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DOI:
[10.3311/PPch.8689](https://doi.org/10.3311/PPch.8689)

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Document Version
Publisher's PDF, also known as Version of record

Citation for published version (Harvard):
Motahari, K, Barati, S & Miri, T 2016, 'Mathematical modelling and simulation of carbon dioxide absorption from N₂ using hollow fiber membrane contactor', *Periodica Polytechnica: Chemical Engineering*, vol. 60, no. 4, pp. 266-272. <https://doi.org/10.3311/PPch.8689>

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Mathematical Modelling and Simulation of Carbon Dioxide Absorption from N₂ Using Hollow Fiber Membrane Contactor

Kazem Motahari¹, Sara Barati^{1*}, Taghi Miri¹

Received 18 October 2015; accepted after revision 22 January 2016

Abstract

In this work, hollow fiber membrane contactor was used to theoretically study absorption carbon dioxide using water. Governing equations were solved by finite element method. The impact of temperature and velocity of liquid phase and velocity of gas phase on CO₂ absorption flux has been investigated. Results showed that by increasing liquid phase velocity, CO₂ absorption flux increases remarkably. Also, CO₂ absorption flux increases only 6 % by increasing gas phase velocity from 0.02 to 0.06. Thus, liquid phase is the controller of mass transfer in gas absorption process. The results also showed that CO₂ absorption flux decreases by increasing liquid phase temperature, due to the reduction of gas solubility in liquid phase, and outlet CO₂ concentration of liquid phase decreases too. To validate of model's results, the data of two experimental works have been used.

Keywords

Hollow fiber, membrane, CO₂ absorption, mathematical model

1 Introduction

The combustion of fossil fuels in power plants worldwide has been responsible for CO₂ emission, and thus it leads to global climate change. In addition, operational and economic problems are the consequences of the presence of CO₂ in natural gas. Hence, several technologies have been developed for CO₂ capture from the gas streams including absorption, adsorption, cryogenic distillation and membrane separation [1, 2]. However, the technologies with high removal efficiency that can reduce overall environmental and economic impacts have the priority. For decarbonation, i.e., to remove CO₂, industries typically employ measures such as aeration, forced draft degasification, and vacuum degasification.

Due to using of heavy-duty tools such as pumps and blowers, applying of these traditional methods costs a lot [3]. So, gas-liquid hollow fiber membrane contactor can be used as a favourable alternative for the CO₂ absorption/stripping processes. In this system, the porous membrane acts as a fixed interface between the gas and liquid phases to avoid dispersing one phase into another. Qi and Cussler were the first who examined these devices, and after that lots of researches have been done about hollow-fiber membrane contactors [4-6]. The mechanism of CO₂ absorption is shown in Fig. 1.

Kreulen et al. [7] investigated the chemical absorption of CO₂ by water/glycerol mixtures in a hollow fiber membrane contactor. Qi and Cussler [6] investigated evolution of a theory of the operation of HFMCs, and estimated mass transfer coefficients in liquid phase. Also, they achieved the overall mass transfer coefficients, which include resistances in both liquid and membrane, and showed the differences of the acting of hollow fibers with that of packed towers [8]. Zhang et al. [9] numerically studied the chemical absorption of CO₂ from flue gas in a gas-liquid hollow fiber membrane contactor, using a mixture of MDEA/PZEA. The result showed that increasing membrane length, fibers number, and porosity, has a positive effect on CO₂ absorption.

Masoumi et al. [10] theoretically compared CO₂ absorption by monoethanolamine (MEA), diethanolamine (DEA), methyl diethanolamine (MDEA) and DEAB solutions in a hollow fiber membrane contactor. Results showed that absorption percentage

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by DEAB solution is lower than MEA solution. Kim and Yang [11] theoretically and experimentally conducted the separation of CO₂/N₂ mixtures using HFMCs. Although there was a good agreement between experimental and modelling results, but researchers assumed a linear decrease of gas velocity for the modelling purposes [8]. Therefore, considering the importance of separating and the good agreement observed between experimental results and simulations of membranes [12, 13], a mathematical model is needed that can provide the public simulation of physical and chemical absorption in hollow fiber membrane contactors. The main objective of this work is to develop a two-dimensional mathematical model to absorb carbon dioxide in hollow fiber membrane. The model equations (equations of mass and momentum transfer) are solved through a numerical method based on finite element method. The model results were validated with the experimental results reported by Mansurzadeh et al. [14, 15]. In this work, physical absorption for „non-wetted mode”, where the membrane pores are full of gas, are considered. Physical absorption is regarded as absorption of CO₂ in water.

