

# Studying the effects of Gas and Liquid velocities in modeling of the simultaneous removal CO<sub>2</sub> and H<sub>2</sub>S using MEA in a hollow fiber membrane contactor

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#### ABSTRACT

A two-dimensional mathematical model was studied for the transport of  $CO_2$  and  $H_2S$  through hollow fiber membrane (HFM) contactors while using MEA (monoethanolamine) as the chemical absorber. The model considered non-wetted where the gas and the solvent liquid fill the membrane pores for counter current gas-liquid flow direction. There is an axial and radial diffusion in the tube, through the membrane and the shell. The model validation was on the physical and chemical absorption of CO<sub>2</sub> using water and MEA, respectively. The percentage of removal of CO2 increased while increasing the absorbent velocity. As for H2S, low flow rate of amine was efficient in complete removal. The percentage removal of CO2 decreased while increasing gas velocity because of lower hold up time, whereas H<sub>2</sub>S removal did not change with increasing gas velocity at low gas velocities. However the effect is more obvious at high gas velocities.

Keywords: CO<sub>2</sub>; Natural gas; H<sub>2</sub>S; MEA

## **1. Introduction**

 $CO_2$  is considered as a major greenhouse gas that causes global warming and lowers the efficiency of several industrial processes. On the other hand,  $H_2S$  is a poisonous gas and

highly toxic and flammable that causes major health concerns upon contact. Several technologies for natural gas treatment in the petrochemical industry include absorption towers and packed and plate columns. Although these conventional processes are being used up to date, these methods possess many disadvantages such as flooding foaming and demand high capital and operating costs [1,2]. In a typical hollow fiber membrane contactor device, a considerable number of hollow fibers membranes are assembled together in a large tube where two phases come into a direct contact with each other for the purpose of mass transfer without the dispersion of one phase into another due to the presence of a very fine barrier (membrane).Membrane contactors offer several practical advantages including high surface area per unit contactor volume, independent control of gas and liquid flow rates, no flooding or foaming formation occurring, and low operation and capital costs[3]. The objective of this paper is to develop and solve a comprehensive 2D mathematical model for simultaneous removal of CO2 and H2S using MEA in hollow fiber membrane contactors.

#### 2. Experimental

#### 2.1. Materials and method

We have studied a 2D mathematical modelling for the removal of both  $CO_2$  and  $H_2S$  through membrane contactor using MEA as the chemical solvent. There is an axial and radial diffusion in the tube, shell, and the membrane as well. The model is useable for none, partial, and complete wetted. A material balance has been used on a shell-and-tube membrane contactor system to develop the main equations for the mathematical model. The system is made of three sections: tube, membrane, and shell side. The gas stream (CH<sub>4</sub>, H<sub>2</sub>S, and CO<sub>2</sub>) flows through the shell side, while the liquid flows through the tube in a counter-current direction. The gas stream is entered to the shell side (at z = L), while the solvent is entered through the tube side (at z = 0).CO<sub>2</sub> and H<sub>2</sub>S are removed

from the gas stream by diffusing through the membrane and then absorbing with the amine. We can see the model development for a segment of a hollow fiber in Figure1, which the solvent flows with a fully- developed laminar parabolic velocity profile.



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

### 3. Results and discussion

1 molar MEA was used as a solvent i the tube side while the gas mixture (10% CO<sub>2</sub>, 10% H<sub>2</sub>S, and 80% CH<sub>4</sub>) was sent through the shell side in a counter-current direction. The model equations with the boundary conditions were solved for different gas, liquid flow rates, and MEA concentrations. The temperature was set at 298 K, and 1 atm pressure. The objective is to study the effect of different gas, and liquid velocities on the removal of CO<sub>2</sub> &amp; H<sub>2</sub>S. The concentration gradient and the total flux vectors of CO<sub>2</sub> in the tube, membrane and shell sides of the contactor are shown in Figure 2. The gas mixture streams in the shell side at (z = L) where the concentration of CO<sub>2</sub> is the highest 20 = 4 mol m-3, while MEA flows in the tube side at (z = 0)



**Fig. 2**: Model solution for CO2.  $r_1 = 0.11$  mm,  $r_2 = 0.15$  mm,  $r_3 = 0.265$  mm, L = 22 cm, CCO20 = 4 mol where CO2 has a concentration amount of zero. The gas in the shell side moves to the membrane due to the concentration difference, and then is absorbed by the MEA in the tube. The steady state material balance for the transport CO2 and H2S in the shell side, may be written as Assuming Happel's free surface model [4].

