

## Density, viscosity and refractive index prediction of binary and ternary mixtures systems of ionic liquid

Gonzalo Astray Dopazo<sup>1,2,\*</sup>, Miriam González-Temes<sup>1</sup>, Dina Larios López<sup>2</sup> and Juan Carlos Mejuto<sup>1</sup>

<sup>1</sup>Department of Physical Chemistry, Faculty of Science, University of Vigo at Ourense, Spain

<sup>2</sup>Department of Geological Sciences, College of Arts and Sciences, Ohio University, Athens, United States of America

**Abstract:** Different artificial neural networks architectures were developed to predict the density, viscosity and refractive index of binary and ternary mixtures of ionic liquids using their individual properties. All neural network implemented were evaluated using the root mean square error (RMSE) and the average percentage deviations (APD) for the training and validation phase. The individual models implemented show great values of  $R^2$  (greater than  $9.995 \cdot 10^{-01}$ ) and low errors in terms of RMSE, that corresponds with an APD less than  $7.766 \cdot 10^{-02}$  %, with the exception of the model for the prediction of viscosity where the error raised to 3.338 %. The results show that the different individual artificial neural networks implemented are a useful tool to predict the density, viscosity and refractive index of binary and ternary mixtures of ionic liquids and with reasonably accuracy.

**Keywords:** Density, viscosity, refractive index, ionic liquid, binary mixtures, ternary mixtures, artificial neural network (ANN), artificial intelligence (IA).

### Introduction

#### Ionic liquid

Ionic liquids (ILs) are similar to molten salts with a wide melting temperature<sup>1,2</sup>. Ionic liquids are good solvents for polar, nonpolar, organic and inorganic compounds<sup>2</sup>. They are nearly non-volatile, non-flammable and, thermally/electrically stable<sup>3,4</sup>. Because of their interesting properties ionic liquids are gaining popularity as an alternative to traditional volatile organic solvents (VOCs)<sup>1,3,5</sup> and they are often called green solvents<sup>1</sup>. Therefore they are involved in many investigations in different areas such as chemistry and biotechnology<sup>3</sup>. Research on their chemical, physical and thermodynamic properties becomes necessary in order to select the most suitable ionic liquid for a particular application<sup>5,6</sup>.

In most cases, ionic liquids are composed of an organic cation and an inorganic, polyatomic inorganic anion<sup>7</sup>. Doing simple changes in the cation and anion combinations or by modifying the chemical structures of the constituent ions, it is possible to develop a huge number of new ionic liquid<sup>3,6,7</sup>, for this reason ionic liquids are called designer solvents<sup>5</sup>. This feature allows to synthesize new ionic liquids to provide a desired density, viscosity, melting point, etc., in order to suit the requirements of a particular process or specific applications<sup>8,9</sup>, we could say that is a simple process<sup>10</sup> but given the large number of potential

\*Corresponding author: Gonzalo Astray

E-mail address: [gastray@uvigo.es](mailto:gastray@uvigo.es)

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new ionic liquids that could be generated, their physicochemical characterization could be complicated because measurements and experiments are not always cheap and easy<sup>10</sup>, it is necessary therefore, the application of predictive models, as presented in this paper, to estimate the properties of different ionic liquids are important to minimize costs, reagents and labour.

### **Artificial neural network**

During the last three decades, Artificial Neural Networks (ANNs) have been widely applied<sup>11</sup>. The first studies about ANNs started in 1940, but their applications were limited until 1980<sup>12</sup>. ANNs has been used in poorly described complex systems, problems with noise or when the input is ambiguous<sup>13</sup>. In recent years researchers has shown that ANNs are good predictive models for engineering problems with a huge number of interaction variables<sup>14</sup> or to mainly nonlinearity problems<sup>15</sup>. There are advantages of this neuronal model compared with other methods based on empirical equation of state or atomistic approaches using molecular dynamics or Monte Carlo. One advantage is the simplicity of the method. The parameters needed to determine the density, viscosity and refractive index of mixtures of ionic liquid, are readily available from the supplier of the compounds of the mixture. Another advantage is that neural networks do not need to perform complex calculations using previous knowledge of molecular parameters, or the design of any type of empirical equations that explain the process.

ANNs are composed of interconnected units, artificial neurons, which are inspired in biological neurons. ANNs are a set of non-linear computational methods<sup>16</sup>, which attempt to simulate the processing ability of the human brain<sup>15,17</sup>. An artificial neural network is formed by a different number of neurons grouped into different interconnected layers<sup>17</sup>. The input data are introduced in artificial neural network through the first layer (input layer), these data are used as input variables to obtain the predicted value (output values) in the output layer, between the input layer and the output layer can be one or more intermediate layers (hidden layers)<sup>13,15,17</sup>. The ANN's structure, called topology or architecture, is given by the number of layers and the number of neurons into each layer, obviously the input neurons corresponds to the variables available, the output neurons depend on variables to predict, and intermediate neurons should be procured by a trial and error approach<sup>18</sup>.

In this paper we implemented a feed forward neural networks, where the information is transferred from the input layer to the first hidden layer up to the output layer, in other words, the input layer distributes the data information to the first intermediate layer, where propagation function processed the input data and the activation functions calculated the first intermediate layer output, which is transferred to output layer. Our neural network, a Multilayer Perceptron are trained with a back propagation algorithm, gradient descent algorithm, that transfers the estimation error back through the network until it reaches an acceptable error by modifying weight values through several iterations<sup>11,13-15,17</sup>.

