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Mathematical modeling for the simultaneous absorption of CO_2 and H_2S using MEA in hollow fiber membrane contactors

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ABSTRACT

A comprehensive two-dimensional mathematical model was developed for the simultaneous transport of carbon dioxide and hydrogen sulfide through hollow fiber membrane (HFM) contactors while using monoethanol amine (MEA) as the chemical solvent. The model considered non-wetting and partial-wetting conditions where the gas mixture and the solvent fill the membrane pores for countercurrent gas–liquid flow arrangement. Axial and radial diffusion were considered inside the fiber, through the membrane, and within the shell. The model was validated for physical and chemical absorption of CO_2 using water and MEA, respectively. The model results were in excellent agreement for the physical absorption while considering non-wetting conditions. However, for chemical absorption the model showed excellent agreement with the experimental data while considering 10% wetting for polypropylene (PP) when 0.005 M MEA was used, and 50% wetting for polyvinyllidene fluoride (PVDF) when 2 M MEA was used. The effect of MEA concentrations, gas and liquid velocities were studied on the simultaneous removal of CO_2 and H_2S. The % removal of CO_2 increased while increasing the MEA concentration. As for H_2S, low concentration of MEA was efficient in complete removal. The % removal of CO_2 decreased while increasing gas velocity, whereas, H_2S % removal did not change with increasing gas velocity when operating at low gas velocity. However, the effect is more pronounced while operating at high gas velocities. Both CO_2 and H_2S % removal was increased with increasing the liquid velocity. CO_2% removal increased slightly, while H_2S % removal increased substantially while operating at high gas velocity.

1. Introduction

Global warming and climate changes are believed to be caused by the rise of the greenhouse gases emissions, because of the global expansion of industrial activities over the past three decades [1]. Carbon dioxide is representing about 80% of greenhouse gases. It is reported that half of the CO_2 emissions are produced by industry and power plants using fossil fuels [2]. These emissions create the need for low energy-consumption, and efficient technologies for the capture and removal of CO_2 from gas mixtures produced by industrial sources. Another important gas produced in these industrial and domestic processes is Hydrogen Sulfide. H_2S is a highly toxic and corrosive gas and is considered as one of the major sources of the environmental problems such as acid rains. Usually natural gas refinery streams contain H_2S and CO_2 as major impurities. Therefore, in order to utilize these fuels for chemical processing or energy generation, H_2S and CO_2 must be removed.

Both H_2S and CO_2 are acidic in nature and are similar in many physical and chemical aspects. Hence the processes used for the removal of CO_2 in gas treating processes also absorb H_2S. The most common processes for removal of these acid gases are an absorption into a solvent using conventional gas–liquid contactors such as packed or plate absorption towers. Simultaneous absorption of H_2S and CO_2 using packed towers has been extensively studied, both experimentally and theoretically [3]. Using alkaline solution as the absorption medium, H_2S selectivity was reported to be in the range of 10–30 [4]. However, these conventional chemical absorption processes for the removal of H_2S and CO_2 suffer many drawbacks such as flooding, foaming, entraining, channeling, and high capital and operating costs. Therefore, many researchers have looked for new technologies to enhance the efficiency of these processes. Hollow fiber membrane (HFM) contactors attracted the attention of many researchers as a new technology for gas separation [5–10].

Membrane contactors are devices that allow two fluids to come into direct contact with each other, for the purpose of mass transfer without dispersion of one phase into the other. In the membrane contactor, the gas mixture flows in one side of a hydrophobic microporous membrane while the liquid absorbent flows in the other side. A gas–liquid interface will be formed at the pores opening adjacent to the liquid when the membrane pores are not wetted

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with liquid absorbent but filled with gas. In the absorption module based on microporous fibers, gases diffuse from the gas phase to the gas–liquid interface through the pores and are absorbed in the liquid. The rich solution may be sent to another membrane contactor for stripping to remove the absorbed acid gases. The lean solution is then recycled to the absorption module. This type of process offers several practical advantages including high surface area per unit contactor volume, independent control of gas and liquid flow rates without any flooding, loading, weeping, or foaming, known gas–liquid interfacial area, modularity, capably linear scale-up, low corrosion problems, low operation costs and capital cost [11]. Membrane gas absorption has been considered to be a promising and potential large-scale application technology for the recovery and removal of CO_2 [12].

Much work has been done to study the removal of H_2S and CO_2 using amine solutions such as DEA, and MDEA, and a mixture of both. Mandal et al. [13] studied the selective removal of H_2S from a gas mixture containing CO_2 and H_2S using MDEA and AMP aqueous solutions in a wetted column. Haimour and Sandall [14] modeled the simultaneous absorption of H_2S and CO_2 in DEA using penetration theory. Say et al. [15] have presented a process development work using hindered amine as the promoter of hot carbonate solutions for simultaneous removal of H_2S and CO_2. Rascol et al. [16] numerically interpreted the simultaneous mass transfer of CO_2 and H_2S into aqueous blends of MDEA and DEA. They have used the film theory to describe the mass transfer phenomena within the liquid phase. Rinker [17] studied theoretically and experimentally the simultaneous absorption of CO_2 and H_2S into blends of MDEA and DEA. Al-Marzouqi et al. [18] studied CO_2 absorption using MEA and NaOH in hollow fiber membrane contactors. Atchariyawut et al. [19] used MEA and NaOH as the absorbent solvents for CO_2 absorption using Membrane contactors. They both concluded that NaOH and MEA wet the prospective membranes used in their experiments.

