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# Prediction of acid gas solubility in amine, ionic liquid and amino acid salt solutions using artificial neural network and evaluating with new experimental measurements

Mohammad Ehsan Hamzehie, Hesam Najibi*

*Faculty of Petroleum Engineering, Petroleum University of Technology (PUT), P.O. Box 63431, Ahwaz, Iran*

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ABSTRACT

In this work presented here attempt is prediction of acid gases (carbon dioxide and hydrogen sulfide) loading capacities by employing artificial neural network (ANN) model in 51 single and blended alkanolamine, ionic liquid and amino acid salt solutions as commonly and new industrial absorbents in large domain of operational conditions. Also for evaluating extrapolation capability of ANN, new experimental data on $CO_2$ solubility in aqueous solutions of Potassium Glycinate blended with Piperazine (PZ) and 2-amino-2-methyl-1-propanol (AMP) at different temperatures and pressures are measured. It should be mention that $CO_2$ solubility data for these two solutions are not available in literature. For developing ANN, solution pH, total mass concentration, partial pressure of $CO_2$ and $H_2S$, apparent molecular weight, critical temperature, critical pressure and temperature are assumed as inputs. A band of 2982 experimental data points for $CO_2$ and $H_2S$ loading capacities have been collected from literature to create the suggested ANN. The best structure of the suggested network is achieved by employing these literature data points. The network is trained by algorithm of Levenberg–Marquardt back-propagation, consists of 9 and 6 neurons in first and second hidden layers, respectively. For the hidden and output layers, Tan-sigmoid transfer function is utilized. The output results of developed network show that suggested network that is created with solubility data of single and blended alkanolamine, ionic liquid and amino acid salt solutions has capability to predict accurately $CO_2$ and $H_2S$ loading capacities in dissimilar commonly and new industrial solutions with Average Relative Deviation (ARD %) equal to 2.7992, Mean Square Error (MSE) value of $3.7468 \times 10^{-5}$ and correlation coefficient ($R^2$) equal to 0.9984.



## 1. Introduction

Currently significant value of hydrogen sulfide ($H_2S$) and carbon dioxide ($CO_2$) are made and released from industrial activities like landfill management, wastewater treatment, gas refinery, petrochemical units, paper manufacturing and several other industrials. In many cases, industry has main role in environmental crises. For example, more than 60 percent of annual greenhouse gas emissions are related to the industrial activities in the world (transportation fuels and distribution 25.3%, power stations 21.3% and industrial process 16.8%) (Valipour et al., February; Valipour et al., 2013; Valipour et al., 2012a). Attendance of acid gases is a main issue that causes difficulties in use of gas streams.

For example, presence of acid gases in natural gas streams cause reduction in heating value and corrosion in process equipment. $H_2S$ and $CO_2$ contents in the gas fuels for utilization must be less than 4 ppm and 2 mol %, respectively. Therefore, removal or reduction in the concentration of acid gases from gas mixtures is important (Startsev et al., 2013).

Among technologies to remove acid gas from gas fuels, chemical absorption as flexible, proper and relatively low cost method with common solvents like alkanolamine and ionic liquid solutions is considered (Rufford et al., 2012). Also amino acid salt solutions as recent chemical absorbents have potential alternative for alkanolamines and ionic liquids. Although amino acid salt solutions are relatively expensive than alkanolamines, but due to polarized nature, they have low viscosities, low vapor pressures, proper binding energy with acid gases, high stability against oxidative degradation and good chemical reactivity (Senthil Kumar et al., 2001). Solubility

* Corresponding author.
*E-mail addresses:* hesamnajibi60@gmail.com, najibi@put.ac.ir (H. Najibi).

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data of acid gases into commonly and new chemical solvents have an important role in the simulation and design of refinery units and can determine the total cost of the chemical plants (Maddox, 1985). Therefore, a true estimation of equilibrium loading capacities of $CO_2$ and $H_2S$ in chemical absorbents is vital.

Experimental data for acid gas ($CO_2$ and $H_2S$) loading capacities into vast solutions of single and blended alkanolamine, ionic liquid and amino acid salt solutions as commonly and new industrial absorbents are collected from literature and they are limited in accuracy and range. Fundamentally, the loading capacity of a gas into a chemical solution is a function of some variables, like type of solution, pressure, temperature and concentration. For developing ANN, many collected experimental data points from previous literature works for training data set are given in Table 1 (Park et al., 2002; Kundu et al., 2003; Baek and Yoon, 1998; Kamps et al., 2007; Ma'mun et al., 2005; Aroun et al., 2011; Addicks et al., 2002; Sandall and Park, 2001; Rho et al., 1997; Jou et al., 1998, 1996; Derks et al., 2005; Kennard and Meisen, 1984; Rebolledo-Libreros and Trejo, 2004; Murrieta-Guevara et al., 1998; Tong et al., 2012; Shen and Li, 1992a; Henni and Mather, 1995; Cheng et al., 2010; Austgen et al., 1991; Dawodu and Meisen, 1994; Jenab et al., 2006; Cheng et al., 2010; Hosseini Jenab et al., 2005; Seo and Hong, 1996; Jane and Li, 1997; Kundu and Bandyopadhyay, 2006; Rahmati-Rostami et al., 2009; Sakhaeinia et al., 2010; Rebolledo-Morales et al., 2012; Jalili et al., 2010, 2013; Safavi et al., 2013; Sidi-Boumedine et al., 2004; Huttenhuis et al., 2007; Feng and Mather, 1993; Guevara et al., 1988; Kamps et al., 2003; Xia et al., 2000; Jou et al., 1990; Haghtalab et al., 2014; Murrieta-Guevara et al., 1992, 1994; Rufford et al., 2012), also collected data from literature for testing and validation sets are presented in Table 2, selected experimental data are limited in ranges of pressure, temperature and concentration. Therefore, equilibrium loading capacity data are needed to evaluate the reliability of chemical systems. Also experiments are expensive in some cases, therefore solubility measurements are not always practical. Based on Tables 1 and 2, literature data points are limited in type of chemical solutions and operational conditions. Therefore, prediction of $CO_2$ and $H_2S$ loading capacities in single and blended alkanolamine, ionic liquid and amino acid salt solutions is vital. Therefore, prediction methods according to available data points have been suggested in this work.