After ensuring the correctness of results of the model, the effects of various parameters such as liquid temperature, gas and liquid phase velocity on mass transfer of carbon dioxide were investigated. Physical absorption was considered here for CO₂, and the concentration distribution in the membrane contactor was predicted.

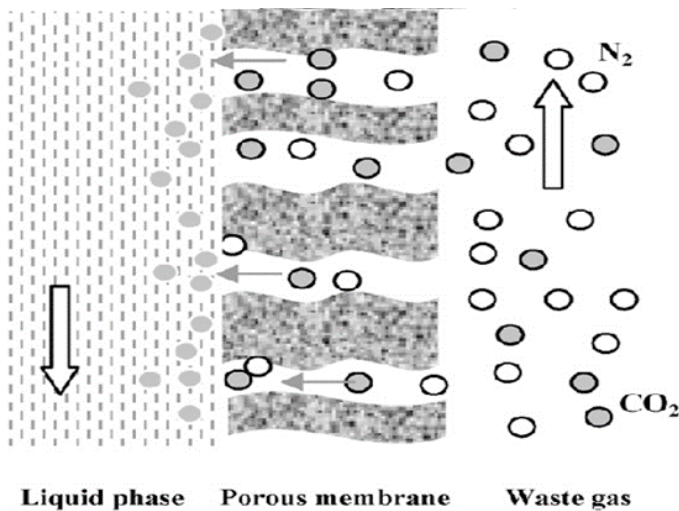


Fig. 1 Schematic of CO₂ absorption mechanism in gas-liquid membrane contactors [15]

2 Theory

A 2D mathematical model was developed to predict mass transfer of CO₂ through the hollow fiber membrane contactors in physical absorption process. In this paper, the separation of CO₂ from CO₂/N₂ gas mixture using water as absorbent in a HFMC has been studied. The liquid stream containing pure water was fed inside the lumen (tube)-side to the bottom of the module and flowed upwards and the gas stream was fed through the shell-side in counter-current manner.

The following assumptions have been considered in the model (1) incompressible fluids; (2) laminar flow; (3) non-wetted condition; (4) steady state and isothermal condition; (5) constant tube-side pressure (6) constant physical properties of the fluid; and (7) Henry's law was applicable for gas-liquid interface.

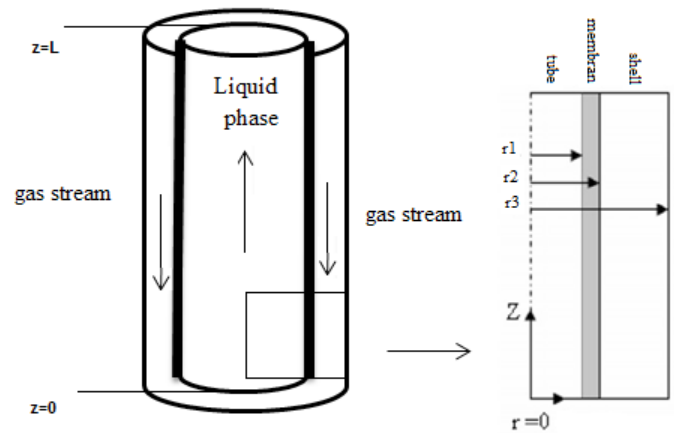


Fig. 2 A schematic diagram for the hollow-fiber membrane contactor used for absorption of CO₂.

