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COMPARISON BETWEEN DIFFERENT ARTIFICIAL NEURAL NETWORK ARCHITECTURES IN VLE PREDICTION MODELS USING EXPERIMENTAL AND SIMULATED DATA (VIA MODIFIED RAOULT'S LAW) OF BINARY SYSTEMS

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Abstract. *In this present work, a comparison between two different Artificial Neural Networks (ANN) where made in VLE predictions. It was used three sets of VLE data containing the molar fraction of liquid phase of each substance and the pressure of the system. The objective is to determine the molar fractions of vapor phase and temperature. Three training types where compared:(a) using only simulated data with various pressure values (obtained via thermodynamic model); (b) using only monobaric experimental data; and (c using a hybrid prediction that utilizes an ANN trained with simulated data and an ANN trained with experimental data. Three binary systems where used to evaluate the quality of regression given by ANN predictions when compared with experimental data. The results have shown that it's possible to replace simulated predictions given by thermodynamic models with correctly trained ANNs. Also, the ANNs were capable to interpolate experimental data with reasonable accuracy.*

Keywords: *Artificial Neural Networks, Vapor Liquid Equilibrium, Modified Raoult's Law, Wilson Equation.*

1. INTRODUCTION

Separation of chemicals and molar fraction predictions of substances in Vapor Liquid Equilibrium (VLE) are topics of great interest in chemical engineering. There are several thermodynamic models that can predict the behavior of substances in VLE conditions, but they require obtaining parameters via VLE data analysis and regression over the measurements.

Many times, hence the parameters where obtained, these thermodynamic models require iterative methods to compute a solution; and this procedure could require some computational effort as well as some time to be evaluated with significant precision. As pointed by Ganguly (2003), this approach is not the ideal when fast prediction of a dynamic behavior is required.

In other hand, ANN presents mapping capabilities, and many authors has shown that they can be used to acquire knowledge about the VLE topology of multicomponent systems through a relatively small data set, and perform reliable predictions. The ANN can extract this knowledge directly from presented data and comprehends nonlinear relations between variables. Hence the ANN is correctly trained, their predictions are not obtained by iterative methods; therefore, its execution is quick, demanding less evaluating time when compared with iterative methods.

Ganguly (2003) used two schemes of radial basis function network to evaluate the molar fraction of liquid phase and pressure of four binary systems and two ternary system, and compared the results with predictions given by UNIQUAC thermodynamic model. Although UNIQUAC has shown to be more precise, the ANNs have shown good fitting to the experimental data.

Azari, *et al.* (2013), trained an artificial neural network with feedforward back propagation algorithm with experimental data to estimate VLE for eight binary refrigerant systems containing CO_2. The input data consisted of state variables (temperature, pressure, mole fraction of vapor and liquid phase) for each system and pure component properties (normal boiling point, critical temperature, critical pressure and acentric factor). The objective was to determine the molar fractions of vapor and liquid phases of CO_2. The prediction obtained by ANN where compared

with Madani Thermodynamic Model. The results have shown that Madani and ANN approaches present similar results, and both agreed reasonably with experimental data.

Nguyen *et. al.*(2007) used a back propagation network trained with experimental data to predict and estimate VLE of three ternary systems saturated with salt. It was used eight input variables: liquid mole fraction of solvents, critical temperature of solvent, critical pressure of solvent, acentric factor, cation radius and anion radius. These input variable should be capable to map four output variables: vapor molar fractions of solvents and bubble point temperature. The results were compared with Tan-Wilson and Tan-NRTL models. The ANN approach has shown better agreement with experimental data. In this work, it was explored the use of ANN in three approaches: (i) a set of six ANNs with two different architectures were trained with artificial data of three binary systems (benzene+cyclohexane, cyclohexane+n-heptane and hexane+heptane) generated with Raoult's modified Law, with Wilson equation for liquid phase (two ANN of each type for each system). This data was generated in a wide range of pressure, and a single VLE example consisted of six variables: liquid molar fraction of each substance, vapor molar fraction of each substance, temperature and pressure of the system. Using the thermodynamic model, pressure and liquid molar fractions were used in order to obtain vapor molar fractions and temperature of the system. Then the ANNs were trained with liquid molar fraction and pressure as input data (over the range of pressure stipulated), and should be able to predict vapor molar fractions and temperature; (ii) a set of monobaric experimental data containing liquid and vapor molar fractions and temperature was used to train another set of six ANNs. The ANNs should be able to predict vapor molar fractions and temperature of the system, given the liquid molar fractions (in this case, the pressure was not used as input parameter, hence all data was under the same pressure); and (iii) a hybrid model containing one ANN trained with simulated data and one ANN trained wit experimental data, for each binary system.