The use of predictive models based on artificial intelligence are widely used in many fields such as; Food Chemistry to optimization of ultrahigh pressure extraction of green tea polyphenols<sup>19</sup>, Medicine for automatic electrocardiogram analysis<sup>20</sup>, Engineering to Active pulse structural control to control civil engineering structures under dynamic loading<sup>21</sup>, Mathematics<sup>22</sup>, Physics to predict maximum temperature cooling in single chips<sup>23</sup>, Environmental Sciences for monitoring and diagnosis of a combined heat and power plant<sup>24</sup>, Hydrology for flow prediction<sup>25</sup>, Food authenticity<sup>26</sup>, Aerobiology<sup>27</sup>, or in Chemistry to

analysis of chromatographic behavior of indinavir and its degradation products<sup>28</sup>, prediction of solid solubilities in supercritical carbon dioxide<sup>29</sup>, determining the rejection of neutral organic compounds by polyamide nanofiltration and reverse osmosis membranes<sup>30</sup>, predict of ethene + oct-1-ene copolymerization ideal conditions<sup>31</sup>, prediction density in ionic liquid<sup>32</sup>, conductivity<sup>33</sup>, viscosity<sup>34</sup> and to estimate the water content<sup>35</sup>.

The ultimate goal of this paper is to develop a predictive model to determine accurately the density, viscosity and refractive index of binary and ternary mixtures of ionic liquids using their individual properties, avoiding unnecessary waste of economic resources, reagents and labour. We must emphasize that the aim of this paper is to develop a simple model for the determination of the three mentioned properties: density, viscosity and refractive index, using data reported for commonly used ionic mixtures. We could try to develop a model where the ionic liquid could be characterized in its basic components and apply it to the prediction of the three studied variables. However, this would increase the complexity of the ANN model, and could decrease the predictive power of the ANN due to the increase in number of implicit variables.

## Material and methods

The first step to develop the different ANN models is to have experimental values (data set) of different ionic liquids to allow the ANN generalization from the training cases. Because the number of combinations of possible ionic mixtures is very large, the results of this work are limited to the modelled ionic liquids and their possible mixtures with organic components. In Table 1 we can see the compounds that can be part in the different mixtures collected.

**Table 1.** Composition of the different mixtures studied<sup>5,36-45</sup>. The meaning of abbreviation names is explained in the text.

Comp. 1	Comp. 2	Comp. 3	Ref.	Comp. 1	Comp. 2	Comp. 3	Ref.
Ethyl acetate	Ethanol	[C <sub>4</sub> mim][NTF <sub>2</sub> ]	5	Ethanol	Water	[Bmim][MeSO <sub>4</sub> ]	40
Ethyl acetate		[C <sub>4</sub> mim][NTF <sub>2</sub> ]	5	Water		[Bmim][MeSO <sub>4</sub> ]	40
Ethanol		[C <sub>4</sub> mim][NTF <sub>2</sub> ]	5	Ethanol		[Bmim][MeSO <sub>4</sub> ]	40
Ethyl acetate	Ethanol	[C <sub>8</sub> mim][NTF <sub>2</sub> ]	36	Ethanol	Water	[MMIM][MeSO <sub>4</sub> ]	41
Ethyl acetate		[C <sub>8</sub> mim][NTF <sub>2</sub> ]	36	Water		[MMIM][MeSO <sub>4</sub> ]	41
Ethanol		[C <sub>8</sub> mim][NTF <sub>2</sub> ]	36	Ethanol		[MMIM][MeSO <sub>4</sub> ]	41
Methyl acetate	Methanol	[C <sub>8</sub> mim][NTF <sub>2</sub> ]	37	Ethanol	Water	[C <sub>6</sub> mim][Cl]	42
Methyl acetate		[C <sub>8</sub> mim][NTF <sub>2</sub> ]	37	Ethanol		[C <sub>6</sub> mim][Cl]	42
Methanol		[C <sub>8</sub> mim][NTF <sub>2</sub> ]	37	Water		[C <sub>6</sub> mim][Cl]	42
ETBE	Ethanol	[emim][EtSO <sub>4</sub> ]	38	Ethanol	Water	EMISE	43
Ethanol		[emim][EtSO <sub>4</sub> ]	38	Ethanol	Water	[C <sub>8</sub> mim][Cl]	43
Isopropyl acetate	Isopropanol	[C <sub>8</sub> mim][NTF <sub>2</sub> ]	39	Ethanol	Water	[C <sub>4</sub> mim][Cl]	44
Isopropyl acetate		[C <sub>8</sub> mim][NTF <sub>2</sub> ]	39	1-Propanol	Water	EMISE	45
Isopropanol		[C <sub>8</sub> mim][NTF <sub>2</sub> ]	39	2-Propanol	Water	EMISE	45

## Data Set

We have compiled a database of 1053 experimental binary and ternary mixtures taken from the literature<sup>5,36-45</sup> and their properties were used to implement the different artificial neural networks. The experimental values of density, viscosity and refractive index were reported at 298.15 K and at atmospheric pressure<sup>5,36-45</sup>. The authors of different experiments report that all reagents used in the mentioned literature were of the highest purity and supplied by Fluka, Merck and Sigma-Aldrich. The ionic liquids used for the development of neural networks are as follows; i) 1-butyl-3-methylimidazolium methylsulphate ([Bmim][MeSO<sub>4</sub>]), ii) 1-butyl-3-methylimidazolium chloride ([C<sub>4</sub>mim][Cl]), iii) 1-hexyl-3-methylimidazolium chloride ([C<sub>6</sub>mim][Cl]), iv) 1-octyl-3-methylimidazolium chloride ([C<sub>8</sub>mim][Cl]), v) 1-ethyl-3-methylimidazolium ethylsulphate ([emim][EtSO<sub>4</sub>] or EMISE), vi) 1,3-dimethylimidazolium methylsulphate ([MMIM][MeSO<sub>4</sub>]), vii) 1-butyl-3-methyl-imidazolium bis(trifluoromethylsulfonyl)imide ([C<sub>4</sub>mim][NTF<sub>2</sub>]) and viii) 1-octyl-3-methyl-imidazolium bis(trifluoromethylsulfonyl)imide ([C<sub>8</sub>mim][NTF<sub>2</sub>]). The mole fraction of the compounds in the mixture varied for the different experiments and resulted in different binary and ternary mixtures of one ionic liquid.