2.1 Equations of model

2.1.1 Equations of lumen side

The continuity equation for all species in absorption system can be expressed as [11]

$$\nabla(-D_i C_i + C_i u) + \frac{\partial C_i}{\partial t} = R_i \quad (1)$$

Where C_i denotes the concentration of solute (mol/m³), D_i denotes its diffusion coefficient (m²/s), u the velocity vector (m/s) and R_i denotes the reaction term (mol/m³.s). By applying Fick's law for calculation of diffusive flux, the steady-state continuity equation for CO₂ transport in the lumen side of membrane contactor is obtained as follows:

$$D_{\text{CO}_2\text{-tube}} \left[\frac{\partial^2 C_{\text{CO}_2\text{-tube}}}{\partial r^2} + \frac{1}{r} \frac{\partial C_{\text{CO}_2\text{-tube}}}{\partial r} + \frac{\partial^2 C_{\text{CO}_2\text{-tube}}}{\partial z^2} \right] = V_{z\text{-tube}} \frac{\partial C_{\text{CO}_2\text{-tube}}}{\partial z} - R_{\text{CO}_2} \quad (2)$$

Where r and z represent the radial and axial coordinates, respectively. Because there is no chemical reaction, the reaction term in the lumen side is not discussed. The convective mass transfer in radial direction is neglected, because the feed and solvent are flown in axial direction (z -direction). The velocity distribution in the lumen side is assumed to follow Newtonian laminar flow [16]

$$V_{z\text{-tube}} = 2\bar{u} \left[1 - \left(\frac{r}{r_1} \right)^2 \right] \quad (3)$$

Where u (m/s) is the average velocity in the lumen and r_1 is the fiber inner radius. The boundary conditions assumed for the lumen are given below:

$$\text{At: } Z = L, \quad C_{\text{CO}_2\text{-tube}} = 0 \quad (\text{Inlet boundary}) \quad (4)$$

$$\text{At: } Z = 0, \quad \text{convective flux} \quad (5)$$

$$\text{At: } r = r_1, \quad C_{\text{CO}_2\text{-tube}} = C_{\text{CO}_2\text{-membrane}} * m \quad (\text{Henry's law}) \quad (6)$$

$$\text{At: } r = 0, \quad \frac{\partial C_{\text{CO}_2\text{-tube}}}{\partial r} = 0 \quad (\text{Axial symmetry}) \quad (7)$$

2.1.2 Equations of shell side

The steady-state continuity equation for the CO₂ transfer in the shell side of HFMC is given as follows:

$$D_{\text{CO}_2\text{-shell}} \left[\frac{\partial^2 C_{\text{CO}_2\text{-shell}}}{\partial r^2} + \frac{1}{r} \frac{\partial C_{\text{CO}_2\text{-shell}}}{\partial r} + \frac{\partial^2 C_{\text{CO}_2\text{-shell}}}{\partial z^2} \right] = V_{z\text{-shell}} \frac{\partial C_{\text{CO}_2\text{-shell}}}{\partial z} \quad (8)$$

To characterize the out hollow fibers velocity distribution, Happel's free surface model is used [17]. In fact, Happel's model has been widely used for HFMC, although the flow in the real hollow-fiber modules is not exactly in agreement with this model. The laminar parabolic velocity distribution outside the hollow fibers is:

$$V_{z\text{-shell}} = 2u \left[1 - \left(\frac{r_2}{r_3} \right)^2 \right] \times \frac{(r/r_3)^2 - (r_2/r_3)^2 + 2 \ln(r_2/r)}{3 + (r_2/r_3)^4 - 4(r_2/r_3)^2 + 4 \ln(r_2/r_3)} \quad (9)$$

Where u (m/s) is average velocity in the shell side, r_3 (m) is the radius of free surface and r_2 is the outer radius of fibers (see Fig. 2). The radius of free surface can be estimated as follows:

$$r_3 = \left(\frac{1}{1-\varphi} \right)^{1/2} r_2 \quad (10)$$

In which φ is the volume fraction of the void. It can be calculated as:

$$1 - \varphi = \frac{nr_2^2}{R^2} \quad (11)$$

Where R is the module inner radius and n is the number of fibers. Boundary conditions for shell side are given as:

$$\text{At: } z = 0, \quad C_{\text{CO}_2\text{-shell}} = C_0 \quad (\text{Inlet boundary}), \quad (12)$$

$$\text{At: } r = r_3, \quad \frac{\partial C_{\text{CO}_2\text{-shell}}}{\partial r} = 0 \quad (13)$$

$$\text{At: } r = r_2, \quad C_{\text{CO}_2\text{-shell}} = C_{\text{CO}_2\text{-membrane}} \quad (14)$$

$$\text{At: } z = L, \quad \text{convective flux} \quad (15)$$

2.1.3 Membrane equations

The steady-state continuity equation for the transfer of CO₂ inside the membrane, which is considered to be appropriate to diffusion alone, is written as:

$$C_{\text{CO}_2\text{-membrane}} \left[\frac{\partial^2 C_{\text{CO}_2\text{-membrane}}}{\partial r^2} + \frac{1}{r} \frac{\partial C_{\text{CO}_2\text{-membrane}}}{\partial r} + \frac{\partial^2 C_{\text{CO}_2\text{-membrane}}}{\partial z^2} \right] = 0 \quad (16)$$

