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Research paper

Thermodynamical and artificial intelligence approaches of H_2S solubility in *N*-methylpyrrolidone

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ABSTRACT

Artificial Intelligence approach is utilized for developing the auto-design model of neural network based genetic algorithm (GA) to manipulate the obtained experimental data of H_2S solubility measurements. The solubility of hydrogen sulfide (H_2S) in low vapor pressure organic solvents *N*-methylpyrrolidone (NMP) has been measured experimentally. The experimental input data included operating temperature and pressure, and solubility values regarded as output data. Design of topology and parameters of the neural networks as decision variables have been achieved by GA, which enhances the effectiveness of the forecasting scheme. The precision of the GA is compared with experimental data and also with thermodynamical modeling approach in which Peng-Robinson-Stryjek-Vera (PRSV) cubic equation of state has been chosen for this purpose. The results reveal that the thermodynamical and GA–artificial neural networks (ANN)-based model, both have sound agreement with experimental values.

1. Introduction

These days, Artificial intelligence (AI) approaches are widely well established as tools contribute a voluntary scheme to solve difficult and ambiguous problems. ANNs are data processing methods, and because of their fundamental concept, they have been used in different realms of engineering demands [1]. Such networks may now be developed for number of different prediction application including greenhouse effect modeling, simulation of gas sweetening plant, and evaluation of density at wide range of pressure and temperature [2]. Neural networks have a better refining capability than empirical approaches because of their micro feature concept, because each node decodes only a micro character of the overall input–output pattern. This micro concept signifies that each node only affects the input–output pattern marginally. Just all the nodes are put together into an individual integrated network these micro features outline the macroscopic input–output motif [3–5]. Artificial intelligence has been used for variety application of solubility estimation in ionic liquids and water dew points prediction [6–12].

The purpose of present work is: (1) to experimentally research the solubility of hydrogen sulfide in *N*-methylpyrrolidone (NMP) and compared to that of values reported for other organic solvents reported by our research group [13–17], (2) to employ genetic algorithms (GAs) to determine the number of neurons in the hidden layers, the momentum, the learning rates for minimizing the time and effort required to find the optimal architecture and parameters of the back-propagation algorithm in order to predict the solubility of hydrogen sulfide (H_2S) in *N*-methylpyrrolidone (NMP), (3) to investigate the quality of genetic algorithm in H_2S solubility in physical solvents via comparison with thermodynamical approach such as cubic equation of state [18–20].

Sulphur-contained acid gases are poisonous, corrosive, and flammable and for economical and operational purposes, they have been extremely recommended to be removed from natural gases or biogases streams. Organic solvents or even aqueous solutions of organic solvents have been used for removing the undesirable Sulphur-contained acid gases [15,21–26].

NMP is applicable in commercial solvent formulation in removal of sour gas from natural gas, fuel gas and syngas and has the excellent selectivity for H_2S over CO_2 [23,27].

2. Experimental section

2.1. Materials

The sample information including chemical specifications and origins of hydrogen sulfide and NMP are summarized in Table 1.

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Nomenclature

NIST	National Institute of Standards and Technology
NMP	*N*-methylpyrrolidone
PRSV EoS	Peng-Robinson-Stryjek-Vera equation of state
V_{gc}	volume of the gas container (or gas sample)
V_{EC}	volume of equilibrium cell
P_t	total absolute pressure
P_{VP}	vapor pressure of pure solvent
P_{H2S}	partial pressure of H_2S at equilibrium state
P^e_{ag}	partial pressure of acid gas at equilibrium state in equilibrium cell
n^l_{ag}	amount of acid gas in the liquid phase at equilibrium state
M_S	molar mass of pure solvent
$U\ (n_i)$	uncertainty of amount of species i
m_j	molality of component j, mol·kg^{-1}
$w_{Solvent}$	weight of solvent charged into cell in g
ρ_{ag}	density of acid gas in gas phase at equilibrium state
x_{H2S}	mole fraction of loaded gas
x_s	mole fraction of solvent
T_r	reduced temperature
T_c	the critical temperature
P_c	the critical pressure
$\alpha(T)$	temperature dependent parameter in cubic equation of state
b	co-volume constant
R^2	correlation coefficients
ARD	average of relative deviations
MRD	maximum relative deviation
$h^{(0)}_{H,x}$	the Henry's law constant on mole fraction base
$h^{(0)}_{H,m}$	the Henry's law constant on molality base
$f_{COS}(T,p)$	fugacity of COS in gas phase
γ_x	the activity coefficient of gas on the mole fraction scale
V^{∞}_{H2S}	molar volume of dissolved gas at infinite dilution
$\Delta_{sol}G^{\infty}_{m,x}$	the partial molar Gibbs free energy of solubility at reference state
$\Delta_{sol}H^{\infty}_{m,x}$	the partial molar enthalpy of solubility at reference state
$\Delta_{sol}S^{\infty}_{m,x}$	the partial molar entropy of solubility at reference state