All data set have been divided into two groups, the first group for training the different artificial neural networks (75% data cases), and the second group (25% data cases) to validate the neural networks and the good power accuracy (Table 2). For viscosity was necessary to eliminate some cases with not available output data, likewise, for the training and validation phase, viscosity cases greater than 100 mPa·s was eliminated.

**Table 2.** Summary of data reported in the literature and used for training and validation phases of the different neural networks implemented<sup>5,36-45</sup>.

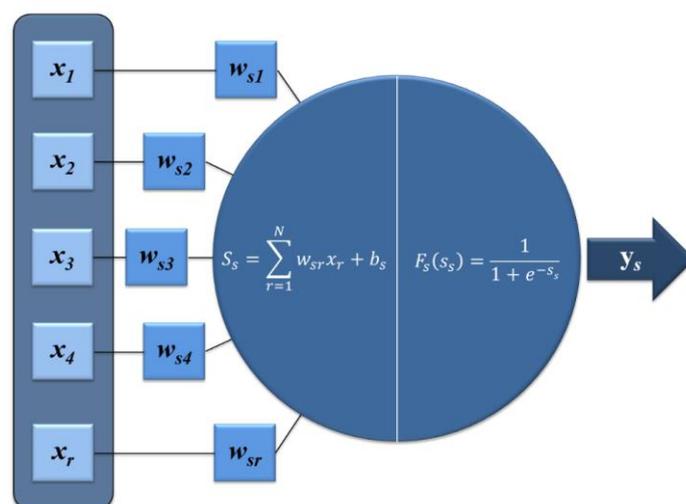
	Training	Validation
Density	789	263
Refractive Index	789	263
Viscosity	684	227

## Mixture chemical descriptor variables

Descriptor variables are used to differentiate the substances involved and their mixtures. The different mixture chemical descriptor variables used as input variables were taken from literature<sup>5,36-47</sup>. The following descriptor variables for the input were selected: i) Mole fraction ( $x_1$ ), density of pure compound ( $\rho_1$ ) and Molecular weight ( $M_{w1}$ ) for the first component of the mixture, ii) Mole fraction ( $x_2$ ), density of pure compound ( $\rho_2$ ) and Molecular weight ( $M_{w2}$ ) for the second component of the mixture, and iii) Mole fraction ( $x_3$ ), density of pure compound ( $\rho_3$ ) and Molecular weight ( $M_{w3}$ ) for the ionic liquid. A total of 9 input variables were used for all ANN implemented including individual neural networks and neural network with three outputs. The ionic liquid is always identified with the third component in the ANN. The other two components of the ternary solutions are identified with the components 1 and 2. In the case of a binary solution component 2 stays empty in the ANN matrix.

### Artificial neural units

The basic unit of all ANN is the artificial neuron (Figure 1). The artificial neuron has inputs, denoted as  $x_r$ , each input has a weight ( $w_{sr}$ ) that corresponds to the specificity of each pair of related neurons, and finally, each neuron has an output, denoted as  $y_s$ .



**Figure 1.** Structure and rules for the neural network in the first intermediate layer.

### Artificial neural network rules

We used a multilayer neural network, as the most commonly used in other studies of our research group<sup>48,49</sup>. In this case we use a neural network with an input layer with nine neurons to collect the training data. This type of neural networks is governed by two different functions; i) the propagation function, and ii) the activation function. The propagation function (Equation 1) whose mission is to convey the information between the different neurons spreads on different layers of the ANN (Figure 1).

$$S_s = \sum_{r=1}^N w_{sr} x_r + b_s \quad (1)$$

In this equation  $N$  corresponds with the number of neurons in input layer,  $w_{sr}$  corresponds with the weight (importance that characterizes the union of two neurons) between the neuron  $r$  (neuron in the input layer) and the neuron  $s$  (neuron in intermediate layer), finally,  $b_s$  corresponds the value of the neuron “bias” associated to each the intermediate neuron  $s$ . The value obtained is used by the activation function (Equation 2), a sigmoidal function, to provide an output value for each input entered into the ANN system.

$$F_s(S_s) = \frac{1}{1 + e^{-S_s}} \quad (2)$$

The neural network training continues until it reaches the minimum error previously set by the system operator. This error is used to modify the weights between neurons using for this purpose the back propagation rule. This error is calculated according to equation 3, where  $M$  is the number of neurons in the output layer,  $d_t$  represents the experimental value and  $y_t$  represents the output value predicted by the ANN.