Boundary conditions are given as:

$$\text{At: } r = r_2, \quad C_{\text{CO}_2\text{-membrane}} = C_{\text{CO}_2\text{-shell}} \quad (17)$$

$$\text{At: } r = r_1, \quad C_{\text{CO}_2\text{-membrane}} = C_{\text{CO}_2\text{-tube}}/m \quad (\text{based on Henry's law}) \quad (18)$$

Where m is the solubility of CO₂ in the solution. The distribution coefficient of CO₂ in pure water was taken from Versteeg and Van Swaaij [18]:

$$m_{w,\text{CO}_2} = 3.59 \times 10^{-7} RT \exp\left(\frac{2044}{T}\right) \quad (19)$$

The diffusivity of CO₂ in pure water, D_{w,CO_2} was taken from Versteeg and Van Swaaij [18]:

$$D_{w,\text{CO}_2} = 2.35 \times 10^{-6} \exp\left(\frac{-2119}{T}\right) \quad (20)$$

2.2 Numerical solution of model equations

The characteristics of the membrane contactor simulated are presented in Table 1. A 2D mathematical model was developed to predict transfer of CO₂ from CO₂-loaded water solutions (Fig. 2). The separation of CO₂ from CO₂/N₂ gas mixture using water as absorbents in three sections of the single fiber was investigated using a Computational Fluids Dynamics (CFD) approach which was based on the Finite Element Method (FEM). Using finite difference method, the equations governing on absorption process are separated from each other, and an enormous linear system of equations is created which can be described as follows:

$$FP = S \quad (21)$$

Matrices F and S are known here, and matrix P is unknown. One approach to solve this matrix is to calculate the inverse of F . But this approach is not suggested, because it'll create a plethora of non-zero indexes, which needs a high memory in addition to the bunch of calculations. To overcome this problem, the method of breaking the matrix into U and L members (Upper triangular and Lower triangular matrix) or in other words, using direct solving methods is used, and different algorithms such as PSRED, UMF, HJL, HGTR, HGT and so on have been presented, all in which U and L members are tried to have the minimum amount of non-zero components. PARSOL's method has been used in this work to solve the equations. Matrix F is turned

into two L and U matrices using graph theory, and the coefficient matrix is tried to have the minimum amount of non-zero components, so the calculations get accomplished faster and need the minimum of memory.

The model was assumed incompressible, isothermal, laminar, non-wetted condition, steady state condition, and Henry's law was applicable for gas-liquid interface.

Table 1 Module parameters of Mansourizadeh's experiments [14]

Module parameters	
Module i.d.(mm)	14
Fibero.d. (mm)	0.95
Fiberi.d.(mm)	0.60
Effective fiber length (mm)	150
Number of fiber	30
Module length (mm)	250
Packing density (m ² /m ³)	582

3 Results and Discussion

Figure 3 indicated the concentration distribution of CO₂ in all three sections of the membrane contactor. The N₂/CO₂ mixture flows in shell side the contactor where the concentration of CO₂ is the highest in $z = 0$ (C_0), whereas the pure water flows in lumen side ($z = L$) i.e. the concentration of CO₂ in inlet is zero. As the gas flows along the shell side, due to the gradient concentration CO₂ moves the membrane, then it is absorbed by water in lumen.

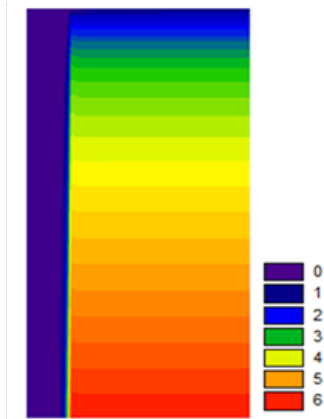


Fig. 3 A representation of the concentration distribution of CO₂ in the membrane contactor for the absorption of CO₂ in water. Gas velocity = 0.02 [m/s], liquid velocity = 0.85[m/s]; CO₂ inlet concentration = 6 mol/m³, n = 30; r₁ = 0.3 mm; r₂ = 0.475 mm; r₃ = 1.278 mm; L = 150mm.

