



# Experimental and theoretical study of carbon dioxide solubility in aqueous solution of potassium glycinate blended with piperazine as new absorbents



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## ABSTRACT

In this work aqueous solution of potassium glycinate, piperazine and potassium glycinate blended with piperazine have been utilized through CO<sub>2</sub> solubility measurements in order to investigate the possible use of these salts of amino acid blended with alkanolamine for CO<sub>2</sub> absorption. The equilibrium solubility of CO<sub>2</sub> for aqueous solutions of potassium glycinate, piperazine and potassium glycinate blended with piperazine at 1.0, 4.0 and 10.0 wt.% overall mass concentration are experimentally measured with an equilibrium cell at CO<sub>2</sub> partial pressure ranging from 5.1 to 2508.7 kPa and temperatures between 293.15 and 323.15 K. Obtained data show that loading capacity decreases with increase in temperature and concentration of potassium glycinate and potassium glycinate blended with piperazine. Obtained CO<sub>2</sub> loadings are very high at low concentration of potassium glycinate. In order to predict CO<sub>2</sub> solubility in aqueous solutions potassium glycinate, piperazine and potassium glycinate blended with piperazine, Deshmukh-Mather model is extended. The values of MSE, ARD and R<sup>2</sup> for the extended Deshmukh-Mather model are 0.0741, 8.9582 and 0.9826, respectively. In addition, an artificial neural network (ANN) is developed to predict CO<sub>2</sub> solubility in aqueous solutions of piperazine, potassium glycinate and potassium glycinate blended with piperazine. The values of MSE, ARD and R<sup>2</sup> for the optimal trained ANN are 0.0179, 4.1528 and 0.9962, respectively.

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## 1. Introduction

Many industries release a large value of greenhouse gases (GHG) that is not proper for environment. Global warming is a phenomenon that is harmful for environment and amplifies with GHG propagations; therefore, decrease of GHG is vital to avoid climate changes. 55% of global warming is due to presence of Carbon dioxide (CO<sub>2</sub>) in atmosphere. Thus, decrease of emission of CO<sub>2</sub> from industries is very important [1].

Absorption process is one of the most important processes to decrease CO<sub>2</sub> from flue gas streams [2]. Also aqueous solution of alkanolamines are applicable chemical solvents to decrease CO<sub>2</sub> from flue gases due to low cost, high resistance to thermal degradation, low hydrocarbons loading capacity and high absorption rate [3]. However, alkanolamines have some disadvantages including high-energy requirements for process, degradation in

presence of oxygen, also exploiting alkanolamines can cause corrosion, foaming and fouling [3].

Newly, ionic liquids (ILs) have been introduced as new absorbents for CO<sub>2</sub> absorption. Rate of vaporization of ILs is low due to low vapor pressure, but using ILs have some difficulties containing high viscosity, high cost and low CO<sub>2</sub> loading [4]. Recently aqueous solutions of amino acid salt as new kind of solutions are introduced [5]. Amino acid salt solutions as new absorbents have a potential alternative for alkanolamines and ionic liquids. Although alkanolamines are cheaper than amino acid salt solutions, but due to ionic nature of amino acid salts, they have suitable properties like, high chemical reactivity, low vapor pressures, high stability toward oxidative degradation, suitable binding energy with CO<sub>2</sub> and low viscosities [6]. Precipitation may occur when CO<sub>2</sub> was absorbed in to amino acids salt solution. Solid formation in the liquid phase through CO<sub>2</sub> absorption cause increase in the CO<sub>2</sub> absorption with shift the reactions towards the formation of more solid phase [7]. Also the glycine as a common amino acid is being widely used especially for making various polymeric and liquid membranes for CO<sub>2</sub> separation.

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### Nomenclature

T	Temperature (K)
$P_{CO_2}$	Partial pressure of CO <sub>2</sub> (kPa)
X	Overall mass concentration of the mixture
$M_{app}$	Apparent molecular weight (g/mol)
$MW_i$	Molecular weight component i
N	Number of data point
NC	Number of component in the mixture
i	Integer counter
I	Ionic strength
a	Activity
H	Henry coefficient

### Symbols

$\alpha$	CO <sub>2</sub> solubility (mole CO <sub>2</sub> /mole solution)
$\bar{\alpha}$	Average CO <sub>2</sub> solubility (mole CO <sub>2</sub> /mole solution)
$\beta$	Binary interaction parameter
$\gamma$	Activity coefficient
$\varepsilon$	Dielectric constant of the solvent
MEA	Monoethanolamine
PZ	Piperazine

### Subscripts

calcd	Calculated
exp	Experimental
carbAmA	Carbamate that produced by salt of amino acid
carbPZ	Carbamate that produced by piperazine
AmA	Amino acid
w	Water
MLFNN	Multi-layer feed-forward neural network
ANN	Artificial neural network
ARD	Average Relative Deviation
MSE	Mean Square Error
R <sup>2</sup>	Correlation Coefficient

