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RAPID COMMUNICATION

Application of artificial neural network for vapor liquid equilibrium calculation of ternary system including ionic liquid: Water, ethanol and 1-butyl-3-methylimidazolium acetate

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Abstract−A feed forward three-layer artificial neural network (ANN) model was developed for VLE prediction of ternary systems including ionic liquid (IL) (water+ethanol+1-butyl-3- methyl-imidazolium acetate), in a relatively wide range of IL mass fractions up to 0.8, with the mole fractions of ethanol on IL-free basis fixed separately at 0.1, 0.2, 0.4, 0.6, 0.8, and 0.98. The output results of the ANN were the mole fraction of ethanol in vapor phase and the equilibrium temperature. The validity of the model was evaluated through a test data set, which were not employed in the training case of the network. The performance of the ANN model for estimating the mole fraction and temperature in the ternary system including IL was compared with the non-random-two-liquid (NRTL) and electrolyte non-random-two-liquid (eNRTL) models. The results of this comparison show that the ANN model has a superior performance in predicting the VLE of ternary systems including ionic liquid.

Key words: Vapor-liquid Equilibrium, Ionic Liquid, Ternary System, Artificial Neural Network

INTRODUCTION

Ionic liquids (ILs) are being investigated because of their potential as alternatives for organic solvents. They have unique physical properties such as high thermal stability, low melting point, non-flammability, high solubility for polar and non-polar organic and even inorganic substances and negligible vapor pressure [1]. ILs are essential as entrainers in extractive distillation in order to separate azeotropic or close boiling mixtures [2,3], as an appropriate solvent in the separation and the extraction processes, for example extraction of aromatic hydrocarbons from mixtures of aromatic-aliphatic hydrocarbons and recovery of amino-acids from aqueous media, and as a solvent or a catalysis component for chemical reactions, membrane separation processes where ILs are used as in-between porous support membranes, etc. [4,5]. They typically include large organic cations and smaller inorganic or organic anions. There is no limit to the number of possible cations and anions of ILs. Therefore, they are named as "designer solvents" for chemical reactions, as solvents for electrochemical applications or as selective solvents (entrainers) in separation processes [6-8]. There is some literature with detailed information about the synthesis and application of ILs [9,10].

When an IL is introduced into a mixture of volatile compounds in order to separate them, the non-ideality and the activity coefficients of the components are changed and they are affected to a different extent. It is desired that these changes result in an increase of relative volatility. The phase equilibrium behavior of the systems containing ILs is fundamental for studies and the design of extractive distillation processes, when an IL is used as entrainer.

It is necessary to understand the vapor-liquid phase behavior of a mixture containing ILs, especially the increase of the relative volatility by addition of IL. For a better understanding of their thermodynamic behavior and for the development of a new thermodynamic model, trustworthy experimental phase equilibrium data are required. Therefore, several research groups started with systematic measurements of the required properties of the ILs. For example, Doker et al. [11] researched measuring and prediction of vapor liquid equilibrium of ternary systems containing ionic liquids. Shen et al. [12] studied the effect of ionic liquid 1-methylimidazolium chloride on the vapor liquid equilibrium of water, methanol, ethanol, and water+ethanol mixture. Banerjee et al. [13] modeled the ternary systems including the following ILs: [bmim][PF_6], [omim][Cl], [emim][BF_4], [bmim][BF_4], [hmim][BF_4], [omim][BF_4], [bmim][PF_6], [hmim][PF_6] and [bmim][TfO]; Simoni et al. [14] correlated six ternary systems including the ILs [emim][BF_4], [emim][$EtSO_4$], [emim][Tf_2N], [dmim][Tf_2N], [bmim][PF_6] and [bmim][Tf_2N]; Pereiro and Rodriguez [15,16] correlated ternary systems containing the ILs [hmim][PF_6] and [omim][PF_6]; Heintz [17] and Vega et al. [18] presented excellent reviews of phase equilibria of ILs.