Both chemicals were reagent grade and used without further purification. NMP as solvent for experimental trials was provided by calibrated balance (Mettler model AE 200) with an uncertainty of ±0.0001 g.

2.2. Apparatus and procedure

The experimental method for measuring the gas solubility have been presented elsewhere [28,29]. The whole experimental equipment including gas sample, equilibrium cell, water recirculation bath, temperature and pressure sensors and their uncertainty values sum up in previous paper and only a short description will be provided here.

The double wall equilibrium cell was connected to a water recirculation bath (model T 2500 PMT Tamson) with temperature stability within ±0.02 K and the temperature was measured using a model TM-917 Lutron digital thermometer with a 0.01 K resolution equipped with a Pt-100 sensor inserted into the cell. The equilibrium cell pressure was measured using a model PA-33X KELLER pressure transmitter sensor in the range of (0–25) bar, which was accurate to within 0.01% of full scale, and that of the gas container was measured using a Baroli type BD SENSORS digital pressure gauge in the range of (0–25) bar, which was accurate to within 0.01% of full scale. Equilibration between liquid and vapor phases inside the cell were normally achieved within about 2 h after beginning of stirring. The number of moles of gas in the liquid phase was determined, n^l_{ag} [29–31],

$$n^l_{ag} = \frac{(\rho_i - \rho_f)\cdot V_{gc} - (V_{EC} - V_S)\cdot \rho_{ag}}{1 - V^{\infty}_{ag}\cdot \rho_{ag}} \quad (1)$$

where V_{gc}, V_{EC} and V_S are respectively the volume of the gas container, equilibrium cell and gas free liquid solvent. ρ_i, ρ_f are the mole density corresponding to the initial and final pressures, P_i and P_f, respectively, in the gas container before and after transferring gas. Densities of gas were calculated using NIST Scientific and Technical Databases [32]. ρ_{ag} is the density of acid gas in gas phase in equilibrium cell at equilibrium state (equilibrium temperature T, and partial pressure, P^e_{ag}). The partial pressure of gas in this situation, P^e_{ag}, was calculated as follow:

$$P^e_{ag} = P_t - P_{VP} \quad (2)$$

where P_t and P_{VP}denote the total pressure and vapor pressure of solution. Total pressure obtained from pressure sensor.

The molality and mole fraction of the loaded gas in the liquid phase is defined as:

$$m_{ag} = \frac{n^l_{ag}(\text{mol})}{w_S(\text{kg})} \quad (3)$$

$$x_{ag} = \frac{n^l_{ag}(\text{mol})}{n^l_{ag} + \frac{w_S(\text{kg})}{M_S(\text{kg})}} = \frac{n^l_{ag}}{n^l_{ag} + n_s} \quad (4)$$

In which, M_S and n_s are the molar mass of solvent in kg per mole and the number of mole of solvent, respectively.

The error propagation theory was used to estimate the uncertainties of final results [33]. The main contributions to the uncertainty of the solubility are attributed to errors in the pressure sensor for equilibrium cell and gas container (both are equal to 0.003 MPa), temperature sensors (0.10 K), and scale for the amount of solvent in equilibrium cell (0.001 g). The volumes of the gas sample and equilibrium cell were obtained by performing pressure swing experiments. The pressure swing experiments consisted of measuring the pressure drop when valve between unknown and reference volume get opened, where prior to its opening, the reference volume was pressurized (atmospheric value) and the unknown volume was evacuated. Using the several measurements

Table 1
Sample information used in this work.

Chemical name	Molecular formula	CAS registry number	Purity	Source
Hydrogen sulfide	H_2S	[7783-06-4]	99.95% (mol%)	Roham Gas Company
N-methylpyrrolidone	C_5H_9NO	[872-50-4]	>99.5% (w_t %)	Merck Schuchardt OHG, Germany