3.1 Effect of liquid velocity on the CO₂ absorption

The equation below can be used to calculate the percentage removal of CO₂:

$$\% \text{ removal CO}_2 = 100 \frac{(v \times C)_{\text{inlet}} - (v \times C)_{\text{outlet}}}{(v \times C)_{\text{inlet}}} = 100 \left(1 - \frac{C_{\text{outlet}}}{C_{\text{inlet}}} \right) \quad (22)$$

The effect of the liquid absorbent velocity on the CO₂ concentration in outlet liquid was investigated (in Fig. 4). At a constant gas velocity of 0.02 m/s, by increasing the liquid velocity from 0.2 to 0.9 m/s, significant decrease in the CO₂ concentration in outlet liquid was observed, about 38 %. The solvent moves faster, as the liquid phase velocity increases, so, the absorbent contact time with inner surface of the fiber along the length of the contactor decreases. Because CO₂ in liquid does not have enough time for mass transfer. Thus, CO₂ concentration decreases in outlet liquid.

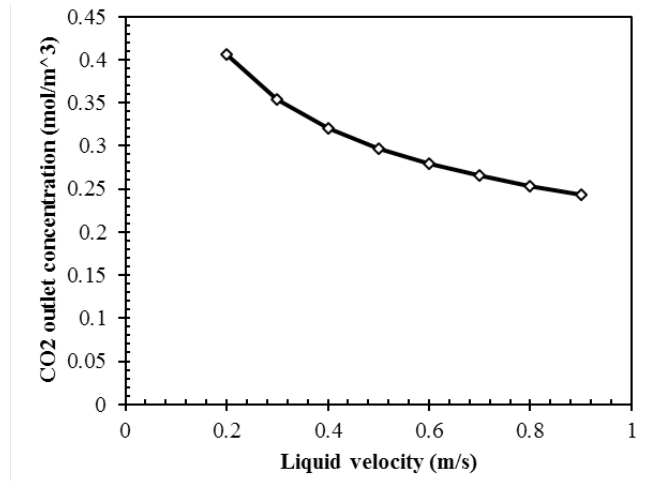


Fig. 4 Relationship between CO₂ outlet concentration in liquid phase and liquid velocity; temperature = 299 K; n = 30; gas velocity = 0.02[m/s]

Figure 5 shows the relation between CO₂ outlet concentration in gas phase (shell side) and liquid velocity at constant temperature of 299 °K. As it is seen in the figure, CO₂ outlet concentration in gas phase decreases by increasing liquid velocity, because the resistance of mass transfer decreases. Therefore, the absorbent absorbs more amount of CO₂ and thus, the percentage removal of CO₂ increases.

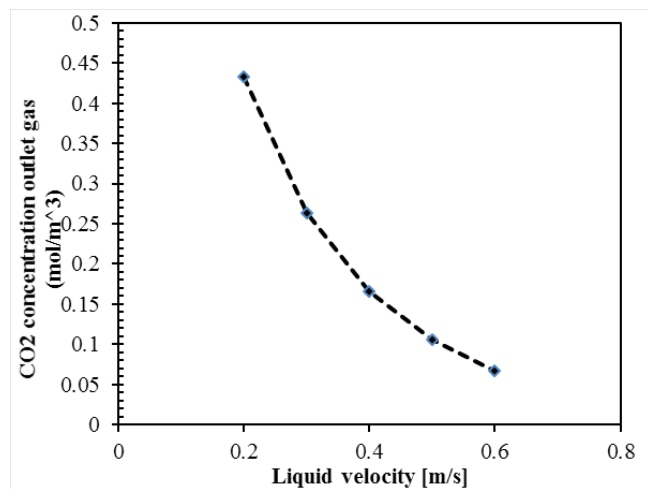


Fig. 5 Relationship between CO₂ outlet concentration in gas phase and liquid velocity; temperature = 299 K; n = 30; gas velocity = 0.02[m/s]

Figure 6 illustrates the variation CO_2 absorption flux as a function of liquid velocity. As the absorbent velocity increases, the mass transfer rate of carbon dioxide into the liquid solvent increases because the concentration gradients of CO_2 and absorbent in the liquid increase, thus the CO_2 outlet concentration in gas decreases and the percentage removal of CO_2 increases (Fig. 7).