The experimental data was taken from literature, and can be found in DONG-SYAU JAN (1994) and Hargreaves (1944).

2. MODIFIED RAOULT'S LAW AND WILSON MODEL

There are many thermodynamic models that describe systems fluid phase behavior. Some of these models are known as Gibbs free energy models or activity coefficients models (Lima and Platt, 2010). The coexistence between liquid and vapor phase in binary systems can be described by the system in Eq. (1):

$$\begin{cases} y_i = x_i \gamma_i P_i^{sat} \\ \sum_{i=1}^{c} x_i = 1 \qquad i = 1, 2, \dots, c \\ \sum_{i=1}^{c} y_i = 1 \end{cases} \tag{1}$$

where x_i and y_i are the liquid and vapor phase of the *i*-th substance of the system, respectively; P is the system pressure; γ_i represents the activity coefficient of the *i*-th substance and P_i^{sat} represents the vapor pressure of the *i*-th pure component.

The Wilson Equation can describe the non-idealities of liquid phase. The expression of Gibbs Free Energy for a binary system is given by Eq. (2).

$$\frac{G^E}{RT} = -x_1 \ln(x_1 + x_2 \Lambda_{12}) - x_2 \ln(x_2 + x_1 \Lambda_{21}) \tag{2}$$

The expression corresponds to the following expressions, in order to obtain the activity coefficient of each component:

$$\begin{aligned} \ln(\gamma_1) &= -\ln(x_1 + x_2 \Lambda_{12}) + x_2 \left(\frac{\Lambda_{12}}{x_1 + x_2 \Lambda_{12}} - \frac{\Lambda_{21}}{x_2 + x_1 \Lambda_{21}} \right) \\ \ln(\gamma_2) &= -\ln(x_2 + x_1 \Lambda_{21}) - x_1 \left(\frac{\Lambda_{12}}{x_1 + x_2 \Lambda_{12}} - \frac{\Lambda_{21}}{x_2 + x_1 \Lambda_{21}} \right) \end{aligned} \tag{3}$$

where the coefficient Λ_{ij} is evaluated as described in Eq. (4)

$$\Lambda_{ij} = \frac{V_j}{V_i} \, exp\left(\frac{-A_{ij}}{RT} \right) \tag{4}$$

The A_{ij} is a binary interaction parameter and depends of the pairs of substances of the system.

3. ARTIFICIAL NEURAL NETWORKS AND ANN ARCHITECTURES

Artificial Neural Networks (ANNs) consists in sets of simple processing units (called neurons) attached by weighted connections. A network is formed by arranging the neuron in layers, where each neuron of a previous layer is connected

to each neuron of the next layer. Neurons are the fundamental process unit of an ANN which arrangement defines the network architecture. The capability of an ANN to understand the topology described by input data is widely used in prediction systems, where it's necessary to process the relationships (nonlinear in many cases) between the variables present in the particularly studied phenomena (Azari, *et al.*, 2013).

The ANN input layer (first layer) receives the information to be processed, and the number of neurons present in this layer depends on the variables involved in the prediction. Hidden layers connect the input and output layers and contributes to a more generalized learning about the relations between variables presented to the network and input/output correlation. The transfer function, that operate over the weighted inputs and bias of a neuron, describes the input/output relations, and describes the nonlinearity over the data. The output layer has the dimensionality of the target variables that the network should be able to map.

Different ANN architectures are different approaches in phenomena topology learning and mapping, and different architectures are motivated by the model of information and processing flow through the network. While in some types of ANN the information flows only from the input layer to output layer, other types includes more intricate connections, such as feedback paths. Neural network architectures are often evaluated by how well they separate the hyperspace for a given number of weights (Smith, 1997). In this work, it was used the software Matlab R2007B in all stages and computational routines.

