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Prediction of refractive index of binary solutions consisting of ionic liquids and alcohols (methanol or ethanol or 1-propanol) using artificial neural network

Allan N. Soriano[a], Karen Faith P. Ornedo-Ramos[a], Carla Angela M. Muriel[a], Adonis P. Adornado[a], Vergel C. Bungay[b], Meng-Hui Li[c,*]

[a] School of Chemical Engineering and Chemistry, Mapúa Institute of Technology, Intramuros, Manila, Philippines
[b] Chemical Engineering Department, De La Salle University, Taft Avenue, Manila, Philippines
[c] R&D Center for Membrane Technology and Department of Chemical Engineering, Chung Yuan Christian University, Chung Li, Taiwan

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ABSTRACT

In recent years, a new class of solvent called ionic liquids had successfully demonstrated potential applications in industrial chemistry and chemical technology due to its desirable properties. To this end, understanding their physico-chemical properties is of high importance. The current study presents a model for predicting the refractive index of binary ionic liquid system containing alcohol (methanol or ethanol or 1-propanol) using the artificial neural network (ANN) algorithm. The refractive index data were correlated as function of temperature, mole fraction, number of carbon atoms in the cation, number of atoms in the anion, number of hydrogen atoms in the anion and number of carbon atoms in the alcohol. Refractive index data from ThermoIL Database were used. Using ANN, a total of 752 data points were used in the calculation and to obtain the optimum neural network parameters. The 6-6-9-1 neural network architecture was found to be the best network using two hidden layers as shown by mean absolute error of 0.00783 and an overall average percentage error of 0.55%. The obtained correlation satisfactorily represents the experimental refractive index data and can be reliably used to predict the refractive index of other binary systems containing the considered cation and anions and the studied alcohols.

1. Introduction

Novel solvent called ionic liquids (ILs) had been the interest of some researchers in recent years due to their desirable properties such as negligible vapor pressure, non-flammability, large range of densities and viscosities, high potential for recycling, and highly solvating capacity for organic, inorganic and organometallic compounds [12]. ILs have found their possible applications in industrial chemistry and chemical technology as solvents for chemical synthesis and catalysis, gas–liquid absorption, liquid membrane separations, fuel and solar cells, nanomaterial technologies, preparation of polymer-gel catalytic membranes, and generation of high conductivity materials [11]. These applications resulted to the need of understanding the properties of these solvents; properties as basic as density, refractive index, viscosity, heat capacity, etc. Knowledge of these properties is required in both physical chemistry and chemical engineering calculations involving fluid flow and heat and mass transfer analysis [15].

Experimental studies seem to be expensive as the prices of most ILs are still high so the best options to study its properties is via computer simulation. Predictive computing is one way of evaluating these properties using available experimental data from reliable database in generating model for predicting these properties [13] and studying their molecular behaviors [14]. In these calculations, one of the essential factors to consider is the choice of computer algorithm such as the artificial neural network (ANN). The ANN algorithm has been used extensively for several applications, e.g. electronics [9,26,32], forecasting [2,10,28], pharmaceutical research [1], financial institutions [39], etc. but limited use of such algorithm has been made for chemical engineering applications.

The ability to predict the physical properties of ionic liquids is extremely important for the rational design of proper ionic liquids with specific properties. In practice, processes involving ionic liquids are usually mixed with one or more compounds to enhance their properties. Therefore, in addition to pure component properties, knowledge of the physical properties of binary or ternary mixtures is also crucial for various applications.

* Corresponding author. Tel.: +886 3 265 4109; fax: +886 3 265 4199.
E-mail address: mhli@cycu.edu.tw (M.-H. Li).

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Nomenclature

A	parameter of Eq. (11) which is a function of weights and bias weights
$B_{n'}$	parameter of Eq. (11) which is a function of weights and bias weights
C_n	parameter of Eq. (11) which is a function *T*, *X*, *NCC*, *NA*, *NH*, *NCA*
E_l	experimental or actual value
MAE	mean absolute error
MSE	mean square error
n'	number of nodes in the hidden layer 2
N	number of nodes in the hidden layer 1
NA	number of atoms in the anion
NCA	number of carbon atoms in the alcohol
NCC	number of carbon atoms in the cation
NH	number of hydrogen atoms in the anion
T	absolute temperature (K)
($w_{b\text{-}n}$, $w_{b\text{-}n'}$, or $w_{b\text{-}Y}$)	empirical parameters of ANN called bias weights
($w_{l'\text{-}n}$, $w_{n\text{-}n'}$, or $w_{n'\text{-}Y}$)	empirical parameters of ANN called weights
X	mole fraction of the IL
Y_l	predicted value
Y_l'	output node

