



# Removal of carbon dioxide by aqueous amino acid salts using hollow fiber membrane contactors



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## ARTICLE INFO

### Article history:

Received 6 September 2015

Received in revised form 7 January 2016

Accepted 26 May 2016

Available online xxx

### Keywords:

CO<sub>2</sub> capture

Hollow fiber membrane contactor

Amino acid

Alkanolamines

## ABSTRACT

The promising absorbent based on amino acid salt has good potential in capturing CO<sub>2</sub>. Amino acids can be considered as a proper solvent because of characteristics such as low volatility, high resistance to degradation, high surface tension and appropriate reactivity with CO<sub>2</sub> in the removal of carbon dioxide. In the present work, the process of carbon dioxide absorption into two aqueous amino acid salts (potassium glycinate and potassium sarcosine) using hollow fiber membrane contactor was modeled. In this work, potassium sarcosine solution was used for the first time as an absorbent for removing CO<sub>2</sub> in HFMC. The performance regarding their absorbing quality of these amino acids was compared with renowned amines. The partial and ordinary equations were solved numerically in liquid, membrane and gas phases. The model results were validated using the available experimental data in the literature for PG, MEA, DEA and MDEA. The results showed that PG has a better performance when it comes to the absorption of CO<sub>2</sub> in comparison to MDEA, DEA and PS at a relatively high CO<sub>2</sub> partial pressures. The absorption flux can be enhanced by increasing the system temperature, amino acids concentration, gas and liquid flow rates. Simulation results indicated that potassium glycinate has a better performance in relatively high partial pressures of CO<sub>2</sub> over MEA.

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## 1. Introduction

Any substance in air which could be harmful to the health, human welfare and the environment is considered as air pollutant. Carbon dioxide has been found to be one of the most important pollutant gases in the atmosphere due to the greenhouse effect and climate changes it causes. With regard to the increasing amount of carbon dioxide production, the use of an effective and economical manner to absorb this gas is highly valuable. In fact, separation processes, their apparatus and equipment are so important that in many industries, the main budget allocated to the product is spent on the separation and purification. Therefore, finding an easier and less expensive separation process can be of a great importance. Although for decades, towers and other traditional contactors had played the main role in chemical industries [1], some problems

arise in these devices due to the need for direct contact between two fluids. These problems include foaming, flooding, channeling and entrainment. Membrane contactors are alternative devices which have solved these problems and provide much larger contact area. Hollow fiber membrane module is one of these contactors [2].

Gabelman and Hwang [1] presented an extensive review on hollow fiber membrane contactors (HFMCs), which is one of the most applicable references in this area due to discussing different modules and their applications. Keshavarz et al. [2] proposed a mathematical model to predict the module performance and the effects of shell side channeling. It was found that the model based on random distribution of fibers is a suitable method for predicting the performance of commercial modules. Many researchers used different solvent for capturing carbon dioxide [3–11]. Lu et al. [12] used two aqueous solutions of MDEA-AMP and MDEA-PZ for absorbing carbon dioxide. They concluded that the mass-transfer fluxes of activated MDEA solutions are significantly higher than that of the non-activated MDEA solution. Marzouk et al. [13] worked on simultaneous removal of CO<sub>2</sub> and H<sub>2</sub>S from pressurized CO<sub>2</sub>-H<sub>2</sub>S-CH<sub>4</sub> gas mixture using HFMC. They found that the liquid phase mass transfer resistance reigns at low pressures whereas gas

*Abbreviations:* AMP, 2-amino-2-methyl-1-propanol; DEA, diethanolamine; DEAB, 4-diethylamino-2-butanol; MEA, monoethanolamine; PG, potassium glycinate; PS, potassium sarcosine; PTFE, polytetrafluoroethylene; PVDF, polyvinylidene fluoride.

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## Nomenclature

$C$	Concentration (kmol m <sup>-3</sup> )
$d_p$	Pore diameter (cm)
$D$	Diffusivity (m <sup>2</sup> s <sup>-1</sup> )
$D_{kj}$	Knudsen diffusivity of species $j$ (m <sup>2</sup> s <sup>-1</sup> )
$H$	Henry's constant (kmol kmol <sup>-1</sup> )
$J$	Absorption flux (kmol m <sup>-2</sup> s <sup>-1</sup> )
$k$	Reaction rate constant
$M$	Molecular weight (kg kmol <sup>-1</sup> )
$P$	Total pressure (kPa)
$R$	Radial coordinate (m)
$R_e$	Free surface radius of fiber (m)
$R_i$	Inner radius of fiber (m)
$R_j$	Reaction rate of component $j$ (kmol m <sup>-3</sup> s <sup>-1</sup> )
$R_M$	Module inner diameter (m)
$R_o$	Outer radius of fiber (m)
$R_w$	Radius of the gas-liquid interface (m)
$T$	Temperature (K)
$U$	Velocity (m s <sup>-1</sup> )
$z$	Axial coordinate (m)

