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AN ARTIFICIAL NEURAL NETWORK MODEL FOR PREDICTING THE H_2S REMOVAL PERFORMANCE OF PIPERAZINE SOLVENTS IN GAS SWEETENING PLANTS

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ABSTRACT

A feedforward artificial neural network (FFANN) model is proposed for accurate prediction of acid gas absorption capacity of piperazine solvent employed in oil and gas industries. The network has been trained, validated and tested by using 70%, 15% and 15% of all measured equilibrium H_2S solubility datapoints respectively. A network with one hidden layer comprising seven nodes is found to be accurate enough for plant design. The Levenberg–Marquardt back propagation training algorithm is used to train the network while tansigmoid transfer function and a linear transfer function are applied to the hidden and output layers, respectively. The overall average absolute deviation percent (AAD%), mean square error(MSE) and correlation coefficient (R^2-value) of predicted results are about 1.38 %, $4.6X10^{-4}$ and 0.9941 respectively. The presented FFANN model can be utilized for accurate prediction of H_2S removal efficiency of piperazine solvent in sour gas sweetening plants.

Keywords: *neural network, H_2S solubility, gas sweetening, piperazine.*

INTRODUCTION

Sour gas sweetening is often achieved by absorption of acid gases in aqueous solution of amines. The basic design of such absorption processes requires an accurate model for the solubility of sour gases, e.g. carbon dioxide and hydrogen sulfide, in aqueous solutions of one or more amines, e.g. methyldiethanolamine (MDEA), piperazine ($PIPH_2$), monoethanolamine (MEA), diglycolamine (DGA), and diethanolamine (DEA). Piperazine ($PIPH_2$) is a new activator which is used in combination with selective amine solvents such as MDEA to increase the removal capacity of H_2S and CO_2. In this work, the H_2S absorption capacity of aqueous piperazine solvent in gas sweetening plants is studied. The ability of a solvent to remove hydrogen sulfide is dictated by its equilibrium solubility as well as mass transfer and chemical kinetics characteristics.

The equilibrium solubility of hydrogen sulfide in aqueous piperazine solvent as well as the kinetic of absorption has been experimentally studied [1]. The equilibrium solubility of hydrogen sulfide in aqueous MDEA and piperazine solutions has also been experimentally investigated [1-5]. Because of the inherent complexity of such systems, thermodynamic models such as electrolyte solution models are often failed to describe the phase equilibrium of strong electrolyte systems especially at high temperatures and pressures. Xia et al. [1] have used a G^E model to calculate the activity coefficients of both molecular and ionic species of aqueous electrolyte solution of mixed components H_2O, piperazine, MDEA, and H_2S. The calculation requires the knowledge of the temperature

dependent chemical equilibrium constants as well as interaction parameters. However, the predicted results are not accurate enough for design purposes. Bishnoi et al. [6] studied the absorption of carbon dioxide into aqueous solutions of piperazine and corresponding absorption rate was measured from mass transfer and chemical equilibrium reaction data.

Although thermodynamic models can be implemented for modeling the phase equilibria of the electrolyte system, these models may give inaccurate predictions at high solvent concentrations. There are also strong deviations between predicted and measured values at different temperature and pressures. In order to overcome these difficulties, the artificial neural network approach has been employed in this work.

MODEL DEVELOPMENT

In this work a feedforward neural network (FFNN) [7-11] has been developed for prediction of H_2S removal capacity of piperazine solvent in oil and gas industries. Each layer of the proposed network except input layer, receive an input from each neuron in the previous layer and deliver an output to the neurons in the next layer after passing its weighted sum inputs plus a bias value through an activation function according to the following equations:

$$y_j = f_h(a_j) = f_h\left(\sum_{j=1}^{n} w_{ji} x_i + b_j\right) \quad (1)$$

where w_{ji} is the weight between the j-th hidden neuron and the i-th input node in the input layer, x_i denotes the i-th input, b_j is the bias value of the j-th hidden node, n is the number of hidden neurons, f represents the hidden layer transfer function, and y_j is the output of the j-th hidden neuron. In this work the hyperbolic tangent sigmoid activation function is used for the hidden layer which can be written in the following form:

$$f_h(a_j) = \frac{e^{aj} - e^{aj}}{e^{aj} + e^{aj}} \quad (2)$$

For the output layer, a linear transfer function is used:

$$f_o(a_k) = a_k \quad (3)$$

where a_k is the output from the k-th output node and f_o denotes the transfer function of the output layer. The backpropagation algorithm is employed for network training. It uses the error between the real network output and the target values to adjust the weight and bias of the layers. The error function can be expressed as:

$$E = \frac{1}{N}\sum_{i=1}^{N}(O_i - t_i)^2 \quad (4)$$

where N is the number of elements in the output vector, O_i is the i-th element of the network output and t_i represents the according target value. The backpropagation algorithm minimizes the error function so that the weight and bias values can be updated using an updating rule. Consequently, a gradient of the error is considered, where,

$$\nabla E = \frac{\partial E}{\partial w_{kj}} \quad (5)$$

Now, by the steepest gradient the weight adjustment can be written as

$$w_{kj}^{m+1} = w_{kj}^{m} + \Delta w_{kj}^{m} \quad (6)$$

where m is the iteration number, w_{kj} denoting the weight between the k-th neuron in the output layer and neuron j in the hidden layer, Δw_{kj} can be defined by the steepest descent procedure:

$$\Delta w_{kj} = -\eta \frac{\partial E}{\partial w_{kj}} \quad (7)$$

where η is the learning rate and $\frac{\partial E}{\partial w_{kj}}$ separately defined for hidden and output layers:

$$\frac{\partial E}{\partial w_{kj}} = -\phi_k y_j(p-1) \quad (8)$$

And by eqs.7 and 8:

$$\Delta w_{kj} = \eta \phi_k y_j(p-1) \quad (9)$$

where j denoting the j-th input to neuron k of the output layer (p), and ϕ_k can be written as:

$$\phi_k = y_k(1-y_k)(t_k - y_k) \quad (10)$$

y_k is the output of the k-th neuron in the output layer. Therefore, the derivation of the setting of output layer weights can be completed as follow:

$$\Delta w_{kj}(p) = \eta \phi_k(p) y_j(p-1) \quad (11)$$

Accordingly, for the j-th neuron of the hidden layer, $\phi_j(p-1)$ term can be rewritten as:

$$\phi_j(p-1) = y_j(p-1)\left(1 - y_j(p-1)\right)\sum_k \phi_k(p) w_{kj}(p) \quad (12)$$

And from Eq. (7), weight adjustment of the hidden layer can be expressed as

$$\Delta w_{ji}(p-1) = \eta \phi_j(p-1) y_i(p-2) \quad (13)$$

In this work, Levenberg–Marquardt back propagation algorithm is used for network training.

RESULTS AND DISCUSSION

The most influencing parameters such as temperature, pressure and piperazine concentration are defined as input variables of the proposed neural network. The equilibrium H_2S sour gas solubility in the aqueous solution of piperazine is the network output. Network has been trained, tested and validated by using 70%, 15% and 15% of all measured equilibrium H_2S solubility data respectively. A feedforward neural network with seven neuron in one hidden layer is found to be the most accurate structure as shown in Fig 1. The prediction performance regarding the training data is shown in Fig 2. According to this figure, it is evident that the prediction error between the experimental and predicted network output is very low. The accuracy of the trained network has been examined by testing and validation calculations as shown in Figs. 3 and 4 respectively. The correlation coefficient (R^2-value) for training, validation and testing calculations are 0.9933, 0.9982 and 0.9942 respectively, suggesting that the proposed network is well trained and validated. The MSE value of the network for the training, validation, and testing data are about $5X10^{-4}$, $2.7X10^{-4}$, and $4.7X10^{-4}$, respectively. The optimum calculated values of network parameters to be used in simulations are given in Table 1.

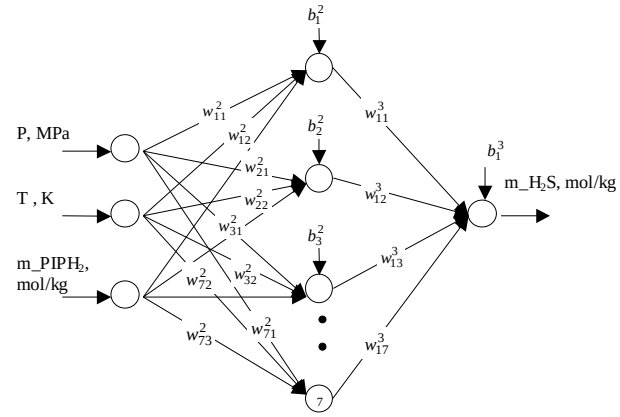

Fig. 1. Proposed Feedforward neural network structure.

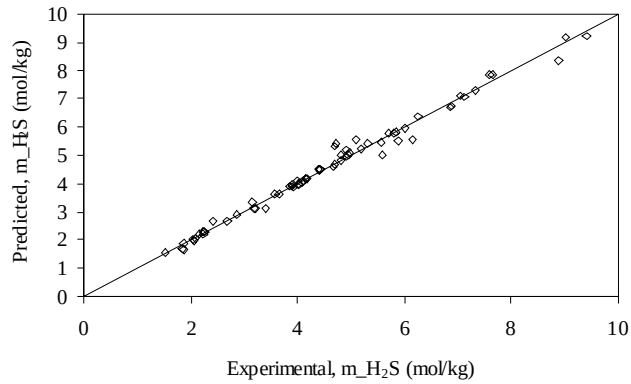

Fig. 2. Comparison of the proposed neural network model predictions with the experimental H_2S solubility data (training).