Feed-Forward Artificial Neural Networks (FF-ANN) with back propagation algorithm are the most popular and widely used ANN models in many practical applications. In a FF-ANN, the information flows from the input layer to the output layer (forward): from the input nodes, the data goes through the hidden layers (if any) and reach the output layer, and no loops or feedback paths can be encountered. Each neuron evaluates a weighted sum of the inputs, usually adds a bias value in this stage, and this result is passed to an activation function. This process occurs through all neurons through the hidden layers. The activation function is usually a smooth step function. The sigmoid function is an example (Eq. 5).

$$sigmoid(x) = \frac{1}{1+e^{-x}} \tag{5}$$

Radial Basis Functions are a special class of functions. Their responses decrease (or increase) monotonically with the distance from a central point. The center, distance scale and the shape of the radial function are parameters that depend of the model (Orr, 1996). A typical radial function is the Gaussian function described in Eq. (6).

$$h(x) = exp\left(-\frac{(x-c)^2}{r^2}\right) \tag{6}$$

Radial Basis Function Artificial Neural Networks (RBF-ANN) could be, in theory, applied in sort of model (linear or nonlinear) or network (single layer or multilayer), but, traditionally, RBF-ANN have been associated with radial functions in single layer networks (Orr, 1996).

These two ANN architectures mentioned above were utilized in this work.

4. METHODOLOGY AND RESULTS

Three monobaric measurements of three binary systems found in literature were utilized as experimental training set for ANNs and also for validation. The experimental data consists of liquid and vapor mole fractions, temperature and pressure (fixed) of the system. The first analysis consisted to determine the degree of agreement between thermodynamic model and experimental data. In order to verify the agreement, it was calculated, for each prediction (two predictions of mole fraction for vapor phase and one for temperature), the mean absolute error (MAE), the standard deviation (SD) of the mean square error and the magnitude of both measures: mean absolute error magnitude (MAEM) and standard deviation magnitude (SDM), as described in Eq. (7), where p experimental measures were available, y_i^{exp} and y_i^{calc} are the experimental measure and calculated value (through thermodynamic model or ANN) of the i-th component (i = 1,2 denotes vapor mole fractions and i = 3 is temperature).

$$\begin{aligned}
E_i &= \left|y_i^{exp} - y_i^{calc}\right| \\
MAE_i &= \frac{\sum_{i=1}^{p} E_i}{p} \\
SD_i &= \sqrt{\frac{\sum_{i=1}^{p}(E_i - MAE_i)^2}{p}} \\
\text{MAEM} &= \sqrt{\sum_{i=1}^{3} MAE_i{}^2} \\
\text{SDM} &= \sqrt{\sum_{i=1}^{3} SD_i{}^2}
\end{aligned} \tag{7}$$

Figure 1 shows the behavior of the thermodynamic model compared to the experimental data for the benzene+cyclohexane, cyclohexane+n-heptane and hexane+heptane systems, respectively. Qualitatively, it's possible to observe that the model prediction is coherent with experimental data.

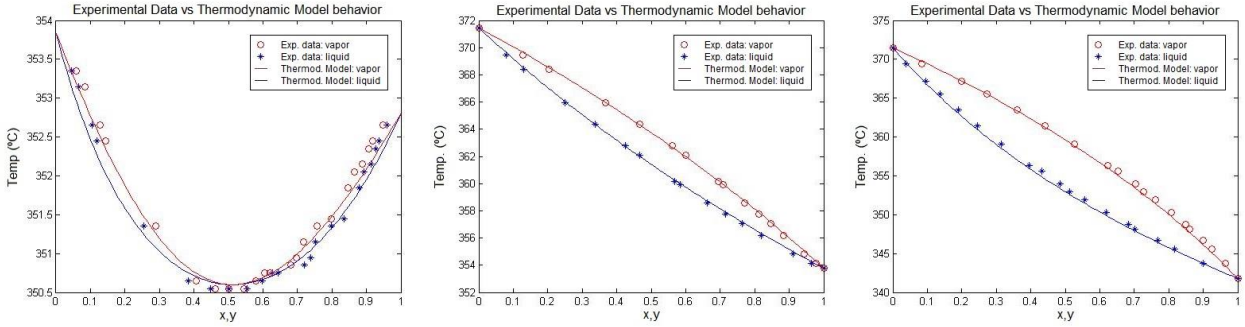

Figure1: (from left to right) thermodynamic model behavior over experimental data for benzene+cyclohexane, cyclohexane+n-heptane and hexane+heptane systems.