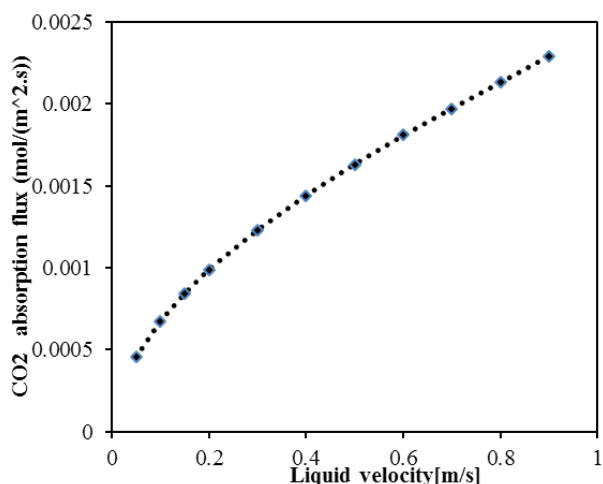


Fig. 6 Relationship between CO_2 absorption flux and liquid velocity, temperature = 299 K, gas velocity = 0.02 [m/s].

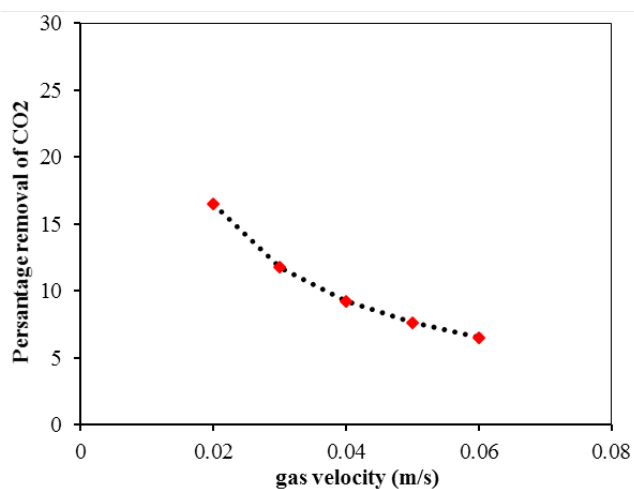


Fig. 7 Effect of gas velocity on the percentage removal of CO_2 for the absorption of CO_2 in water; temperature = 299 K; n = 30; liquid velocity=0.55.

3.2 Effect of gas velocity on the CO_2 absorption

Figure 7 presents the percentage removal of CO_2 in the gas phase of membrane contactor for different values of gas velocity (the effect of convection term). As predicted, while the gas velocity increases, a reduction in the residence time in the membrane contactor happens, which in turn reduces the removal rate of CO_2 in the contactor. As the gas velocity in the membrane contactor changes from 0.02 m/s to 0.06 m/s,

the percentage removal of CO_2 decreases from 16 % to 6 %. Also, Fig. 7 points out that gas flow rate does not affect the CO_2 removal in the membrane contactor very much. The effect of gas velocity on the CO_2 absorption flux was studied at constant temperature of 299 °K in membrane contactor. (in Fig. 8). CO_2 absorption is expected to change by the increase in gas velocity, which is due to the reduction of mass transfer resistance in gas phase. But as it is indicated in Fig. 8, when gas velocity in shell side changes from 0.02 to 0.06 m/s, CO_2 absorption flux increases only 0.14 %. Thus, absorption can be said not to be a strong function of gas velocity.