However in spite of the immense commercial significance of single amines, or aqueous blended amine solvents for simultaneous removal of CO_2 and H_2S from sour natural gas streams, studies on simultaneous removal of CO_2 and H_2S using MEA in HFM contactors have not been reported in the literature so far. The objective of this paper is to develop and solve a comprehensive 2D mathematical model for simultaneous removal of CO_2 and H_2S using MEA in hollow fiber membrane contactors. Because of the lack of literature on the experimental data for simultaneous removal of CO_2 and H_2S using MEA, the model will be validated for physical and chemical absorption of CO_2 using water and MEA respectively for the case of non-wetting and partial-wetting conditions. Once the model has been validated, the effect of MEA concentration, gas and liquid velocities on the simultaneous absorption of CO_2 and H_2S will be studied.

2. Model development

In this work, a comprehensive 2D mathematical model for the transport of both CO_2 and H_2S through membrane contactor using MEA as the stripping solvent has been developed. Axial and radial diffusion is considered in the tube, shell compartments, and within the membrane as well. The model is flexible to account for non-wetting, partial wetting, and complete wetting. However to obtain the best efficiency for the simultaneous removal of CO_2 and H_2S using MEA, non-wetting conditions are considered. Non-wetting conditions can be obtained in real situations by using polytetrafluoroethylene (PTFE) hollow fiber membranes as suggested by several authors because it has high resistance for membrane wetting [20,10]. Kim and Yang [21] used MEA as the absorbent solvent for CO_2 removal while using (PTFE) hollow fiber membranes. His model was in good agreement with the experimental data, when non-wetting conditions were assumed.

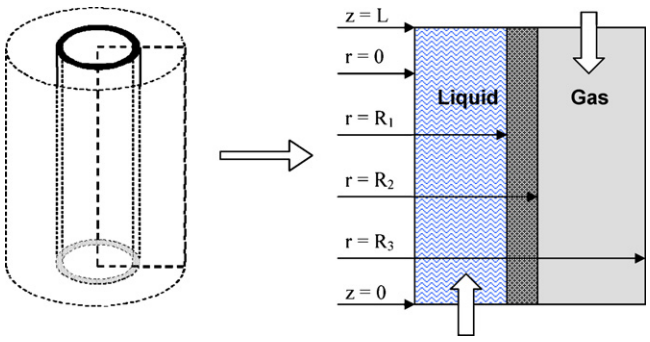

Fig. 1. A schematic diagram for the membrane contactor used for modeling.

2.1. Material balance

A material balance has been carried out on a shell-and-tube membrane contactor system to develop the main equations for the mathematical model. The membrane contactor consists of three sections: tube side, membrane, and shell side. The gas mixture (CO_2, H_2S, and CH_4) flows through the shell side, while the solvent flows through the tube side in a countercurrent arrangement. The steady state two-dimensional material balances are carried out for all three sections. The gas mixture is fed to the shell side (at $z = L$), while the solvent is passed through the tube side (at $z = 0$). Carbon dioxide and hydrogen sulfide are removed from the mixture by diffusing through the membrane and then absorbing/reacting with the solvent.

The model is developed for a segment of a hollow fiber, as shown in Fig. 1, through which the solvent flows with a fully developed laminar parabolic velocity profile. The fiber is surrounded by a laminar gas flow in an opposite direction to that of the liquid. Fig. 2 shows the cross sectional area of the membrane contactor. Based on Happel's free surface model [22], only portion of the fluid surrounding the fiber is considered which may be approximated as circular cross section. Thus, symmetry may be considered at the outer portion of the fluid surrounding the fiber (at $r = R_3$ in Fig. 1).

2.1.1. Shell side

The continuity equation for each species during the simultaneous mass transfer and chemical reaction in a reactive absorption system can be expressed as

$$\frac{\partial C_i}{\partial t} = -\nabla \cdot N_i + R_i \tag{1}$$

where C_i, N_i, and R_i are the concentration, flux, and reaction rate of species i along the length of the membrane, respectively. Fick's law of diffusion can be used for the determination of fluxes of species i as follows:

$$N_i = -D_i \nabla C_i + C_i V_z \tag{2}$$

where D_i and V_z are the diffusion coefficient of species i and the axial velocity along the length of the module, respectively. Combining Eqs. (1) and (2) gives the overall mass balance which includes the

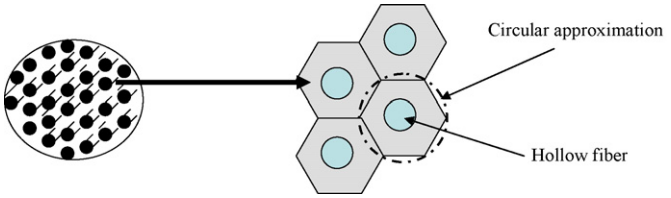

Fig. 2. Cross-sectional area of the membrane contactor and circular approximation of portion of the fluid surrounding the fibers.