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Correlation of solubility data of ammonia in ionic liquids for gas separation processes using artificial neural networks

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ANN, Artificial neural network
EoS, Equation of state
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%Δ, Percent deviation

ABSTRACT

Artificial neural networks have been used for the correlation and prediction of solubility data of ammonia in ionic liquids. This solubility of ammonia is highly variable for different types of ionic liquids at the same temperature and pressure, its correlation and prediction is of special importance in the removal of ammonia from flue gases for which effective and efficient solvents are required. Nine binary ammonia + ionic liquids mixtures were considered in the study. Solubility data (P–T–x) of these systems were taken from the literature (208 data points for training and 50 data points for testing). The training variables are the temperature and the pressure of the binary systems (T, P), being the target variable the solubility of ammonia in the ionic liquid (x). The study shows that the neural network model is a good alternative method for the estimation of solubility for this type of mixtures. Absolute average deviations were below 5.6%, for each isothermal data set and overall absolute average deviations were below 3.0%. Only in the range of low solubility (below 0.2 in mole fraction) did predicted solubility give deviations higher than 10%.

1. Introduction

Ammonia is a poisonous gas, harmful to people's health and is a severe environment contaminant. However, ammonia is necessary for some physiological and biological processes and is the raw material in petroleum refining and fertilizer manufacture. As an undesired material, the removal of ammonia from flue gases becomes an important issue since traditional ammonia–water absorption systems require additional rectification, increasing costs to prohibited limits [1]. Solvents that can absorb high quantities of ammonia and that do not need expensive additional separation processes represent an interesting alternative. Ionic liquids have shown to be that type of solvents and have received especial attention for process, such as synthesis, separations, catalysis, electrochemistry and waste gas separation [2–6]. Detailed information about various applications of ionic liquids is available in the literature [7–13]. The capture of ammonia by ionic liquids has also received some attention in the literature [1,12,13]. Experimental data show that solubility of ammonia in ionic liquids cover different ranges for different types of ionic liquids at the same temperature and pressure, and for several ionic liquids, solubility can be as high as 80% in mole fraction.

Different gas + ionic liquids mixtures have been studied in the literature using various thermodynamic models, mainly equations of state [16–20]. The application of an equation of state (EoS) to mixtures requires the use of

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Nomenclature

P	Pressure
P_c	Critical pressure
T	Temperature
T_c	Critical temperature
x_1	Solubility (component 1)
x^{cal}	Calculated solubility
x^{exp}	Experimental solubility

Greek symbols (super/subscripts)

cal	Calculated
exp	Experimental

mixing rules to represent the dependency of the EoS parameters on concentration and combination rules to represent the interaction between the unlike components in the mixture. The accuracy in correlating vapor–liquid equilibrium obtained by this method depends on the EoS used and the mixing and combining rule employed [21]. Also, binary interaction parameters must be introduced to obtain more accurate results. Such interaction parameters are obtained by fitting experimental phase equilibrium data at each temperature at which VLE is required. Considering also the difficulties of experimental measurements, besides the high cost in some cases, the development of alternative estimation methods, such as artificial neural networks have shown to be very successful for estimating VLE data that are of interest in chemical engineering [22–25].

For the specific case of ammonia + ionic liquids mixtures, some experimental studies at low and moderate pressures have been presented in the literature. Yokozeki and Shiflett [14,15] determined pressure–temperature–composition (P–T–x) solubilities of ammonia at room temperature ILs. The authors presented phase equilibrium data of four NH_3 + IL mixtures: NH_3 with [C4mim][BF4], [C4mim][PF6], [C6mim][C1] and [C2mim][Tf_2N]. In another work, Yokozeki and Shiflett [15] reported P–T–x solubility of ammonia in [C2mim][Ac], [C2mim][SCN], [C2mim][EtOSO3] and [DMEA][Ac]. In both works, the authors observed high solubilities of ammonia in these ionic liquids. More recently, Li et al. [1] reported experimental solubilities of ammonia in four imidazolium-type ILs: [C_2mim][BF4], [C_4mim][BF4], [C_6mim][BF4] and [C_8mim][BF4]. The authors showed that all these ionic liquids have high solvency to capture NH_3. To the best of the authors knowledge, specific applications, such as the one done in this work, in which P–T–x equilibrium data of ammonia + ionic liquids are correlated using artificial neural networks, have not been presented in the literature.

2. Application of artificial neural networks

Artificial neural networks are a computational model inspired in the behavior of natural neurons. A structure of neurons organized in different layers (known as architecture) receives data related to a given property, solubility for instance, and some independent variables that are supposedly related to the dependent main variable (temperature and pressure, for instance). The input and output variables are weighed by weights and shifted by a bias factor specific to each neuron. By optimization, the network learns the relation between the variables and stores the values of the weights and biases that give the lowest error between calculated and experimental data of the dependent variable (solubility in this work).

The artificial neural networks are "neural" in the sense that they have been inspired by neuroscience but not necessarily because they are faithful models of biological neural or cognitive phenomena. A neural network is characterized by:

- its pattern of connections between the neurons (the architecture);
- its method of determining the weights on the connections (training or learning process);
- its activation function (relation between dependent and independent variables).

Good descriptions of ANN are given in the literature [26].

Taskinen and Yliruusi [27] presented a complete list of properties, mostly for organic substances, that have been analyzed in the literature, until 2003, using different approaches of ANN. Properties, such as normal boiling point, critical temperature, critical pressure, vapor pressure, heat capacity, enthalpy of sublimation, heat of vaporization, density, surface tension, viscosity, thermal conductivity, and acentric factor, among others, were thoroughly reviewed. Also, ANN has been previously used for gas solubility and phase equilibrium modelling in mixtures that do not include ionic liquids [28–32].

Applications of neural networks for the prediction of various thermodynamic properties of ionic liquids have been reported in a number of papers during the last ten years. Melting temperature have been studied by Carrera and Aires-de-Sousa [33], Bini et al. [34] and Torrecilla et al. [35], while other properties, such as density, viscosity and heat capacity have been modelled by Palomar et al. [36], Valderrama et al. [37,38] and Lashkarbolloki et al. [39,40]. Other authors have used ANN to model mixture properties, such as concentrations, infinite dilution activity coefficients [41–43]. Solubility modeling and Henry's law correlation using ANN have been done by Palomar et al. [36], Eslamimanesh et al. [25] and Safamirzaei and Modarres [44,45].

Table 1 shows selected papers on VLE calculation in binary mixtures of gas + ionic liquids using ANN. In the table, the type and number of systems studied, the number of data treated, the variable being correlated and predicted, the optimum architecture, and the type of information provided by the authors are clearly indicated. The symbol (*) in the last column indicates papers that provide the weight and bias matrixes, which correspond to the ANN model. As shown in the table, 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