Table 1: Membrane module dimensions, properties, and model parameters for CO2-H2S-MEA system.

Parameter	Value	Reference
Inner tube diameter (mm)	0.22	-
Outer tube diameter (mm)	0.3	-
Inner shell diameter (mm)	0.529	-
Module length L (cm)	22	-
Number of fibers	3600	-
$D_{CO_n-shell}$ (m <sup>2</sup> s <sup>-1</sup> )	$1.8 \times 10^{-5}$	Cussler [36]
$D_{co_n-tube}$ (m <sup>2</sup> s <sup>-1</sup> )	$1.51 \times 10^{-9}$	Paul et al. [37]
$D_{CO_2-membrane}$ (m <sup>2</sup> s <sup>-1</sup> )	$D_{\rm CO_2-shell}(\varepsilon/\tau)$	Calculated
$D_{H_2S-shell}$ (m <sup>2</sup> s <sup>-1</sup> )	$2.01 \times 10^{-3}$	Cussler [36]
$D_{H_2S-tube}$ (m <sup>2</sup> s <sup>-1</sup> )	$1.52 \times 10^{-9}$	Estimated
$D_{H_2S-membrane}$ (m <sup>2</sup> s <sup>-1</sup> )	$D_{H_2S-shell}(\varepsilon/\tau)$	Calculated
$D_{\text{MEA-tube}}$ (m <sup>2</sup> s <sup>-1</sup> )	$9.32 \times 10^{-10}$	Paul et al. [37]
$D_{\text{MEA}^+-\text{tube}}$ (m <sup>2</sup> s <sup>-1</sup> )	DMEA-tube	Assumed
$D_{\text{MEACOO}-tube}$ (m <sup>2</sup> s <sup>-1</sup> )	D <sub>MEA-tube</sub>	Assumed
$D_{HS^{-}-tube}$ (m <sup>2</sup> s <sup>-1</sup> )	DH-S-tube	Assumed
$m_{CO_2}$ (mol mol <sup>-1</sup> )	0.8	Paul et al. [37]
$m_{\rm H,S}$ (mol mol <sup>-1</sup> )	2.3	Estimated
$\varepsilon$ (porosity)	0.4	-
τ (tortuosity)	2	-

Fig. 3 shows the concentration gradient of H2S in the tube, membrane and shell sides of the system. H<sub>2</sub>S is consumed very quickly in the first 20% of the module length. This is because of the higher solubility of H2S in MEA (mH<sub>2</sub>S = 2.3) and the instant reaction with



MEA. The flux vectors in the shell side also disappear after 20% of the module length.

Fig. 3: Model solution for H2S.  $r_1 = 0.11$  mm,  $r_2 = 0.15$  mm,  $r_3 = 0.265$  mm, L = 22 cm, CH2S0 = 4 mol m<sup>-3</sup>, CMEA0 = 1000 mol m<sup>-3</sup>,  $V_{Gas}$ = 3.33 m s<sup>-1</sup>,  $V_{Liq} = 0.67$  m s<sup>-1</sup>.

We can see the deduction in absorption of CO2 by increasing the velocity of gas through the shell side in Figure 4, but it is evident that the increase of gas velocity almost doesn't influence the removal of h2s and all of the H2S is being absorbed through the shell side at considered situations.



Fig. 4: Removal of CO<sub>2</sub> and H<sub>2</sub>S in different gas velocities.

It is obvious that we have a rise of removal efficiency for  $H_2S$  by increasing the speed of liquid in Figure 5, but it is not important to raise the absorption of  $H_2S$  because it is fully absorbed even in lower liquid velocities.



Fig. 5: Removal of CO<sub>2</sub> and H<sub>2</sub>S in different liquid velocities.

### 4. Conclusions

A 2D mathematical model was developed for the simultaneous transport of  $CO_2$  and  $H_2S$  in HFM contactors using MEA as chemical absorber. The model assumed to be non-wetted for countercurrent gas– liquid flow direction. Axial and radial diffusion were considered in the fiber, membrane and the shell. We had no experimental data for validation, so the model was validated for physical and chemical absorption of CO using water and MEA, respectively. The model results have an excellent agreement for physical and chemical absorption while considering non-wetted and partial-wetted conditions, respectively. After the model was validated, the effect of gas and liquid flow rates were studied on the efficiency of simultaneous removal of  $CO_2$  and  $H_2S$ . The % removal of  $CO_2$  increased while increasing the MEA flow rate. But for  $H_2S$ , low flow rate of MEA was more efficient in complete removal. The percentage of 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.

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