In the present study, the vapor-liquid equilibrium for a ternary system containing IL has been investigated. Owing to the low viscosity of 1-butyl-3-methylimidazolium acetate ([bmim][OAc]), which is an attractive solvent for heat and mass transfer in the extractive distillation, we will extend the modeling to the system water (1)+ethanol (2)+[bmim][OAc] (3). Such information is mainly obtained from the experimental measurement of vapor-liquid equilibrium (VLE) data that were done by Deng et al. [19]. The main goal is obtaining the mole fraction of ethanol in vapor phase in the IL-containing mixture in a wide range of IL mass fractions.

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The most convenient way to reach the compositions in vapor phase is to develop the best function of liquid phase compositions. The artificial neural network (ANN) model can be used to predict the compositions in vapor phase over a wide range of the mole fraction of components.

ANN is a suitable method for estimating complex functions in order to evaluate the equilibrium [20] and the transport properties of the mixtures such as solubility [21], diffusion coefficient [22] and thermal conductivity [23]. This work presents a feed forward ANN model, trained by the back propagation algorithm, to predict the characteristics in vapor-liquid equilibrium of a system containing ionic liquid. It predicts the composition of the ethanol in vapor phase and temperature of systems (bulb temperature calculation) as target via mass fraction of IL; the mole fraction of ethanol in the liquid phase and pressure are input data. The modeling is based on T, x, y data in a wide range of IL mass fractions and in a relatively complete composition range for ethanol.

Different excess Gibbs free energy models, such as NRTL, UNIQUAC and NRTL-NRF [24,25], are used and applied successfully for ternary IL-containing systems. The results of this work were compared with NRTL and eNRTL models and a satisfactory advantage was found.

METHODOLOGY

Artificial neural networks are computational systems which mimic the computational abilities of biological systems by using a number of interconnected artificial neurons [26]. Considering the inherent ability of the ANNs to learn and recognize nonlinear and complex relations, they can be used in various fields of chemical engineering [27]. The ANNs consist of a number of interconnected nodes arranged in layers corresponding to input layer, hidden layer and output layer as shown in Fig. 1. The hidden layers encode and arrange the information received from the input layer and deliver them to the output layer. Each neuron of the network is connected with an associated weight, to the others via direct communication links, which finally provides a logical relationship between input and output parameters. The number of neurons for the input and output layers is usually determined by the number of input and output variables, respectively. However, the number of neurons in the hidden layers is variable and it is significant for optimization of the network, which will be explained in the following sections.

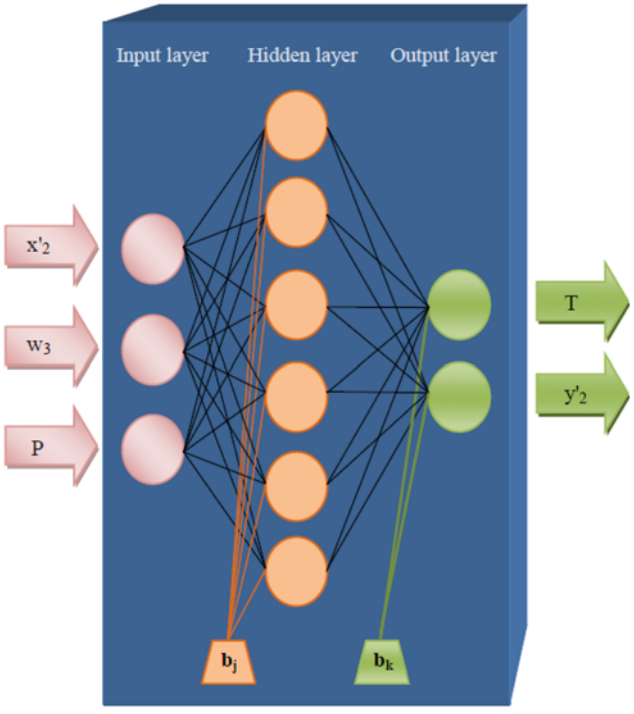

Fig. 1. The layout of the ANN architecture.

