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Solubility of hydrogen sulfide in ionic liquids for gas removal processes using artificial neural networks

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ABSTRACT

Artificial neural networks have been used for the correlation and prediction of solubility data of hydrogen sulfide in ionic liquids. The solubility of hydrogen sulfide is highly variable for different types of ionic liquids at the same temperature and pressure and its correlation and prediction is of special importance in the removal of hydrogen sulfide from flue gases for which effective and efficient solvents are required. Several network architectures were tested to finally choose a three layer network with 6, 10 and 1 neuron, respectively (6, 10, 1). Twelve binary hydrogen sulfide + ionic liquids mixtures were considered in the study. Solubility data (pressure, temperature, gas concentration in the liquid phase) for these systems were taken from the literature (392 data points for training and 104 data points for testing). The training variables are the temperature and the pressure of the binary systems being the target variable the solubility of hydrogen sulfide in the ionic liquid. Average absolute deviations are lower than 4.0% and the maximum individual absolute deviation in solubility is 12.6%. The proposed neural network model is a good alternative method for the estimation of solubility of hydrogen sulfide in ionic liquids for its use in process analysis, process design and process simulation.

1. Introduction

The solubility of gases in low-volatility solvents is of importance in several technological applications such as petroleum refining and natural gas processing [1]. In particular, hydrogen sulfide (H_2S) is frequently present as a contaminant in these processes and must be removed, not to affect downstream processes but also to fulfill environmental requirements [2,3]. Alkanolamine–water solutions are currently used in natural gas processes but the increasing of production costs have caused the search for alternative solvents [4], among which ionic liquids represent good candidates due to their especial solvent qualities [5–9].

If ionic liquids are used as solvents to absorb pollutant gases, some mixture properties such as the solubility of the gases in the potential ionic liquid solvents, must be known. Different gas + ionic liquids mixtures have been studied in the literature using various thermodynamic models, mainly equations of state [10–16]. The application of an equation of state (EoS) to mixtures requires knowing the properties of the mixture components and the use of mixing rules. The accuracy in correlating vapor–liquid equilibrium obtained by the EoS method depends mainly on the mixing and combining rules employed to represent the dependency of the EoS parameters on concentration [17]. Usually, mixing and combining rules include binary interaction parameters to obtain more accurate correlation and predictions. The interaction parameters are obtained by fitting experimental phase equilibrium data using an optimization routine. The optimization process may be complex for some cases and sometimes prediction is not accurate enough, so applications to real processes become difficult to implement [18,19]. Therefore, it is necessary to use new techniques to correlate and predict the behavior of the thermodynamic phase mixtures acid gas with ionic liquids, so that solubility values are applicable to different conditions [20]. The development of alternative estimation methods, such as artificial neural networks (ANN), have shown to be very successful for estimating VLE data that are of interest in chemical engineering [20–23].

Studies on the capture of hydrogen sulfide gases by ionic liquids have received some attention and solubility studies at low and moderate pressures have been presented in the literature. Solubility of hydrogen sulfide in ionic liquids has been reported

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Nomenclature

P	Pressure
P_c	Critical pressure
T	Temperature
T_c	Critical temperature
X_1	Solubility (component 1)
X^{calc}	Calculated solubility
X^{exp}	Experimental solubility

Abbreviations

ANN	Artificial neural network
Eos	Equation of state
N	Number of data

Greek letters

%Δ	Percent deviation

Super/subscripts

cal	Calculated
exp	Experimental

by several authors [22–28]. Some of these authors have used the Krichevsky–Kasarnovsky equation to correlate P-T-x data of H_2S in different ionic liquids. The authors showed that most ionic liquids have high solvency to capture H_2S. Recently, Ahmadi et al. [20] used least square support vector machine (LSSVM) to predict hydrogen sulfide solubility in 11 ILs.

In this paper, solubility data of H_2S in ionic liquids have been analyzed using ANN. The variable of interest is the solubility (the concentration of H_2S in the ionic liquid in the range of low concentration) and the independent variables selected for the study are the temperature and the pressure of the mixture and the critical pressure and the critical temperature of the ionic liquid to distinguish between one ionic liquids and another. To the best of the authors' knowledge, this has not been presented in the literature in the form done in this work.

2. Applications of artificial neural networks (ANN)

A neural network is a connectionist computational system in which information is processed collectively, in parallel throughout a network of nodes (the nodes, in this case, being neurons where the information received is processed). Artificial neural networks are based on the way that biological neurons receive process and transmit information and are constructed in structures formed by layers with a certain number of neurons per layer [29,30].