The first set of ANN was developed using simulated data. Using the thermodynamic model, it was generated VLE data between 50 kPa and 150 kPa (200 samples of pressure in the range) and a grid with 20 values of liquid molar fraction in the interval [0,1]. Therefore, the database created contains 4000 VLE artificial samples (for each pressure fixed, using 20 values of liquid molar fraction, the thermodynamic model can produce 20 vapor molar fractions and 20 temperatures related). 800 randomly chosen VLE examples of this database were employed to train ANNs with simulated data (ANN^{sim}) used in this work. FF-ANN was trained with the following parameters: 2 hidden layers with 12 neurons each, learning rate of 0.001, 70% of the database used for training, 20% for validation and 10% for testing; Levemberg-Marquadt (trainlm) was used as backpropagation algorithm. Transfer function was adjusted to *tansig* for hidden layers and *pureline* for output layers.

Table 1. Thermodynamic model and ANN^{sim}: performance against experimental data.

			MAE	SD	MAEM	SDM
TM	benzene+ cyclohexane	y_1	0.007019	0.003489	0.114351	0.070728
		y_2	0.007019	0.003489		
		$T^{(1)}$	0.113920	0.047754		
	cyclohexane+ n-heptane	y_1	0.001449	0.000810	0.110772	0.058201
		y_2	0.001449	0.000810		
		$T^{(1)}$	0.110753	0.058856		
	hexane+ heptane	y_1	0.006557	0.004628	0.337447	0.186521
		y_2	0.006557	0.004629		
		$T^{(1)}$	0.337320	0.177004		
FF-ANN^{sim}	benzene+ cyclohexane	y_1	0.006828	0.004466	0.113965	0.070479
		y_2	0.006828	0.004487		
		$T^{(1)}$	0.113553	0.070194		
	cyclohexane+ n-heptane	y_1	0.001392	0.000887	0. 111367	0.058613
		y_2	0.001392	0.000891		
		$T^{(1)}$	0.111349	0.058599		
	hexane+ heptane	y_1	0.006562	0.004570	0.337463	0.186505
		y_2	0.006562	0.004570		
		$T^{(1)}$	0.337335	0.186393		
RBF-ANN^{sim}	benzene+ cyclohexane	y_1	0.362417	0.262309	31.762307	0.923756
		y_2	0.362417	0.262309		
		$T^{(1)}$	31.758057	0.845998		
	cyclohexane+ n-heptane	y_1	0.318256	0.177973	4.249891	3.040528
		y_2	0.212417	0.168721		
		$T^{(1)}$	4.232631	3.030622		
	hexane+ heptane	y_1	3.517794	0.276536	16.420102	8.111441
		y_2	3.517794	0.276536		
		$T^{(1)}$	15.648323	8.102007		

$^{(1)}$T in Celsius.

RBF-ANN^{sim} was defined with a spread of 0.95, which was obtained by trial and error, although the results show no agreement with experimental data, and its predictions could not describes correctly the VLE behavior in a pressure that was not present in the training phase (other values was tested, without success). Further investigations could lead to a better choice of spread parameter. Table 1 shows that the thermodynamic model (TM) has a good agreement with experimental data. The same behavior can be observed for FF-ANN^{sim}. RBF-ANN^{sim} performed worse than FF-ANN^{sim}. It's observable that FF-ANN^{sim} can replace thermodynamic model, with the advantage of perform multibaric VLE prediction without the inconvenient of iterative methods or possibly convergence problems associated. Although database contains 4000 examples for training, by trial and error, it was found that 800 examples were enough to led FF-ANN^{sim} to a satisfactory prediction capability.

By observing the results in Tab. (1), it became clear that FFF-ANN^{sim} fits better to the simulated data set than RBF-ANN^{sim}. For all three binary systems, FFF-ANN^{sim} features lower values of MAE, SD, MAEM and SDM than RBF-ANN^{sim}, implying that FF networks are better choices for multibaric predictions when large simulated data sets are available for training.