The ANN used in this study is a multilayer feed forward neural network with a learning order of the back propagation (BP) of errors and the Levenberg-Marquardt algorithm [28-30] for the correction of the connecting weights. The following steps are those required to develop the neuromorphic model to predict characteristics of VLE.

The first step in the modeling, which is one of the important decisions in the development of the neuromorphic model, is compiling an adequate database to train the network and to evaluate its capacity for generalization. Many researchers by various techniques have measured VLE data. Comprehensive analysis on the VLE data has also been published [31]. In the present work, experimental VLE data for an IL-containing mixture consisting of water (1), ethanol (2) and [bmim][OAc] (3) were taken from Deng et al. [19] and were used for training and testing the ANN model. We have not used the VLE data given in some publications that are based on the approximation correlations, and all data points used in this study are the experimental data.

The investigated system was at the equilibrium condition. There were three components in the liquid phase, water (1), ethanol (2) and IL [bmim][OAc] (3).The VLE measurements were done in a way that the IL mass fraction, w_3, changed from high to low value, while the mole fraction of ethanol on IL-free basis, x_2', remained approximately unchanged in each group of the data sets. So, there were six groups that the mole fractions of ethanol on IL-free basis in those are approximately 0.1, 0.2, 0.4, 0.6, 0.8 and 0.98. When the equilibrium was recognized, the vapor condensate was sampled and analyzed. As the IL is nonvolatile, the vapor phase only consisted of water (1) and ethanol (2). In the reference of these experimental data, vapor-phase composition was obtained by analyzing the water content by using the Karl-Fischer method. Then vapor-phase compositions were calculated from the ratio of mixing and the water content. The accuracy of the vapor-phase composition was estimated to be 0.0001 in water mole fraction. Liquid-phase compositions were calculated on the basis of mass balance, using a procedure obtained in the author's previous work [32,33]. A set of 48 experimental data points was used to develop the neuromorphic model.

The data were divided into two parts: training data set and test data set. Two-thirds of the total data were selected for training the ANN and the other were used as the test data set. The training data set was chosen in order to cover all the composition range of the all components in the experimental data.

After collecting and classifying the data set, the next step is the selection of input variables, which are the model's independent variables. The available equations for prediction of the compositions at VLE are essentially based on the assumption that composition of a component in vapor phase and temperature can be described as a

function of pressure and compositions of compounds in liquid phase as follows:

$$(T, y'_2)=f(x'_2, w_3, p) \quad (1)$$

According to Eq. (1), in this study, the ANN input data were the mole fraction of the second component and mass fraction of IL both in the liquid phase, and the pressure of the system. The output of the ANN will be the mole fraction of the second component in vapor phase and the temperature of the system.

The experimental data were measured in six mole fractions of ethanol on IL-free basis (0.1, 0.2, 0.4, 0.6, 0.8, and 0.98) and each mole fraction consists of eight data points (totally 48 data points). Two mole fractions of data points, 0.2 and 0.8 (16 data points), were considered as the test data. The training set consisted four mole fraction data group (0.1, 0.4, 0.6 and 0.98), and it covered low and high composition range of the all components in the experimental data.

1. ANN Modeling and Training

In the last few years more than a few techniques related to the network pruning methods have been provided such as optimal brain surgeon method (OBS) [34,35] and optimal brain damage (OBD) [36] These methods are known as pruning methods.

As far as training of the networks, the input and output have various physical units and range, so it is usually useful to normalize all of the input and output before training. Therefore, all data were normalized in the range of 0-1 to avoid any computational difficulty, by use of the following relation:

$$\text{Normalized data}=\frac{\text{data value}-\text{minimum value}}{\text{maximum value}-\text{minimum value}} \quad (2)$$