Mehrpooya et al. (Mehrpooya et al., 2010) suggested a feed forward artificial neural network (FFANN) for predicting the value of sulfur in sour gas. They were estimated sulfur content from pressure of $H_2S$ data, temperature, concentrations and acid gas gravity. Ibarra-Berastegi et al. (Ibarra-Berastegi et al., 2007) suggested some kinds of artificial networks to decrease $H_2S$ in a biofilter. In their research, elimination of $H_2S$ in a bio filter was modeled. Hemzehie et al. and Bastani et al. (Bastani et al., 2013; Hamzehie et al., 2014, 2015; Hamzehie and Najibi, 2015) suggested some FNN model to predict the $CO_2$ or $H_2S$ loading capacities separately, in some chemical solutions that cover wide ranges of operational conditions. In their research, loading capacities were estimated separately, as a function of total concentration of the solute, solution molecular weight as kind of solution, $CO_2$ and $H_2S$ partial pressure and temperature. Fundamentally, in literature, apparent molecular weight was defined as type of solution (as an input variable) to modeling with ANN.

In literature to our best knowledge all models (Thermodynamic or machine learning algorithm-based models) predicted $CO_2$ or $H_2S$ solubility separately in same family of solutions (family of amines or ionic liquids or amino acids). However, in our work presented here for the first time in literature, $CO_2$ and $H_2S$ loadings simultaneously are accurately predicted in many families of solutions and in wide domain of operating conditions by exploiting a multi-layer feed forward neural network (MLFNN) model. Critical temperature, critical pressure, solution pH, apparent molecular weight, partial pressure of $CO_2$ and $H_2S$, temperature and total mass concentration of the solute are chosen as input parameters of the suggested ANN. New literature experimental data are chosen to create ANN model for various chemical solvents. It should be noted that in this research for defining type of solution, critical properties, pH of solutions and apparent molecular weight of solutions, are selected. In order to creating ANN structure, data exploiting in every steps are not exploited in other steps. Also for evaluating extrapolation capability of ANN, new experimental measurements on $CO_2$ solubility in aqueous solutions of Potassium Glycinate blended with Piperazine (PZ) and 2-amino-2-methyl-1-propanol (AMP) are done. It should be mention that $CO_2$ solubility data for aqueous solutions of Potassium Glycinate + Piperazine and Potassium Glycinate + 2-amino-2-methyl-1-propanol are not available in literature. Results of prediction indicate that developed ANN in this work can predict $CO_2$ solubility in aqueous solution of Potassium Glycinate + Piperazine and Potassium Glycinate + 2-amino-2-methyl-1-propanol accurately; therefore, developed ANN in this work has extrapolation capability to predict $CO_2$ solubility in different mixed solutions. Table 3 shows the large domain of data that exploited to develop the proposed multilayer perceptron neural network (MLPNN).

## 2. Experimental

### 2.1. Materials

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

### 2.2. Apparatus

A schematic of the apparatus that is used in our previously published work (Najibi and Malek, 2013) is shown in Fig. 1 briefly, the main part of the experimental set-up is a cell made of stainless steel 316, which can with stand pressures up to 5 MPa. The equilibrium cell has an inner volume of 31.5 $cm^3$. The cell contents are stirred by a magnetic stirrer, which ensures sufficient agitation to facilitate reaching equilibrium. The cell temperature is controlled by putting it in a bath of oil or water depending on the required temperature. The cell temperature is measured using a platinum resistance sensor (Pt100) which is inserted inside the cell. The uncertainty for temperature measurement is less than 0.1 K. The pressure is measured using a pressure transducer. The uncertainty for pressure measurement is less than 0.01 MPa.

### 2.3. Experimental procedure

In this work, $CO_2$ loadings in all experiments are measured based on difference between primary and secondary pressures. The cell is first evacuated and then 7 ml of previously prepared solution is introduced. After that, $CO_2$ gas is introduced until the cell pressure reached to specified pressure $P_1$ at the initial temperature $T_1$. This initial pressure and temperature are recorded and the cell is put in a bath maintained at controlled temperature T. The cell is leaved in the bath to reach equilibrium conditions and the cell pressure became constant at pressure $P_2$. The amounts of gas at initial and final conditions are calculated by knowing the volumes of cell, connection lines, magnet and liquid solution. Finally by calculating the solution vapor pressure, the moles of $CO_2$ absorbed