The second test consisted of training ANNs with experimental data (ANN^{exp}). It was expected that they could overcome the problem of a small database (23 examples for benzene+cyclohexane system, 19 examples for Hexane+heptane system and 15 examples for Cyclohexane+n-heptane system) and correctly predict VLE. For benzene+cyclohexane system, 9 examples was used in training stage and 14 examples in validation stage, for both FF-ANN^{exp} and RBF-ANN^{exp}. For hexane+heptane system, 7 examples was used in training stage and 12 examples in validation stage, for both FF-ANN^{exp} and RBF-ANN^{exp}. Finally, for cyclohexane+n-heptane system, 7 examples was used in training stage and 8 examples in validation stage, for both FF-ANN^{exp} and RBF-ANN^{exp}.The parameters has shown to be difficult to determine, but both ANNs have overcome the limitation. FF-ANN was trained with the following parameters: 1 hidden layers with 32 neuron2, learning rate of 0.00015, 70% of the database used for training, 20% for validation and 10% for testing; Levemberg-Marquadt (*trainlm*) have led the ANNs to premature convergence of training stage; the function chosen as backpropagation algorithm was *trainbr*, which applies the Levemberg-Marquadt through a process called Bayesian regulation. Transfer function was adjusted to *tansig* for hidden layers and *pureline* for output layers. In this case, the parameter called *net.trainParam.mu_inc* was set to 1.004 (default is 10). Table 2 shows the statistic results of validation analysis of experimental data trained ANNs.

Table 2. FF-ANN^{exp} and RBF-ANN^{exp}: performance against experimental data not included in training stage.

			MAE	SD	MAEM	SDM
FF-ANN^{exp}	benzene+ cyclohexane	y_1	0.255160	0.139373	1.035426	0.855858
		y_2	0.255160	0.139373		
		$T^{(1)}$	0.970512	0.832852		
	cyclohexane+ n-heptane	y_1	0.211487	0.146718	3.950089	2.487363
		y_2	0.211487	0.146718		
		$T^{(1)}$	3.938750	2.478694		
	hexane+ heptane	y_1	0.227200	0.181396	6.880839	4.674528
		y_2	0.227200	0.181396		
		$T^{(1)}$	6.873333	4.667484		
RBF-ANN^{exp}	benzene+ cyclohexane	y_1	0.004591	0.004792	0.108117	0.070711
		y_2	0.004591	0.004792		
		$T^{(1)}$	0.107922	0.070385		
	cyclohexane+ n-heptane	y_1	0.001148	0.001091	0.074249	0.037143
		y_2	0.001148	0.001091		
		$T^{(1)}$	0.074231	0.037112		
	hexane+ heptane	y_1	0.001783	0.001981	0.114295	0.089779
		y_2	0.001783	0.001981		
		$T^{(1)}$	0.114267	0.089735		

$^{(1)}$T in Celsius.

By observing the results in Tab. (2), it becomes clear that RBF-ANN^{exp} fits better to the experimental data set than FF-ANN^{exp}(also, it has a better agreement than thermodynamic model). For all three binary systems, RBF-ANN^{exp} features lower values of MAE, SD, MAEM and SDM than FF-ANN^{exp}, implying that RBF networks are better choices for monobaric predictions when a small data set is available for training.

Figure 2 shows the VLE performed by both ANNs^{exp} (benzene+Cyclohexane system only). In all three systems, it was possible to observe good fitting performed by RBF-ANN^{exp} and a reasonable fitting performed by FF-ANN^{exp}.

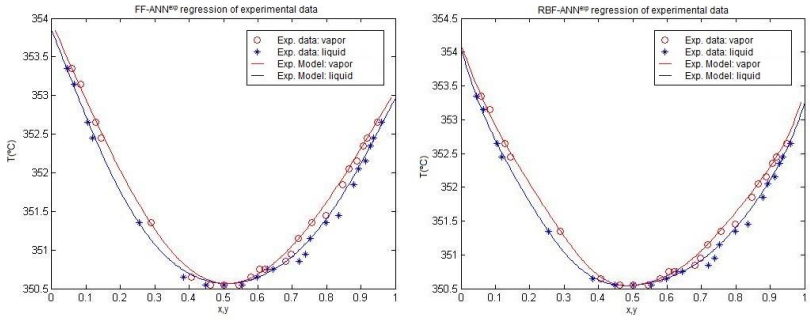

Figure 2. FF-ANN^{exp} and RBF-ANN^{exp} regression compared to experimental data (benzene+ Cyclohexane system).