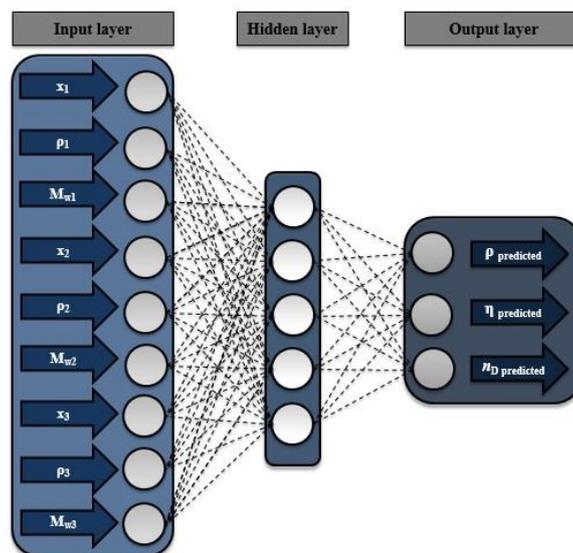
$$Error = \frac{1}{2} \sum_{t=1}^M (d_t - y_t)^2 \quad (3)$$

### Denotation of neural network implemented

The notation used for the identification of the neural network is similar to the notation used in other our papers<sup>49,50,51</sup>, and it is as follows;

$$N_{input} - N_{intermediate} - N_{output}$$

Where  $N_{input}$  and  $N_{output}$  represent the neurons number present in the input and output layer, respectively.  $N_{intermediate}$  represents neurons number in the intermediate layer.



**Figure 2.** Artificial neural network (9-5-3) with an input layer with nine neurons, one Hidden layer with five neurons and one input layer with three neurons to determinate the values of density, viscosity and refractive index in binary and ternary mixtures

Figure 2 shows an ANN example of this nomenclature with 9 neurons in the input layer, corresponds with the input variables (mixture chemical descriptor variables), 5 neurons in the intermediate layers, and finally, 3 neurons in the output layer, and corresponds with the values of density, viscosity and refractive index that we want to predict.

### Software package

Specialized software is used to develop all the neural networks, in this case, we have used the commercial packages provided by Neural Planner Software and Alyuda Research, LLC that allow a variety of activation functions very used in literature such as; sigmoidal function<sup>52</sup>, linear function<sup>53</sup> or hyperbolic tangent<sup>54</sup>. Likewise, it is also possible to use different of training algorithms, also used extensively in literature, such as Back Propagation<sup>55</sup>, Levenberg-Marquadt<sup>56</sup>, quasi-Newton algorithm<sup>57</sup>, etc.

## Results and Discussion

### Individual artificial neural networks for prediction properties

All artificial neural networks were implemented with ten input variables that correspond to the variables necessary to identify the mixture components (three variables for each) and the temperature at which the experiment was performed. Once, the input variables (mixture chemical descriptor variables) in the system are known, it is necessary to determine the number of neurons distributed in the intermediate layers. The following equation

(Equation 4) is used to determine the number of neurons in the intermediate layers. Where  $N$  is the number of mixture chemical descriptor variables used as input variables, and  $n$  is the number of neurons that is necessary to spread in the intermediate layers (may be in one layer, two or even three intermediate layers).

$$1 < n < N+1 \quad (4)$$

The following step is to implement the ANN and proceed to obtain the parameters that indicate the quality of the adjustment, such as: the linear squared correlation coefficient ( $R^2$ ), Root Mean Square Error (RMSE, Equation 5) and the average percentage deviation (APD, Equation 6), where  $d_t$  represents the experimental value,  $y_t$  represents the output value predicted by the ANN and  $M$  represents the cases number. All these parameters, RMSE and APD, are calculated for the training phase, denoted by subscript T, validation phase, denoted by subscript V, and for both phases, average, denoted by subscript AV.

$$RMSE = \sqrt{\frac{\sum_{n=1}^M (d_t - y_t)^2}{M}} \quad (5)$$

$$APD = \frac{\left( \sum_{n=1}^M \left( \frac{y_t - d_t}{d_t} \right) \right) \times 100}{M} \quad (6)$$

With these parameters, the goodness of the power prediction of different ANN models is estimated, in this regard Root Mean Square Error in validation phase ( $RMSE_v$ ) has been used. Table 3 shows the best individual artificial neural networks implemented to predict the values of density, viscosity and refractive index. Considering data shown in Table 3, we can say that the best neural networks for each predicted variable present a good fit ( $R^2_T > 9.996 \cdot 10^{-01}$ ) for training phase. Regarding the error in each of neural networks we can say that networks implemented to predict the density and the refractive index have a lower RMSE ( $RMSE_T < 9.941 \cdot 10^{-04}$ ) and the implemented neural network to predict the viscosity has a RMSE of  $1.703 \cdot 10^{-01}$  mPa·s, corresponds to a 3.124%.

**Table 3.** Best neural network implemented for density, refractive index (Ref. ind.) and viscosity.  $R^2$  is the square correlation coefficient for training ( $R^2_T$ ), validation ( $R^2_v$ ) and global phases ( $R^2_{av}$ ), Root mean square error of training ( $RMSE_T$ ), testing ( $RMSE_v$ ) and global phases ( $RMSE_{av}$ ) and Average percentage deviation for training ( $APD_T$ ), validation ( $APD_v$ ) and global phases ( $APD_{av}$ ) for different artificial neural network models developed. Finally, topology and training cycles for the best neural networks implemented for each property.