Once the input and output have been determined, the next step is to develop the ANN architecture. The neural network toolbox of the MATLAB programming language was used for the ANN modeling. A network with one hidden layer was selected as the network structure. In this work a fixed number of 45 iterations were used as the stopping criterion. This number was obtained through experience, so that the difference between of error values of train and test data set is not so much. In each run of the MATLAB script, different parameters (solutions) were obtained due to the different values of initial parameters. To achieve the best results of parameters, the commonly used method was applied that has already been used in the literature [37]. Because the initial weights may have a large effect on the convergence, runs were repeated 100 times with different randomly generated initial values. The results presented in this work are the best ones obtained from this procedure. Theoretical procedures for choosing the appropriate number of hidden layers are not available. Therefore, the number of neurons in each hidden layer cannot be obtained theoretically. So the trial-and-error method is commonly used to design the neural network. A small number of neurons are used in the hidden layer, and if the error of the trained ANN does not reach the desired tolerance, the number of neurons in the hidden layer is increased and training cycle and performance estimation is repeated [34]. This process is continued until the trained network performs satisfactorily (its training and testing errors are lower than the target aim). Usually when a model is complicated, an increase in the number of neurons in the hidden may cause over fitting. In complicated neural networks, the optimal number of neurons should be determined. Applying the procedure to design the ANN model for the estimation of calculation of VLE of the IL-including system led to a three-layer network with three neurons in input layer and two neurons in output layer.

The final output of the ANN is calculated from the following equation [38]:

$$Y_k=F_l\left\{\sum_{j=1}^{n}W_{kj}\left[F_t\left(\sum_{i=1}^{m}W_{ji}X_i+b_j\right)\right]+b_k\right\} \quad (3)$$

where Y, X, W, b, n, m are the final answer of the network, input value of the network, the weight value, the bias, the number of neurons, the number of input variables; 'i', 'j' and 'k' refer to the input, hidden, and output layer, respectively. F is the transfer function that transforms the sum of the weights and the bias to get the normalized output values. Many types of transfer functions can be used. In this work the "hyperbolic tangent sigmoid" transfer function was considered for the hidden layer and "linear" function was used for the output layer. These functions are defined as follows:

$$F_t(x)=\frac{e^{x}-e^{-x}}{e^{x}+e^{-x}} \quad (4)$$

$$F_l(x)=x \quad (5)$$

In the BP training method, the connection weights W_{ij} and the biases b_j are iteratively adjusted to minimize the output deviation (between predicted by the ANN and the experimental data) from the target values. Moreover, the error values should be calculated for different numbers of neurons in hidden layer.

RESULTS AND DISCUSSION

In this research, the variables considered as the input of the network were selected according to the models that have been previously presented. The ANN with different architectures possibly makes different outputs, so there is more chance to reach the best answer with more trial and error. In this study, various numbers of neurons were used for the hidden layer, and the optimal number of neurons was evaluated. The aim functions, which were used for selecting the best ANN architecture, are the mean relative error (MRE) and root mean square error (RMSE), defined as follows:

$$\text{MRE}(\%)=\frac{100}{N}\sum_{i=1}^{N}\left(\frac{|t_i-y_i|}{t_i}\right) \quad (6)$$

Table 1. Modeling results obtained from different numbers of neurons of the hidden layer

Number of neurons	RMSE		MRE	
	y'_2	T/K	y'_2	T/K
3	0.0264	1.14	3.60	0.27
4	0.0109	0.52	1.25	0.13
5	0.0104	0.53	1.12	0.11
6	**0.0047**	**0.27**	**0.49**	**0.06**
7	0.0070	0.45	0.72	0.08
8	0.0050	0.58	0.41	0.12
9	0.0070	0.38	0.52	0.06
10	0.0063	0.43	0.54	0.07
11	0.0089	0.45	0.70	0.07

Table 2. Parameters (weights and biases) of the ANN

Neuron	W_{ji}			b_j	W_{kj}		b_k	
	x_2'	w_3	P/kPa		y_2'	T/K	y_2'	T/K
1	0.2578	2.9406	0.3650	−3.3300	0.096034	1.4498	0.2942	1.9935
2	2.7880	−1.2981	−0.9073	−2.4207	0.27976	0.16243	-	-
3	2.3743	−0.5801	−4.0784	1.5172	0.80601	0.3534	-	-
4	4.0926	0.4844	0.4564	−2.6109	0.17357	0.005199	-	-
5	1.7253	−1.0577	−5.2407	3.2365	−0.74795	−0.65015	-	-
6	−4.1586	0.27493	0.06038	−0.7895	−0.70732	0.35049	-	-

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(t_i - y_i)^2} \quad (7)$$

where N, t and y are the number of data points, the target (experimental) data, and the ANN predicted values, respectively.