Lashkarblooki et al. [27] have proposed a neural network model able to well predict the ternary viscosity data points by using the predicted values of boiling temperatures of ionic liquids with considerably high accuracy (AARD% = 1.94%, MSE = 2.45×10^{-8}, AAD% = 0.87% and R2 = 0.9998 for all data points). In other publication, Hezave et al. [22] have investigated the applicability of the ANN tool to predict the electrical conductivity of the ternary mixtures of 1-butyl-3-methylimidazolium hexafluorophosphate ([bmim][PF_6]) + water + ethanol and [bmim][PF_6] + water + acetone in the temperature range from 288.15 K to 308.15 K. Results showed a very good agreement between the experimental data points and the predicted values by the optimum network (AARD% = 1.44%, MSE = 2.87×10^{-9} and R^2 = 0.9981). The succeeding year, the same group correlated the bubble points of several ternary mixtures containing an ionic liquid using an artificial neural network modeling approach. The overall average absolute relative deviation (AARD%), mean square error (MSE), maximum deviation (E_{max}), minimum deviation (E_{min}) and correlation coefficient (R2) were calculated to be 0.20%, 0.9953, −0.97, 0.87 and 0.95, respectively [23].

Although the properties of some ILs and ILs mixtures were reported in the literature, but the data points measured using experimental methods are still not fulfilling the requirements for their broad application, especially, due to the lack of data of IL homologs which would be helpful to improve the selection of more appropriate test candidates for different applications. Besides, experimental measurements are not always possible, in some cases they are time consuming and relatively expensive especially if they are needed as function of temperature. To overcome this problem, estimation methods have been developed using limited available data points. With the advancement of technical computing and in conjunction with the fast growing number of binary systems containing ILs with organic solvents, it is required to correlate or predict some important thermophysical properties of ILs. Thus, it is the main aim of this work to develop a model for predicting the refractive index of binary ionic liquid system containing alcohol (methanol or ethanol or 1-propanol). Specifically, it determines the best neural architecture using the ANN along with the parameters associated with it, and it predicts the refractive index of the studied systems from the calculated empirical parameters.

2. Methods

The conceptual framework of the study leading to the prediction of refractive index is shown in Fig. 1. The details of each block were briefly discussed in the succeeding paragraphs.

2.1. Data gathering and data trimming

Since this work is purely computational, all data (experimental) needed for the calculations were taken from ThermoIL Database. The ions comprising the considered ionic liquids are presented in Tables 1a and 1b. A total of 806 experimental data points for refractive index were gathered in which the details are presented in Tables 2–4, including the temperature range, pressure, mole fraction range, number of data points and the references. The gathered data were then trimmed (removal of inaccurate and unreliable data). The details of the data trimming process applied were adopted from previous works [33,34].

2.2. Artificial neural network (ANN) algorithm

Fig. 2 shows the architecture used for the ANN algorithm. The architecture consists of one input layer followed by hidden layer/s and an output layer being the last layer. The nodes in every layer were connected by weights ($w_{l'\text{-}n}$, $w_{n\text{-}n'}$, or $w_{n'\text{-}Y}$) as well as bias weights ($w_{b\text{-}n}$, $w_{b\text{-}n'}$, or $w_{b\text{-}Y}$) where the first subscript represents the source node and the second subscript represents the destination node. Aside from temperature, *T*, and mole fraction of the IL, *X*, structure-related parameters such as number of carbon atoms in the cation, *NCC*, number of atoms in the anion, *NA*, number of hydrogen atoms in the anion, *NH* and number of carbon atoms in the alcohol, *NCA*, was also considered in the input layer [35,37]. A maximum number of two hidden layers and a maximum of 10 nodes per hidden layer were imposed and that there is only one expected output per neural network architecture, i.e., the refractive index.

2.3. Determination of the parameters of the model

In this study, the basis for determining the best neural network architecture is the one having the lowest mean absolute error, MAE as in Eq. (1) and corroborated with the percentage error as in Eq. (2). In Eqs. (1) and (2), L is the total number of data points, Y_l is the predicted value and E_l is the experimental or actual value. The calculation flow diagram for choosing the best neural network architecture is shown in Fig. 3.

$$MAE = \frac{1}{L}\sum_{l=1}^{L}|Y_l - E_l| \tag{1}$$

$$Percentage\ Error = \frac{|E_l - Y_l|}{E_l}\times 100\% \tag{2}$$

The mathematical relationship between inputs, hidden layers and outputs is described by weights, bias weights, and transfer functions as follows [30]:

$$n_j = f\left(\sum_{j=1}^{J} I' \times w_{l-n} + 1 \times w_{b-n}\right) \tag{3}$$

$$n'_k = f\left(\sum_{k=1}^{K'} n_j \times w_{n-n'} + 1 \times w_{b-n'}\right) \tag{4}$$