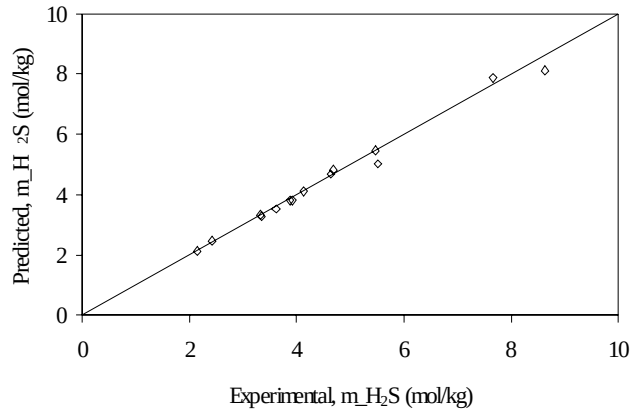

Fig. 3. Comparison of the proposed neural network model predictions with the experimental H_2S solubility data (testing).

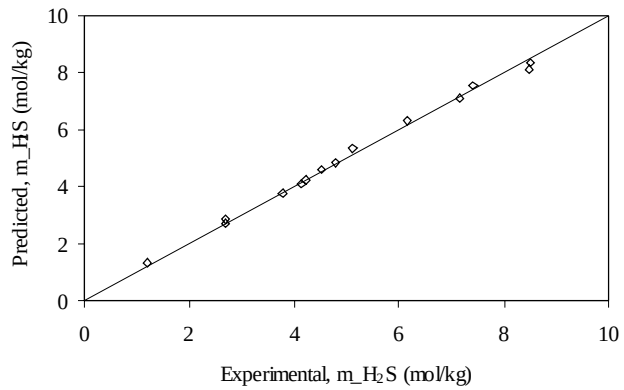

Fig. 4. Comparison of the proposed neural network model predictions with the experimental H_2S solubility data (validation).

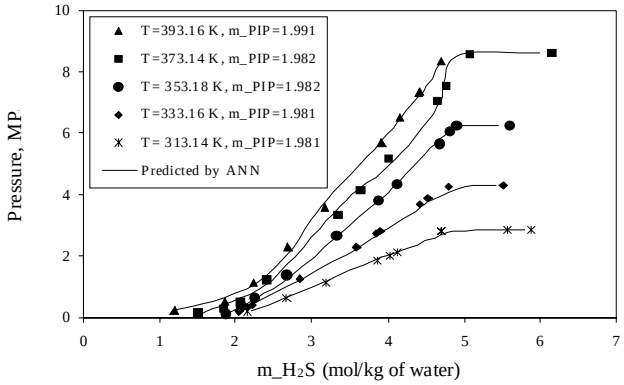

Fig. 5. Comparison of the proposed neural network model predictions with experimental H_2S solubility in aqueous piperazine solution (low piperazine concentration).

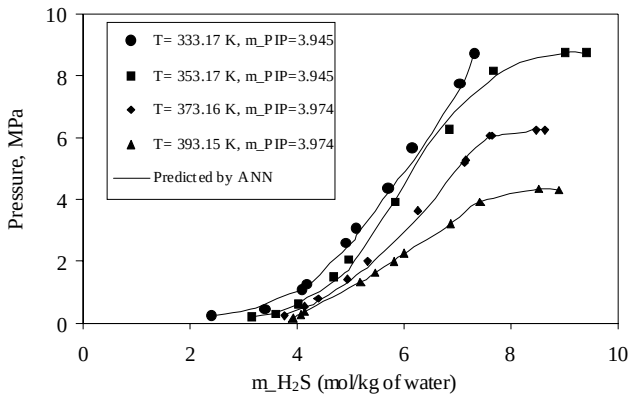

Fig. 6. Comparison of the proposed neural network model predictions with experimental H_2S solubility in aqueous piperazine solution (high piperazine concentration).

Predicted molal solubility of H_2S in aqueous solution of piperazine by using the proposed neural network model are compared with experimental data at different pressures, temperatures and piperazine concentrations in Figs 5 and 6. According to these figures, the deviations between the ANN predicted H_2S solubilities and experimental results are very low for the entire range of operating variables.