The results of the method for the purpose of the optimum number of neurons in the hidden layer have been presented in Table 1. MRE and RMSE values of the trained network have been shown for the different number of the neurons of the hidden layer. The configuration with minimum measured error (MRE and RMSE), is selected as the best network architecture. According to Table 1, the best neural network configuration has one hidden layer with six neurons. The RMSE and MRE values of the ANN for the prediction composition of the ethanol in the vapor phase were 0.0047 and 0.49, and for the temperature of the system were 0.27 and 0.06, respectively. The parameters (weight and bias values) of the best selected neural network are tabulated in Table 2.

In addition to the Levenberg-Marquardt algorithm (trainlm), other algorithms such as traincgb (conjugate gradient back-propagation with Powell-Beale restarts), trainbr (bayesianregularization back-propagation) and traingda (gradient descent with adaptive learning rate back-propagation), were used to train the network. Table 3 shows the RMSE and MRE achieved for each algorithm with optimum hidden neurons and same iterations. The obtained errors in the Table 3 demonstrate that using the Levenberg-Marquardt algorithm leads to the best situation.

The correlation between the simulation results for prediction of mole fraction of vapor phase for the developed ANN and the experimental training data points is illustrated in Fig. 2. The perfect fit (output equal to targets) is shown by the solid line. The close proximity of the best linear fit to the perfect fit, as observed in Fig. 2, shows a good correlation between the network predictions and the experimental data. Also, the performance of the developed network has been evaluated through using the test data set consisting of one-third of data points, which was not previously used for the network training. It can also be seen in Fig. 2. The evaluations indicate that the MRE and RMSE for the training data are 0.29 and 0.0024, respectively, and for the test data are 0.92 and 0.0073, respectively.

Table 3. Best RMSE and MRE values of different training algorithm with optimum hidden neurons

Training algorithm	Number of optimum hidden neurons	RMSE		MRE	
		y_2'	T/K	y_2'	T/K
Trainlm	6	**0.0047**	**0.27**	**0.49**	**0.06**
Traincgb	9	0.0409	2.71	6.10	0.58
Trainbr	7	0.0152	0.55	1.79	0.12
Traingda	9	0.2491	20.39	27.13	4.86

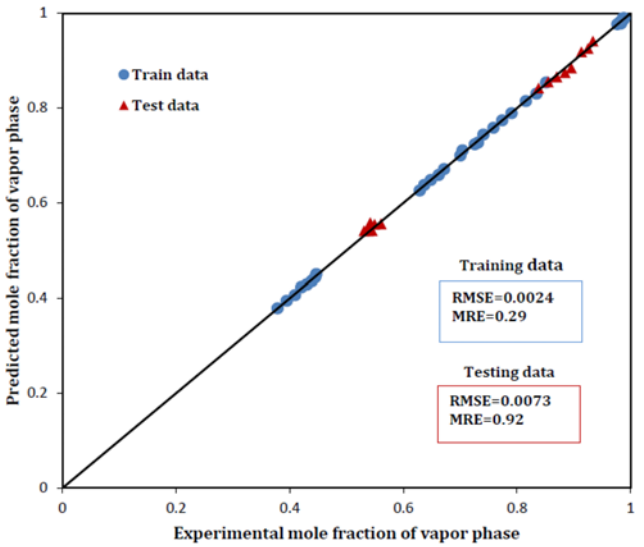

Fig. 2. Comparison between predicted and experimental mole fraction of vapor phase (y_2').