### Greek letters

$\delta_1$	Liquid-filled membrane thickness (m)
$\varepsilon$	Membrane porosity
$\theta$	Packing density
$\tau$	Tortuosity
$\sigma_{\text{CO}_2\text{N}_2}$	Lennard-jones parameter (Å)
$\Omega$	Collision integrals

### Subscripts

$A$	Absorbent
$av$	Average
$e$	Effective
$G$	Gas
$in$	Input
$j$	Any diffusing species
$m$	Membrane
$nw$	Non-wet
$w$	Wet
$P$	Pore
$S$	Solvent

phase mass transfer resistance has a significant role at higher pressures. Mehdi-pour et al. [14] reached a complete removal of CO<sub>2</sub> by 2 M K<sub>2</sub>CO<sub>3</sub> solution at 298 K with HFMC. The results illustrated that potassium carbonate, under its optimum conditions, can give higher CO<sub>2</sub> recovery than diethanolamine solution, especially if DEA wets the membrane. Furthermore, Masoumi et al. [15] studied carbon dioxide separation by amine promoted potassium carbonate solution in a hollow fiber membrane contactor. The higher fluxes of promoted K<sub>2</sub>CO<sub>3</sub> with MEA or DEA as well as other advantages of K<sub>2</sub>CO<sub>3</sub> solution, such as lower cost and easier regeneration, make them good alternative solutions compared to alkanolamines, such as single MEA or DEA, for CO<sub>2</sub> capture in HFMCs. Also they did another investigation using DEAB as a novel absorbent [16]. It was shown that CO<sub>2</sub> absorption flux into DEAB in all calculated range of CO<sub>2</sub> partial pressures is lower than MEA and higher than MDEA.

The removal of CO<sub>2</sub> as an acid gas by aqueous alkanolamine solutions is widely used in the chemical industry. Utilizing alkanolamines in industrial processes has some major problems such as high volatility as well as degradation because of side

reactions with carbon dioxide, oxygen and other contaminants. Use of an appropriate solvent coupled with hollow fiber membrane can be a great idea to improve the absorption of carbon dioxide.

The amino acid salts have certain advantages over alkanolamines such as higher surface tension (this parameter influences on the wetting of fibers which has a direct effect on the performance of absorption) and an almost nonvolatile state (by adding a salt function into amino acid solution). Yan et al. [17] recommended PG as a suitable solvent because of its physical properties. Indeed, it avoided the wetting problem of commercial polypropylene (PP) microporous membranes, also, it had appropriate reactivity towards CO<sub>2</sub> compared with the conventional solvents, MEA and MDEA.

Lu et al. [18] investigated the performance of single solution (Glycine) and novel composite solution (Glycine + Piperazine). They concluded that the efficiency of the composite solution is evidently higher than that of the single glycine salt solution in coupling with hollow fiber membrane contactor. Ghasem et al. [19] applied aqueous amino acid salts for CO<sub>2</sub> absorption. It was clearly seen from the results that the performance of CO<sub>2</sub> absorption using PG is always better than MEA and DEA under the identical operating circumstances. Complete removal was achieved using potassium glycinate (PG) solution as a solvent.

This work is the first study on the CO<sub>2</sub> absorption into potassium sarcosine (PS) solution using hollow fiber membrane contactor. By applying the presented mathematical model, the CO<sub>2</sub> absorption in the case of non-wetted fibers were simulated. Liquid moves through inside the fiber and gas flows in the shell side. The reactions which occurred and velocity equations were explained comprehensively. The partial and ordinary equations were solved numerically in liquid, membrane and gas phases. The model results were validated using the available experimental data in the literature for PG, MEA and MDEA. The effects of some parameters including absorbent concentration, liquid and gas velocities, CO<sub>2</sub> partial pressure and wetting were investigated using the model.

## 2. Model theory

We can see the schematics of hollow fiber membrane contactor in Fig. 1. Hollow fiber membrane contactors consist of three parts: shell, membrane and tube. In parallel co-current flow, both the gas mixture (CO<sub>2</sub> and N<sub>2</sub>) and the absorbent liquid enter the contactor from the same end, but in countercurrent case, gas mixture flows from one end and the absorbent liquid enters from another end into the membrane contactor. Carbon dioxide separates from the gas phase, penetrates through the membrane and comes in contact with absorbent liquid. After dissolution, it diffuses with reaction to the liquid phase. The partial differential equation governs in liquid phase, which flows within the fiber using a fully developed laminar parabolic velocity profile [5]. The membrane has uniform pore size distribution and thickness [5]. An atmospheric gas flows into fibers environs in the same direction based on Happel's free surface model [20]. The steady state and isothermal conditions are assumed for all sections [14]. The ideal behavior can be assumed for gas phase due to low pressure.

### 2.1. Reaction scheme

The zwitterion mechanism is generally acceptable to model the carbon dioxide absorption in aqueous amino acids solutions [21]. Based on the zwitterion mechanism, CO<sub>2</sub> reacts with the amino acid salt (PS and PG in the present case), generating zwitterions, which are intermediate components. Then, some of the

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