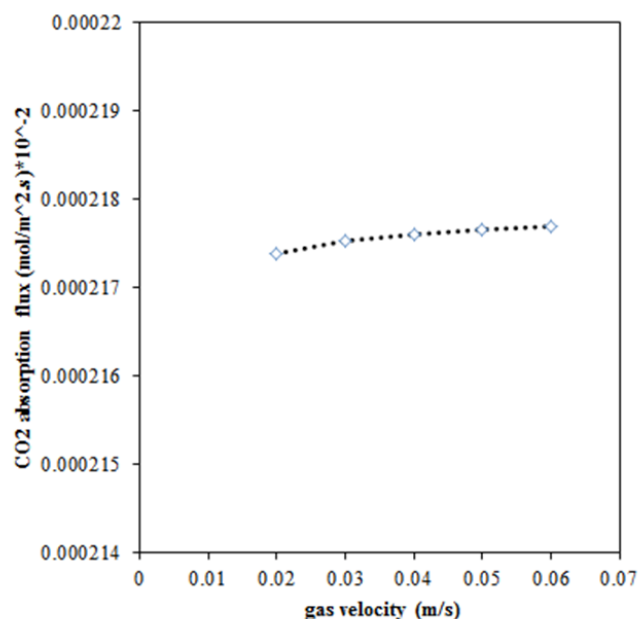


Fig. 8 Effect of gas velocity on the CO_2 absorption flux in membrane contactor.

Figures reveal that the absorption is an intense function of the liquid velocity when the liquid velocity is relatively low. However, the absorption will be less dependent on the liquid velocity at the higher velocities. It can be said that the liquid phase resistance controls mass transfer in absorption process.

Figure 9 shows the effect of liquid phase temperature on CO_2 absorption flux. At constant pressure of 105 Pa and constant velocity of 0.02, by increasing liquid temperature from 300 to 350, an approximate reduction of 38 % is observed in CO_2 absorption flux. Because CO_2 solubility in water decreases by increasing temperature [18]. In addition to this, it must be reminded that water evaporation tendency is more at higher temperatures, and condensed vapor in membrane pores can cause partial wetting. When the temperature of water went up from 20 to 60 °C, physical CO_2 absorption reduced, even when using a highly hydrophobic membranes like PTFE [11]. CO_2 back-pressure increases at higher temperature too, which then reduces mass transfer driving force [20].

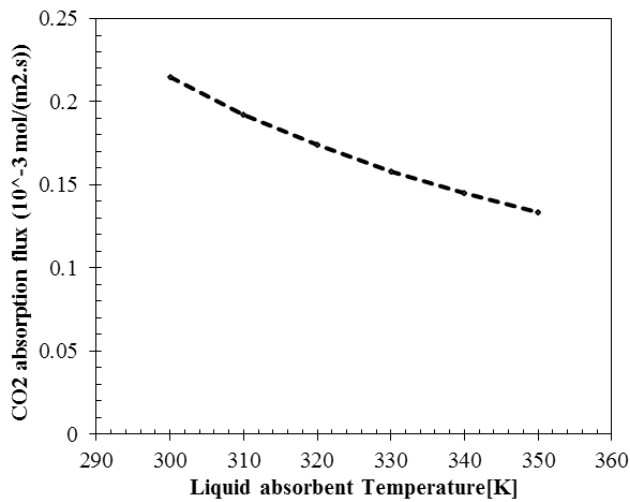


Fig. 9 Effect of liquid temperature on CO₂ absorption flux (U_g= 0.02 m/s; U_l= 0.55 m/s).

3.3 Effect of liquid temperature on the CO₂ absorption

Figure 10 shows the effect of liquid temperature on CO₂ concentration distribution in lumen side of membrane contactor. The comparison of two figures shows that by increasing liquid phase temperature from 300 to 350, an almost uniform distribution is created everywhere but the membrane surface. By increasing temperature, CO₂ concentration in membrane-tube contact decreases, which is observed on the figure by comparing concentration distribution on these two temperatures, because increasing liquid temperature decreases gas solubility in liquid.

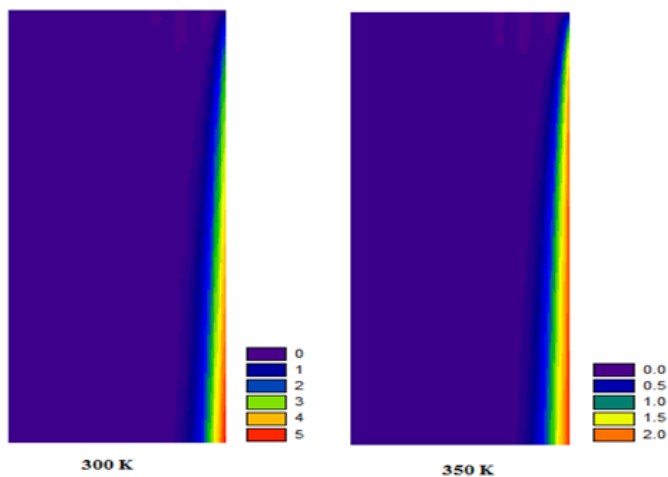


Fig. 10 The effect of liquid temperature on the concentration distribution.