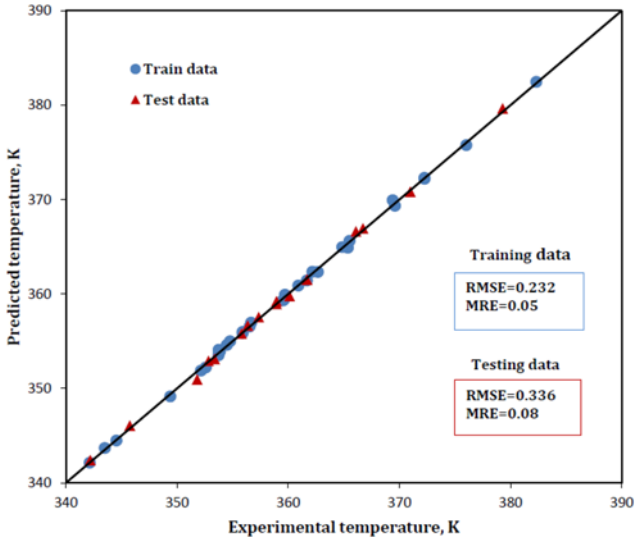

Fig. 3. Comparison between predicted and experimental temperature (T/K).

Table 4. RMSE of the ANN, NRTL and eNRTL models for predicting T and y_2'

Model	RMSE	
	y_2'	T/K
ANN	0.0047	0.27
NRTL[a]	0.0095	0.6
eNRTL[a]	0.008	0.75

[a]Extracted from ref. [19]

In the same way, Fig. 3 shows a comparison between predicted and experimental data for temperature, which is the second purpose of the network. The results indicate that the MRE and RMSE for the training data are 0.05 and 0.0232, respectively, and for the test data are 0.08 and 0.336, respectively. This means that the network was also suitable for predicting the data points which are not used in the training data set.

To have a comparison between the neural network model developed in the present work with others' correlations, there is an estimation of the outputs in the data set by using the developed network as well as NRTL and eNRTL thermodynamic models. Table 4 shows the results of this comparison and the RMSE values of each method separately for composition of ethanol and temperature.

By using the NRTL [39] and eNRTL [40] equations, modeling of the ternary VLE is performed. For the eNRTL equation, Vercher et al. [41] have offered expressions for the liquid-phase activity coefficients of volatile components in a ternary system containing a salt. In the correlations, from literature [42], the binary parameters for water and ethanol mixture are given. With the non-randomness factors and at the most common value of 0.3 for NRTL equation and 0.2 for eNRTL equation, the binary parameters for the ternary system are achieved by minimization of the following function:

$$F = \sqrt{\sum_{n=1}^{N}(y_{1,cal}/y_{1,exp}-1)^2/N} + \sqrt{\sum_{n=1}^{N}(y_{2,cal}/y_{2,exp}-1)^2/N} \quad (8)$$

In which N is the number of data points. The obtained parameters are used for calculation of the ternary VLE data. The NRTL and eNRTL equations provide good correlation for the IL-containing mixture in the experimental composition range [19], but the ANN method's accuracy is better than the accuracy of both of the two well-known models. The degree of accuracy of the ANN model for this system proves the capability of the ANN model for estimating VLE data of IL containing mixtures.

Moreover, two separate ANNs for prediction of T and y_2' were developed. A structure of 3-5-1 with MRE of 0.48 and RMSE of 0.0043 for prediction of T and a network structure of 3-6-1 with MRE of 0.037 and RMSE of 0.25 for estimation of y_2' leads to the best results, respectively. The weight and bias values of the best selected neural networks are given in Tables 5 and 6.