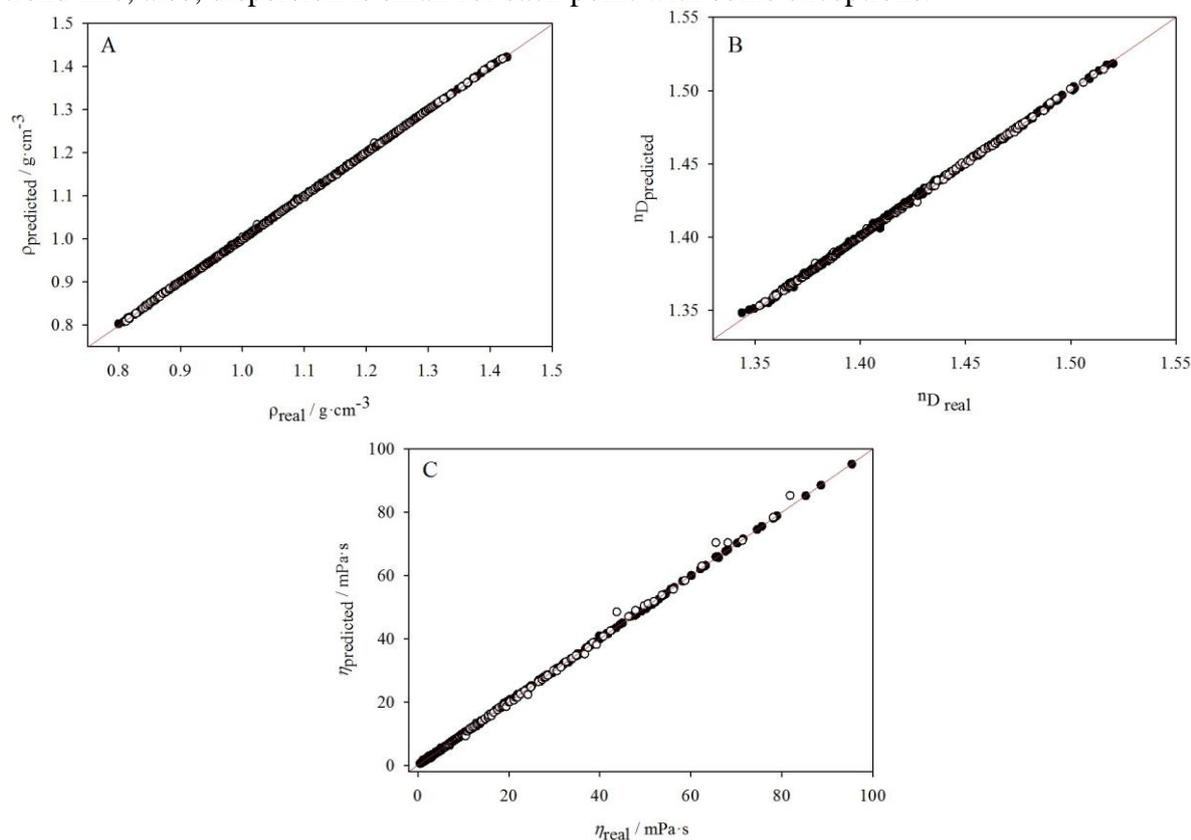
ANN	$R^2_T$	$RMSE_T$	$APD_T$	$R^2_v$	$RMSE_v$	$APD_v$	$R^2_{av}$	$RMSE_{av}$	$APD_{av}$
Density	$9.999 \cdot 10^{-01}$	$9.941 \cdot 10^{-04}$	$7.275 \cdot 10^{-02}$	$9.999 \cdot 10^{-01}$	$1.489 \cdot 10^{-03}$	$9.241 \cdot 10^{-02}$	$9.999 \cdot 10^{-01}$	$1.138 \cdot 10^{-03}$	$7.766 \cdot 10^{-02}$
Ref. Ind.	$9.996 \cdot 10^{-01}$	$9.668 \cdot 10^{-04}$	$5.673 \cdot 10^{-02}$	$9.995 \cdot 10^{-01}$	$9.578 \cdot 10^{-04}$	$5.466 \cdot 10^{-02}$	$9.995 \cdot 10^{-01}$	$9.646 \cdot 10^{-04}$	$5.621 \cdot 10^{-02}$
Viscosity	$9.999 \cdot 10^{-01}$	$1.703 \cdot 10^{-01}$	3.124	$9.988 \cdot 10^{-01}$	$6.088 \cdot 10^{-01}$	3.982	$9.996 \cdot 10^{-01}$	$3.378 \cdot 10^{-01}$	3.338

ANN	Topology	Training Cycles
Density	9-18-1	1,600,00
Refractive index	9-18-1	75,000
Viscosity	9-18-1	600,000

After verification of the correct prediction power of the neural networks to the training phase, we proceeded to calculate the fits for the validation phase using the previously reserved validation data (Table 2). As we can see in Table 3, the best neural network implemented for predict density provides a good prediction power ( $R^2 > 9.999 \cdot 10^{-01}$ ) with a low root mean square error ( $RMSE_V < 1.489 \cdot 10^{-03} \text{ g} \cdot \text{cm}^{-3}$ ) for the validation set. In the case of refractive index neural network the correlation coefficient is  $9.995 \cdot 10^{-01}$  with a low  $RMSE_V$  of  $9.578 \cdot 10^{-04}$ , both neural networks present a low average percentage deviation,  $9.241 \cdot 10^{-02}$  and  $5.466 \cdot 10^{-02}$  respectively. As in the previous case, the neural network to predict the viscosity presents worst correlation coefficient ( $9.988 \cdot 10^{-01}$ ), root mean square error ( $6.088 \cdot 10^{-01} \text{ mPa} \cdot \text{s}$ ) and average percentage deviation (3.982) neural network. All these neural networks were trained with different learning rates, to control the weights variation for a good descent direction<sup>58</sup> and momentum to speed up convergence and maintain generalization power<sup>59</sup>. The maximum number of training cycles was set at the time that the error in the validation phase begins to rise (Table 3).