The process of information consists of finding a relation between the target variable (solubility in this study) and the independent variables (temperature and pressure, in this study). The relation between the target variable and the independent variables is established by training the network. In fact, this characteristic of relating the variables by learning is the key element for the popularity that ANNs have gained in several applications. Good descriptions of ANN are given in the literature and a good review on correlation and prediction of fluid properties using ANN has been presented by Taskinen and Yliruusi [31]. Also ANN have been previously used for gas solubility and phase equilibrium modeling in mixtures containing organic compounds [32,33] and also in mixtures containing ionic liquids [34,35].

In a recent paper, Valderrama et al. [36] presented a list of advantages and disadvantages of ANN for the correlation and prediction of fluid properties. Among the advantages they mention the following: (i) a neural network can perform tasks that a linear program cannot; (ii) if an element of the ANN fails, it can continue without any problem; (iii) and models do not have to be specified in advance. Among the disadvantages, the authors mentioned the following: (i) the neural network needs training to operate; (ii) the best architecture must be known or determined by trial and error; (iii) it requires a large amount of accurate data; and (iv) can suffer from over fitting and overtraining.

Correlation of thermodynamic properties of ionic liquids has using ANN has been reported during recent years. Worthy to mention are the studies of Carrera and Aires-de-Sousa [37]; Bini et al. [38]; Torrecilla et al. [39]; Palomar et al. [40]; Valderrama et al. [41,42] and Lashkarbolloki et al. [43,44]. Properties of mixtures containing ionic liquids have also been correlated by Torrecilla et al. [45]; Nami and Deyhimi [46]; and Alvarez and Saldaña [47]. The modeling of solubility data and of Henry's constants using ANN have been presented by Palomar et al. [40], by Eslamimanesh et al. [22] and by Safamirzaei and Modarres [48,49]. Ahmadi et al. [20] have been investigated the capability of ANN trained with back propagation and particle swarm optimization, to correlate the solubility of H_2S in 11 ILs. Recently, Abdollahi et al. [29] analyzed fabrication modeling of industrial CO_2 ionic liquid absorber by artificial neural networks.

Selected papers on VLE calculation in binary mixtures of gas + ionic liquids using ANN have been recently presented by the authors [50]. Those authors show several characteristics of the publications done by other authors in which VLE calculations of gases + IL mixtures are reported (type of mixtures, number of systems, number of experimental points, and number of layers). Important to mention is the observation related to the availability of the software code use to train the network, so other users can reproduce the calculations and obtain the same results. The authors indicate that "*a drawback of most papers describing applications of ANN is that they do not give detailed information (data, architecture, activation functions, weight and bias matrix, or the program codes) to allow other researchers to reproduce the results and to make appropriate use of the ANN model*" [50]. As known, a given ANN architecture cannot reproduce exactly the same results after each run of the network. The supplementary material provided with this paper includes the ANN model and the files required for running the model and obtain the reported results. Additionally, the files containing the data used for training and testing are given. This information will allow any reader to reproduce the results and to predict the solubility of hydrogen sulfide in the systems studied at other temperatures.

Several authors have also used ANN to correlate VLE in binary mixtures of gas + ionic liquids. Nami and Deyhimi [48] analyzed 24 organic solutes in 16 ionic liquids; Eslamimanesh et al. [22] considered mixtures of CO_2 in 24 ionic liquids; Alvarez and Saldaña [47] studied 2 gases and 5 ionic liquids; Safamirzaei and Modarress [48,49] correlated data for mixtures of gases in [bmim] [PF6] and in [bmim] [BF4]. The authors [50] have also correlated and tested gas–liquid equilibrium data for NH_3 in nine ionic liquids. Liquid–liquid equilibrium data have also been analyzed using ANN by some authors. Lashkarblooki et al. [44,51] considered five liquids and seven ionic liquids, while Shojaee et al. [52] modeled water + ionic liquids mixtures. As mentioned above, most of these papers do not provide the weight and bias matrixes, which define the ANN model. Therefore, it is not possible for other researchers to reproduce the results and apply the model for prediction. In this paper, the ANN model, consisting of the program codes to train the network and to predict the solubility of hydrogen sulfide ionic liquids, and also the files containing the data used for training and testing are provided as Supplementary material.