prevent membrane wetting. On the other hand, the pressure of liquid should be slightly higher than gas phase pressure to prevent dispersion of gas bubbles into the liquid.

Partial wetting can also occur when a hydrophobic membrane such as polypropylene (PP) or polyvinylidene florid (PVDF) is employed for gas absorption or stripping. Capillary condensation of water vapor in the pores, as well as pressure difference between shell side and lumen side, can be possible reasons for partial wetting [17,18]. The presence of organic compounds in the liquid phase, such as amine solutions, can decrease the surface tension of liquid and cause partial wetting. Some inorganic species, complexes, microorganisms or even traces of impurities can have the same effects [19,20]. Mahmud et al. [17,18] and Evren [21] studied the stripping of air from water and stated that the theory and experiments do not agree well in some cases. They attributed this disagreement to the partial wetting and a good fit was searched by changing the wetting fraction in the model. Such disparity between theory and experimental results has also been observed when a chemical reaction occurs in the liquid phase [22–24]. In the recent case, the partial wetting has been assumed again, but the same procedure of Mahmud et al. [17] was applied to search the wetting fraction, without consideration of chemical reaction in the liquid filled pores.

Some researchers try to overcome membrane wetting by using new absorbents or new membrane types. Kumar et al. [25] proposed a new chemical absorbent based on amino acid salts for CO_2 absorption with no wetting effect on polyolefin fibers. Recently, Yan et al. [26] employed aqueous potassium glycinate solution, which has the same influence. deMontigny et al. [27] compared the performance of polypropylene and polytetrafluoroethylene (PTFE) fibers using amine solutions and found that polypropylene membranes suffer a loss in performance over time, while PTFE membranes maintain their initial level of performance. It was attributed to the higher wetting resistance of PTFE compare to PP in amine solutions.

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