Figures 3 show the good fits obtained for the training and validation phase by different ANNs implemented, which are very close to a linear trend with slope 1 and 0 intercept one trend line, also, dispersion is small for each point with some exceptions.



**Figure 3.** Plot of experimental values versus predicted values for training values (●) and testing values (○) by ANN to predict density (A), refractive index (B) and viscosity (C). Red line represents the one trend line.

Our neural networks shows good results but we think that it is possible predicted the three properties at the same time with an only one neural network. In this case, all the data set have been divided into two new groups, the first group for training phase and the second group for validate the neural network model implemented in the before phase (Table 4). As in the

previous case, we used the cases with known output and we excluded the cases with viscosity values exceeding 100 mPa·s.

**Table 4.** Data used for training and validation phases reported in the literature consulted<sup>5,36-45</sup> for different neural networks implemented.

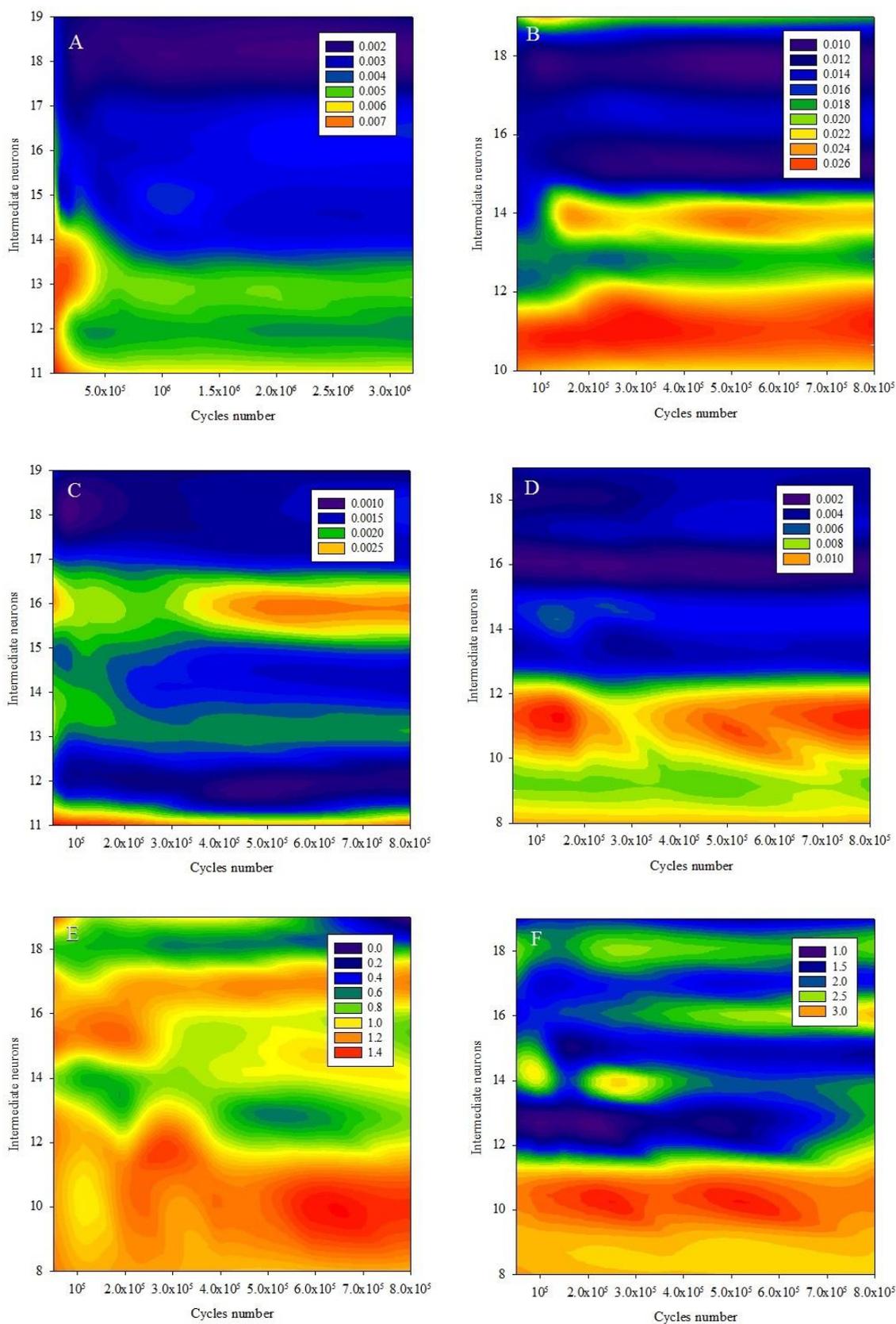
	Training	Validation
<b>Total cases used</b>	684	227

The best neural network implemented has a topology 9-13-3, that's, a single network, with nine input neurons, an intermediate layer with 13 neurons, and finally, and three neurons in the output layer. This ANN 9-13-3 was trained for 100,000 cycles. This neural network was chosen in function of lower RMSE in the validation phase for viscosity. The parameters that show the goodness of fit obtained for the new neural network implemented are shown in Table 5. These parameters can be compared with the same parameters for the individual ANN in Table 3. As we can see, the fits of the network with three outputs lose power prediction in training phase of density, refractive index and viscosity ( $9.941 \cdot 10^{-04} \text{ g}\cdot\text{cm}^{-3}$  vs.  $1.793 \cdot 10^{-02} \text{ g}\cdot\text{cm}^{-3}$ ,  $9.668 \cdot 10^{-04}$  vs.  $4.128 \cdot 10^{-03}$  and  $1.703 \cdot 10^{-01} \text{ mPa}\cdot\text{s}$  vs.  $1.095 \text{ mPa}\cdot\text{s}$  respectively, in terms of RMSE) in compared with individual neural networks. This behaviour also occurs in validation ( $1.489 \cdot 10^{-03} \text{ g}\cdot\text{cm}^{-3}$  vs.  $1.741 \cdot 10^{-2} \text{ g}\cdot\text{cm}^{-3}$ ,  $9.578 \cdot 10^{-04}$  vs.  $4.469 \cdot 10^{-3}$  and  $6.088 \cdot 10^{-01} \text{ mPa}\cdot\text{s}$  vs.  $1.139 \text{ mPa}\cdot\text{s}$  respectively, based on RMSE). Therefore demonstrated that the development of a network with three outputs is not feasible because worsens the predictive power to these three variables, so its use is strongly discouraged, especially in the case of viscosity with an average percentage deviation of 12.45%.