Table 5. Parameters of the ANN for prediction of y_2'

Neuron	W_{ji}			b_j	b_k=−0.88879
	x_2'	w_3	p/kPa		W_{kj}
1	3.4060	−1.2527	−0.2442	0.1039	−2.8644
2	−1.8907	−0.5202	−0.1096	−0.3990	1.4530
3	−4.6681	4.0674	2.9735	0.0056	−0.1906
4	−4.3893	0.3004	−5.2582	0.0340	3.4423
5	3.2798	−0.3297	0.0945	1.6881	1.2770

Table 6. Parameters of the ANN for prediction of T

Neuron	W_{ji}			b_j	b_k=1.4874
	x_2'	w_3	p/kPa		W_{kj}
1	−0.7998	−2.4876	0.2670	2.3527	−0.73541
2	−5.2497	1.5602	0.9721	1.8981	0.027654
3	1.6707	−2.2709	1.8963	−0.8650	−0.13133
4	−3.9384	−2.3725	−6.1637	3.6276	0.069823
5	−0.4709	−6.4363	−10.5112	7.1860	−0.58018
6	−4.3822	−0.0869	−0.2694	−0.9771	0.89246

These error values of prediction results for the two separate ANN show a high accuracy in comparison with the results obtained using only one ANN for both output variables (T and y_2'). On the other hand, a total of 55 weights and biases were employed in the two separate ANNs, while 38 weights and biases were used in the single ANN. Each ANNs can be chosen according to the intended purpose and application.

CONCLUSIONS

An artificial neural network (ANN) has been developed for the prediction of VLE data in ternary system including ionic liquid. The system includes water, ethanol and 1-butyl-3-methylimidazolium acetate, and the ANN predicts the composition of ethanol in vapor phase and temperature as a function of mass fraction of IL and composition of ethanol in liquid phase and the pressure of the system. A set of 48 experimental data points was used for the network training. The best architecture is the feed forward neural network, obtained by trial and error, which consists of one hidden layer with six neurons. To predict ethanol composition in vapor phase, the mean relative error (MRE) and root mean square error (RMSE) of the designed network for training data points are 0.49 and 0.0047, respectively, and for prediction of temperature are 0.27 and 0.06, respectively. The performance of the proposed ANN model was also tested through its application to a test data set consisting of one-third of the experimental data not used in the training. The results of this estimation indicate that the developed ANN model will be able to predict VLE data with a lower MRE and RMSE errors than that of the other well-known thermodynamic models. The results of applying the trained ANN model to the test data show that the method has also a very good prediction of VLE calculation in the mixtures containing IL.

NOMENCLATURE

b : bias
F : transfer function
IL : ionic liquid
m : number of input variables
n : number of neurons
N : number of data points
p : pressure [kPa]
t : target

T : temperature [K]
w : fraction
W : weight
x : mole fraction of liquid phase
X : input value of the network
y : predicted values/ mole fraction of vapor phase
Y : final answer of the network

Superscripts

′ : prime symbol, indicating the quantity on IL-free basis

Subscripts

1, 2, 3 : volatile (1, 2) or nonvolatile (3) component
i : input layer
j : hidden layer
k : output layer

Abbreviations of Ionic Liquids

[bmim][PF_6] : 1-butyl-3-methylimidazolium hexafluorophosphate
[omim][Cl] : 1-octil-3-methylimidazolium chloride
[emim][BF_4] : 1-ethyl-3-methylimidazolium tetrafluoroborate
[bmim][BF_4] : 1-butyl-3-methylimidazolium tetrafluoroborate
[hmim][BF_4] : 1-hexyl-3-methyl-imidazolium tetrafluoroborate
[omim][BF_4] : 1-octil-3-methylimidazolium tetrafluoroborate
[bmim][PF_6] : 1-butyl-3-methylimidazolium hexafluorophosphate
[hmim][PF_6] : 1-hexyl-3-methyl-imidazolium hexafluorophosphate
[bmim][TfO] : 1-butyl-3-methylimidazolium trifluoromethanesulfonate
[emim][$EtSO_4$] : 1-ethyl-3-methylimidazolium ethylsulfate
[emim][Tf_2N] : 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imid
[dmim][Tf_2N] : 1,2-dimethylimidazolium bis(trifluoromethylsulfonyl)imide
[bmim][Tf_2N] : 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide
[bmim][OAc] : 1-butyl-3-methylimidazolium acetate

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