However, an interesting result can be observed by utilizing the combined predictions of both FF-ANN^{sim} and RBF-ANN^{exp} in a unique output. Let VLE predictions be calculated in accordance with the following: by choosing a α in the interval (0,1), for an experimental input (x_1, x_2) under pressure P (fixed in the experimental measurements), evaluate VLE prediction using Eq. (8).

$$ANN^{hyb}(x_{1i}, x_{2i}, P) = \alpha.FF\text{-}ANN^{sim}(x_{1i}, x_{2i}, P) + (1-\alpha).RBF\text{-}ANN^{exp}(x_{1i}, x_{2i}, P) = (y_{1i}, y_{2i}, T_i)^{hyb} \quad (8)$$

This hybrid VLE prediction combines the best results obtained in simulated data regression and experimental data regression performed by ANNs. Furthermore, ANNs aren't iterative methods, thus avoiding iterative methods and convergence problems, which can be desirable when both reliable and fast predictions are needed. Table 3 shows that the hybrid model can even fit slightly better than ANN^{sim} and ANN^{exp} single predictions, when compared with Tab. (1) and Tab. (2). The values of α were chosen by trial and error, and no optimization routine was created to determine the best values for each systems. Further investigations could lead to determine better choices for this parameter. In this present work, the parameter α was set to 0.002, 0.21 and 0.056 for benzene+cyclohexane, cyclohexane+n-heptane and hexane+heptane binary systems, respectively.

Table 3. ANN^{hyb}: performance against experimental data not included in training stage.

			MAE	SD	MAEM	SDM
FF-ANN^{sim} + RBF-ANN^{exp}	benzene+ cyclohexane	y_1	0.004590	0.004791	0.108103	0.070696
		y_2	0.004590	0.004791		
		$T^{(1)}$	0.107907	0.070371		
	cyclohexane+ n-heptane	y_1	0.001115	0.000972	0.080177	0.036375
		y_2	0.001115	0.000977		
		$T^{(1)}$	0.080161	0.036349		
	hexane+ heptane	y_1	0.001665	0.001957	0.116691	0.074044
		y_2	0.001665	0.001957		
		$T^{(1)}$	0.116668	0.073992		

(1)T in Celsius.

Figure 3 shows that the hybrid model behavior is coherent with experimental data.

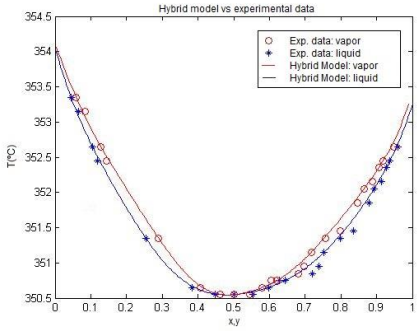

Figure 3. Hybrid model regression compared to experimental data (benzene+ Cyclohexane system).

5. CONCLUSIONS

Using ANN to replace usual thermodynamic models solvers has shown to be a reliable alternative in multibaric VLE predictions. The agreement of FF-ANN^{sim} with the experimental data was practically identical to Raoult's Modified Law outputs. In situations where fast responses of a prediction model are needed, the ANN model can be used as a trustable option that lacks the inconvenience of convergence problems in iterative methods.

Also, the experimental data was reasonable described by RBF-ANN^{exp} and FF-ANNexp, having RBF based network featured the most accurate results in this analysis.

The hybrid model used an ANNs trained with simulated data and an ANN trained with experimental data. The result observed is that was possible to create an output slightly more precise than outputs given by a single ANN. Although this behavior was not deeply verified, future studies can discuss the hypothesis that using both topologies (experimental and mathematical), it's possible to deliver more precise results in VLE predictions.

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