3.4 Experimental validation

The validation of the model was obtained, using the results achieved experimentally by Mansourizadeh et al. [14, 15]. In this section, a comparison between the simulation results with the experimental values is done to validate the model. Figure 11 presents the CO₂ absorption flux along the membrane contactor for different values of liquid velocities. As the liquid velocity increases, the CO₂ absorption flux in the membrane increases,

as well [14]. Figure 12 indicate that model predictions is in accordance with the experimental values. The maximum error in this regard was 6 % with solution time of 5 min.

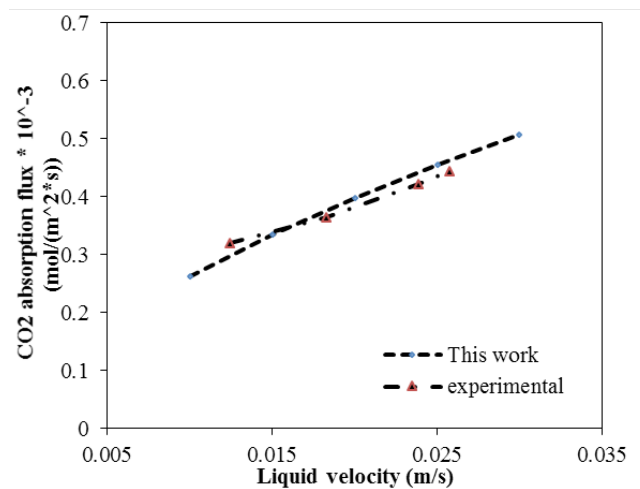


Fig. 11 Comparison of experimental results with simulation results for influence of liquid velocity on the CO₂ absorption flux.

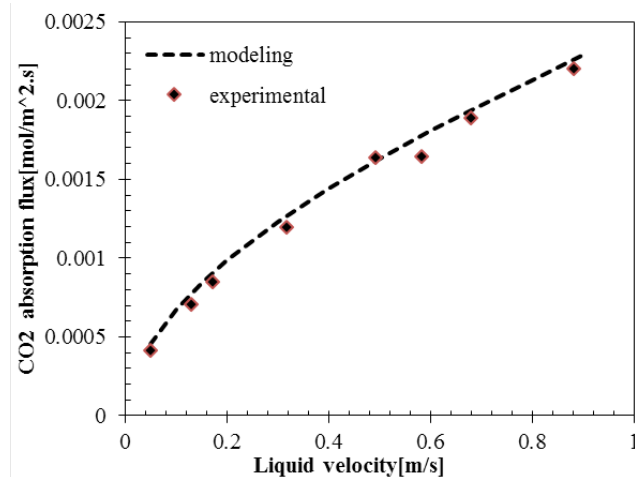


Fig. 12 Comparison of experimental results with simulation results for influence of liquid velocity on the CO₂ absorption flux.

To ensure the exactness of output results, they have been compared with another experimental result of Mansoorzadeh et al. [15]. An error of 4 % has been observed in this validation, therefore the model can be said to be highly accurate.

4 Conclusions

A 2D mathematical model was developed in this work to predict CO₂ transportation from N₂/CO₂ mixing gas. The steady-state two dimensional mass balances for carbon dioxide were taken for all three sections of single fiber, i.e. lumen side, membrane, and shell side. The finite-element method was used to solve model equations. The results showed that by increasing liquid velocity, CO₂ absorption flux increases remarkably, but it decreases only 10 % by increasing gas phase velocity from 0.02 to 0.06. Thus, liquid phase can be concluded to be

controlling mass transfer in the absorption process of gas by liquid. Also, the amount of absorption decreases by increasing liquid phase temperature, which is due to reduction of gas solubility in that phase. To validate the results, the results of two works done by Mansoorzadeh et al. at 2012 and 2014, have been used, and a less than 6 % error has been observed.

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