Ho [8] invented a hydrophilic polymer that was suitable for CO<sub>2</sub> capture from flue gases. For polymer preparation, he used at least one amino acid salt, with mass concentration between 10 and about 80%wt. Obuskovic and Sirkar [9] used hydrophilic porous hollow fiber membranes to study appropriate immobilized liquid membranes (ILMs) for selective CO<sub>2</sub> capture from anesthesia breathing circuit gas mixture containing nitrous oxide (N<sub>2</sub>O), carbon dioxide (CO<sub>2</sub>), oxygen (O<sub>2</sub>) and halogenated hydrocarbons (HHCs). Their results indicated that CO<sub>2</sub> in a breathing apparatus was significantly decreased. Chen et al. [10] prepared glycerol-based immobilized liquid membranes (ILMs) for the selective separation of CO<sub>2</sub> from a mixed-gas (CO<sub>2</sub>, N<sub>2</sub>) feed having low CO<sub>2</sub> concentrations in space-walk and space-cabin atmospheres. They used Glycine as an additive in membrane preparation, their results indicate that prepared membrane can suitably separate CO<sub>2</sub> from gas mixture.

A few researches on CO<sub>2</sub> absorption in amino acid salt solutions are reported in literature. Song et al. [11] measured CO<sub>2</sub> loading capacity in aqueous solutions of 10 wt.%, 20 wt.% and 30 wt.% of sodium glycinate at CO<sub>2</sub> partial pressure between 0.1 and 200 kPa and operating temperature from 303 to 323 K. Their measurement indicated that loading capacity decreased with increase in amino acid salt concentration. Munoz et al. [12] measured the CO<sub>2</sub> solubility into 1 M solution of the potassium salts of arginine, glycine, proline, taurine, threonine, ornithine serine, and histidine. Their experiments were done at 293 K and 100 kPa. Their obtained results indicated that solubility of salts of amino acid were similar

to MEA. Portugal et al. [13] experimented CO<sub>2</sub> solubility in potassium salt of threonine and glycine. Their experiments indicated that solubility of salts of amino acid were similar to MEA but in their experiments, no solid phase was produced. Their measurements indicated that with increase in concentration of salt of amino acid, CO<sub>2</sub> solubility decreased. For modeling, Portugal et al. [13] used simplified Deshmukh-Mather model to correlate the CO<sub>2</sub> solubility in aqueous solution of potassium glycinate at limit ranges of temperature and pressure, in equilibrium calculation, they assumed that vapor phase is ideal phase and binary interactions in liquid phase are not function of temperature, therefore their model is not applicable in wide ranges of temperature and pressure. Majchrowicz and Brillman [14] measured the CO<sub>2</sub> equilibrium solubility in aqueous solution of potassium L-proline. Their measurements indicated that CO<sub>2</sub> loading decreased with increase in the molar concentration. Recently Mazinani et al. [15] measured the CO<sub>2</sub> equilibrium solubility in potassium lysinate at low partial pressures of CO<sub>2</sub>. Their investigations indicated that with increase in concentration and operating temperature of aqueous solution of potassium lysinate, CO<sub>2</sub> solubility decreased. Also other novel solvents such as dendrimers which have amine groups have been introduced for CO<sub>2</sub> absorption [16] but glycine as a common and applicable amino acid has potential for selective CO<sub>2</sub> capturing.

Experimental data that presented in literature for equilibrium absorption of CO<sub>2</sub> into some salts of amino acid are limited in types of amino acid and range.

In current work, CO<sub>2</sub> equilibrium loading capacity in solutions of piperazine, potassium glycinate and potassium glycinate blended with piperazine (amino acid salt families like potassium glycinate are new kind of solutions that seem suitable for utilizing in industrial applications) are measured at different overall mass concentration and operating conditions. Also by using these experimental data, Deshmukh-Mather model is extended, in modeling, vapor phase is not considered as an ideal phase and binary interactions in liquid phase are considered as function of temperature. Also, an artificial neural network is developed to predict CO<sub>2</sub> solubility in potassium glycinate and potassium glycinate blended with piperazine. To our best knowledge, literature data for CO<sub>2</sub> solubility in potassium glycinate blended with piperazine are not available and it is the first time in literature Deshmukh-Mather model is extended for an aqueous solution of amino acid salt blended with alkanolamine. It should be noted that apparent molecular weight of potassium glycinate and potassium glycinate blended with piperazine, temperature, overall mass concentration of the solute and partial pressure of CO<sub>2</sub> are selected as input variables of the proposed ANN. For developing neural network based on some literature models [17–21] apparent molecular weight is selected as type of solutions.

## 2. Experimental

### 2.1. Materials

Monoethanolamine (99.5%), Glycine (extra pure, 98.5%), Sarcosine (extra pure, 98.5%), Alanine (extra pure, 98.5%), Potassium Hydroxide (extra pure, 99%), Sodium Hydroxide (extra pure, 99%) and Piperazine hexa hydrate (extra pure, 98%) were obtained from Scharlau. 2-amino-2-methyl-1-propanol (95%) was also purchased from Merck.

### 2.2. Experimental set-up

A schematic of the experimental set-up that is exploited in our previously published work [22] is briefly illustrated in Fig. 1.

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