**Table 5.** Best neural network implemented for density, refractive index (Ref. ind.) and viscosity.  $R^2$  is the square correlation coefficient for training ( $R^2_T$ ), validation ( $R^2_V$ ) and global phases ( $R^2_{av}$ ), Root mean square error of training (RMSE<sub>T</sub>), testing (RMSE<sub>V</sub>) and global phases (RMSE<sub>av</sub>) and Average percentage deviation for training (APD<sub>T</sub>), validation (APD<sub>V</sub>) and global phases (APD<sub>av</sub>) for artificial neural network model developed with three outputs. Finally, topology and training cycles for the best neural networks implemented.

Property	$R^2_T$	RMSE <sub>T</sub>	APD <sub>T</sub>	$R^2_V$	RMSE <sub>V</sub>	APD <sub>V</sub>	$R^2_{av}$	RMSE <sub>av</sub>	APD <sub>av</sub>
Density	$9.928 \cdot 10^{-1}$	$1.793 \cdot 10^{-2}$	1.337	$9.912 \cdot 10^{-1}$	$1.741 \cdot 10^{-2}$	1.267	$9.923 \cdot 10^{-1}$	$1.780 \cdot 10^{-2}$	1.320
Ref. Ind.	$9.917 \cdot 10^{-1}$	$4.128 \cdot 10^{-3}$	$2.324 \cdot 10^{-1}$	$9.813 \cdot 10^{-1}$	$4.469 \cdot 10^{-3}$	$2.351 \cdot 10^{-1}$	$9.893 \cdot 10^{-1}$	$4.216 \cdot 10^{-3}$	$2.331 \cdot 10^{-1}$
Viscosity	$9.957 \cdot 10^{-1}$	1.095	$1.210 \cdot 10^{+1}$	$9.949 \cdot 10^{-1}$	1.139	$1.353 \cdot 10^{+1}$	$9.955 \cdot 10^{-1}$	1.106	$1.245 \cdot 10^{+1}$

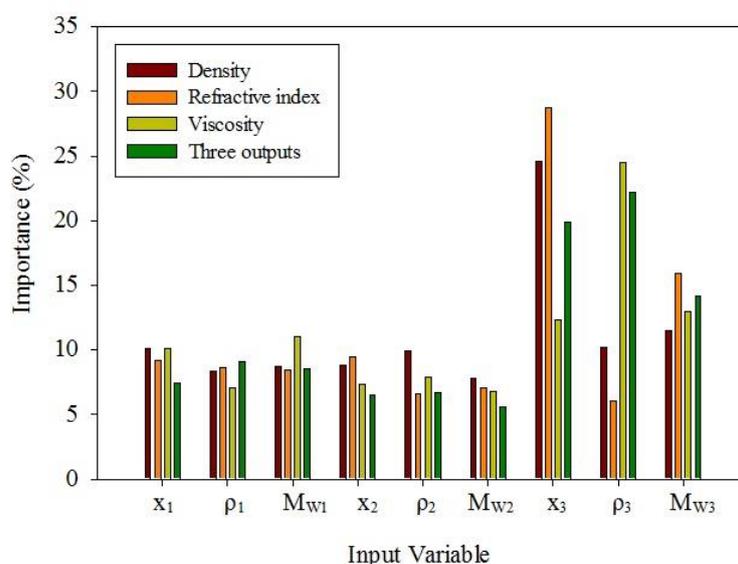
In Figures 4 we can see the errors distribution for all different neural networks as a function of training cycles and intermediate neurons used. It was necessary limit the area graphs to appreciate the difference between models. We can see that for a small number of neurons in the intermediate layers, the fits of different neural networks are worse than if the number of neurons is greater. It can be seen that each chosen neural network coincide with the lower RMSE zone, also we can see as the individual networks (located to the left) have minor errors in terms of RMSE than ANN with three outputs (located to the right), it also seeing, as the variation of errors across the bonnet test is more heterogeneous.



**Figure 4.** Variation of the error in the validation phase as a function of training cycles number for the best ANNs implemented for density (A and B), refractive index (C and D) and viscosity (E and F). Left (individual ANN), right (ANN with three outputs).

The relative importance of the different mixture descriptor variables was obtained from the sum of the weights of the neurons of the input layer that's corresponds to the strength of each neuron interconnection. In this case the importance of nine variables can be seen in Figure 5.

Taking into account the relative contribution of each input parameter, we can say that in general the most important mixture chemical descriptors are; the molar fraction in the mixture ( $x_3$ ) the density ( $\rho_3$ ) and the molecular weight ( $M_{w3}$ ) of the ionic liquid. The three most influential chemical descriptors are those that characterize the ionic liquid and they affect different aspects as the solutes diffusion or the solution behavior under stirring. Those input variables are strongly influenced by the structure of the ions forming the ionic liquid and they are good descriptor of the mixtures.