As shown in Figs. 5 and 6, the solubility of H_2S is very sensitive to the piperazine molality in the solution such that whenever stoichiometric H_2S molality in the solution is less than that of piperazine, the sour gas is practically completely dissolves in ionic form, as hydrogen bisulfide. However when, the stoichiometric H_2S molality surmounts that of the piperazine, H_2S dissolves physically in the solution. Therefore, the liquid can be regarded as an aqueous mixture of two strong electrolytes, so that traditional electrolyte models fail to describe the system as a whole. As shown in Figs 5 and 6, at a fixed temperature, adding H_2S to a piperazine-containing aqueous solution increases total pressure above the solution. When the stoichiometric molality of the H_2S surmounts that of the base piperazine, the total pressure increases steeply as the H_2S can no longer be absorbed chemically and the physical absorption dominates [1]. As H_2S is added to the solution, total pressure increases steeply reach to a plague and remain nearly constant. This phenomenon is due to the formation of the second liquid phase in equilibrium with first liquid phase. The above experimental observations are well predicted by the proposed neural network as shown in Figs 5 and 6. It should be noted that the reaction of H_2S with piperazine is a much faster than the H_2S reaction with conventional carbamate-

Table 1. Optimum values of the proposed neural network parameters.

w_{j1}^2	w_{j2}^2	w_{j3}^2	b_j^2	w_{1j}^3	b_j^3
0.4904	-2.8018	-0.994	3.1647	-0.075	
0.2451	-2.2707	2.2922	-1.6661	0.706	
-0.0038	1.8577	3.2794	-0.5529	-0.0725	
-2.9065	0.3507	-1.4966	0.5924	-0.2809	-0.0839
0.021	0.0616	-0.7322	-0.9063	-1.0498	
2.3719	-1.0802	2.7844	0.9187	0.0203	
0.5151	-3.0105	-0.2864	2.9313	0.0572	

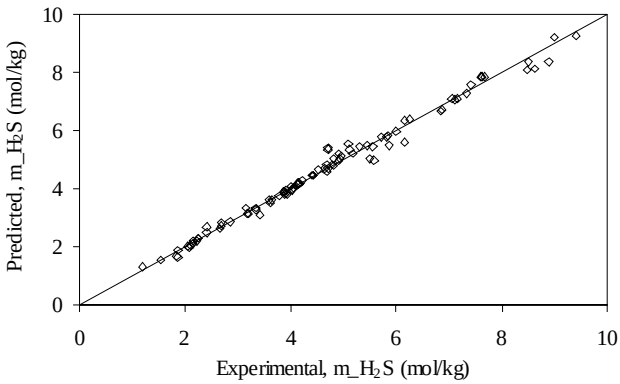

Fig. 7. Comparison of the proposed neural network model predictions with all experimental H_2S solubility data.

Table 2. Overall deviations between predicted and experimental H_2S solubilities in piperazine solution.

Error	m_H_2S (mol/kg)
AAD%	1.38
MSE	4.6×10^{-4}
R^2	0.9941

forming amines such as MEA, DGA, or DEA. The behavior of the system H_2S + piperazine + H_2O is very similar to that observed for the system H_2S + MDEA + H_2O [1, 12].

Fig 7 shows the comparison of the proposed neural network model predictions with all experimental H_2S solubility data with correlation coefficient of R^2=0.9941. Deviations between predicted and experimental data are given in Table 2. The overall AAD%, MSE and R^2-values are about 1.38 %, $4.6X10^{-4}$ and 0.9941 respectively. As shown in Fig 7, the presented neural network model can be used for accurate prediction of the equilibrium H_2S solubility in aqueous solution of piperazine over wide ranges of temperature, pressure, and solvent concentrations.

CONCLUSIONS

A feedforward artificial neural network (FFANN) with one hidden layer comprising of seven nodes has been proposed for accurate prediction of H_2S solubility in aqueous piperazine solvents. The overall average absolute deviation percent (AAD%), mean square error(MSE) and correlation coefficient (R^2-value) of predicted results are about 1.38 %, $4.6X10^{-4}$ and 0.9941 respectively. It is found that the proposed neural network prediction results are much more accurate than those obtained by using the available thermodynamic models. The presented FFANN model can be utilized for accurate prediction of H_2S removal efficiency of piperazine solvents in oil and gas industries.

NOMENCLATURE

f transfer function
w_{ji} weight value between i-th input and j-th neuron of the input and hidden layers
w_{kj} weight value between j-th hidden neuron and the k-th neuron of the output layer
b_j bias value of the j-th neuron
AAD% average absolute deviation percent
MSE mean squared error
E error function
y_j net output from j-th neuron
m iteration number
n hidden nodes number
N number of elements in the input-output vectors
m_H_2S molal solubility of H_2S (mol/kg of water)
k index of k-th neuron in the output layer
O_i overall network output for the i-th element
t_i target values of the i-th element
a_j output from j-th neuron
T absolute temperature
R correlation coefficient
x_i i-th input to the network Greeks
η learning rate
Φ_j error term back-propagated from the j-th node
Subscript
h hidden layer
o output layer Superscript
Exp experimental value
Pred predicted value

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