**Figure 5.** Importance of variables for each different networks implemented in this paper, that is; i) Mole fraction ( $x_1$ ), density of pure compound ( $\rho_1$ ) and Molecular weight ( $M_{w1}$ ) for the first component of the mixture, ii) Mole fraction ( $x_2$ ), density of pure compound ( $\rho_2$ ) and Molecular weight ( $M_{w2}$ ) for the second component of the mixture, iii) ii) Mole fraction ( $x_3$ ), density of pure compound ( $\rho_3$ ) and Molecular weight ( $M_{w3}$ ) for the ionic liquid

The error of the designed individual neural networks for viscosity prediction is due in part to the database (training and validation). In the selection of data sets, the viscosity values are distributed heterogeneously. In fact, if we analyse the results for intervals of viscosity values (Table 6), we can see, for the individual neural network, both in the training and validation phase, the interval of viscosity values with largest errors (in terms of percentage) is the interval (0-20], when we increase the range of the interval, the mistake is lower, except in the intervals [40-60] and [80-100] of validation phase where the error increases but this behaviour is not statistically representative. The largest errors in prediction of viscosity ANN compared with errors in the variables density and refractive index, suggests that the errors are due to the wide viscosity value range. Thus, the error in the prediction of the viscosity should be improved if new mixtures are included. This fact was contrasted in previous implementations, where training and validation phases had a smaller number of cases; in these neural networks the errors for density, refractive index and viscosity are bigger. The most significant case was the viscosity (583 training cases and 195 validation cases) where the average percentage deviation was 9.023, by contrast, the artificial neural network presented in this paper

(684 training cases and 227 validation cases) has an APD of 3.982. Other factors that could improve the errors are; the study of the behaviour of viscosity with temperature, expansion of additional neural networks for binary mixtures, ternary mixtures and mixtures lacking ionic liquid, or increase mixtures with two or more ionic liquids, all in order to get a universal tool for predicting these variables in any kind of mixtures.

**Table 6.** Fits for viscosity to individual ANN according to intervals of viscosity values (mPa·s).  $R^2$  is the square correlation coefficient for training ( $R^2_T$ ) and validation ( $R^2_V$ ), Root mean square error of training (RMSE<sub>T</sub>) and validation (RMSE<sub>V</sub>), the Average percentage deviation for training (APD<sub>T</sub>) and validation phase (APD<sub>V</sub>) and cases number for training ( $n_T$ ) and validation ( $n_V$ ) phases for the best artificial neural network model developed to predict the viscosity.

Training phase				Validation phase					
$R^2_T$	RMSE <sub>T</sub>	APD <sub>T</sub>	$n_T$	Intervals of viscosity value	$n_V$	$R^2_V$	RMSE <sub>V</sub>	APD <sub>V</sub>	
$9.991 \cdot 10^{-01}$	$1.436 \cdot 10^{-01}$	3.716	555	[0-20)	184	$9.983 \cdot 10^{-01}$	$1.938 \cdot 10^{-01}$	4.560	
$9.984 \cdot 10^{-01}$	$2.649 \cdot 10^{-01}$	$7.404 \cdot 10^{-01}$	77	[20-40)	26	$9.930 \cdot 10^{-01}$	$5.375 \cdot 10^{-01}$	1.062	
$9.975 \cdot 10^{-01}$	$2.838 \cdot 10^{-01}$	$4.689 \cdot 10^{-01}$	31	[40-60)	11	$9.384 \cdot 10^{-01}$	1.563	1.862	
$9.993 \cdot 10^{-01}$	$1.613 \cdot 10^{-01}$	$1.536 \cdot 10^{-01}$	18	[60-80)	5	$8.808 \cdot 10^{-01}$	2.462	2.521	
$9.999 \cdot 10^{-01}$	$8.899 \cdot 10^{-02}$	$5.788 \cdot 10^{-02}$	3	[80-100]	1	--	$1.253 \cdot 10^{+01}$	4.327	
$9.999 \cdot 10^{-01}$	$1.703 \cdot 10^{-01}$	3.124	684	(0-100]	227	$9.988 \cdot 10^{-01}$	$6.088 \cdot 10^{-01}$	3.982	

## Conclusions

In the present work, an artificial intelligent based approach was applied to investigate the possibility of the artificial neural networks model to predict the density, refractive index and viscosity of binary and ternary mixtures in the presence of ionic liquids.

Data have been compiled from cases reported in the published literature (1052 cases were used), and we have used the most studied variables to see the suitability of ANN models. In this sense, for future work it could be possible to study other properties, such as solubility or conductivity, if there is enough data reported in the literature.

The data cases were divided in two different groups, a training (75% data cases) group for developed different models and a validation group (25% data cases) to check the best model development. The mixture chemical descriptor variables employed are able to encode the complexity of binary and ternary system.

All individual neural networks developed, to predict the density, refractive index and viscosity of binary and ternary mixtures of ionic liquids, have good fits in terms of  $R^2$  (greater than  $9.995 \cdot 10^{-01}$ ). Likewise presents a low error in terms of RMSE, corresponding to an APD very low (less than  $7.766 \cdot 10^{-02}$  %) with the exception of the model to predict of the viscosity where the error rate rises to 3.338%.

We can recommend artificial neural networks as a suitable prediction method for density, viscosity and refractive index of binary and ternary mixtures of ionic liquids. It has been demonstrated that the models can predict chemical values and physical properties, however it is recommended continuous improvement of these models with new cases investigated in order to get a greater power of prediction.

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