Prediction of Vapor-liquid Equilibrium Data by Using Radial Basis Neural Networks

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Most of the Chemical Engineering processes are nonlinear and complex in nature. They often require conventional modeling and simulation techniques based on certain simplified transport, kinetic and thermodynamic assumptions. These assumptions may, however, alter the exact nature of the system and would provide misleading information about the complex behavior of the system. An artificial neural network has the ability to overcome these limitations of the conventional approach by extracting the desired information directly from the data. In this paper radial basis network, a new generation of artificial neural network, has been successfully incorporated for the prediction of vapor liquid equilibrium data for binary systems including two azeotropes and a ternary system. Radial basis networks require lesser neurons than standard feed forward backpropagation and they can be trained in a fraction of time. From this work it is been proved that radial basis neural network has been successfully used for the prediction of vapor liquid equilibrium (VLE) data.

Key words:
Artificial neural network, vapor liquid equilibrium, radial basis network

Introduction

Artificial neural network is an established tool for effortless computation and its application in Chemical Engineering field is very promising. It has gained extensive interest. Artificial neural network has been successfully employed in solving problems in areas such as fault diagnosis,[1] process identification, property estimation, data smoothing and error filtering, product design and development, optimization, dynamic modeling and control of chemical processes,[2] design of mixing rule for the prediction of vapor-liquid equilibrium (VLE) data,[3] estimation of activity coefficients and prediction of VLE conditions.[4] The purpose of using artificial neural network in VLE data prediction is to reduce the number of experiments that are being carried out to characterize the system.

The well-known multi-layer feed forward backpropagation technique had been proposed earlier for the prediction of VLE data. The method of employing backpropagation technique requires an even spread of data for better VLE prediction and more training epochs. Radial basis networks require lesser neurons than the standard feed forward backpropagation networks and they can be trained in a fraction of time.[5] In this paper, radial basis network function has been successfully incorporated for the prediction of VLE data for binary and ternary systems. The proposed technique of using radial basis function requires only limited experimental data to predict the behavior of the system.

Artificial neural network in VLE data predictions

Vapor liquid equilibrium data are generally estimated using thermodynamic models based on the fundamental phase equilibrium criterion of equality of chemical potentials in both the phases. Most of these thermodynamic models are sets of equations, empirical and semi-empirical, with each having several constants determined using mixing rules for the mixtures.

The conventional methods used for the prediction of vapor liquid equilibrium (VLE) data are tedious and involve rigorous calculations in evaluating the mixture constants. The existing equations of state (EOS) approach apply well to hydrocarbon systems and are handicapped for systems containing polar compounds. The EOS are neither able to describe the critical region satisfactorily for mixtures nor estimate the liquid properties accurately. Activity coefficients are generally used for determining the liquid properties by several standard techniques. Each has its limitations in application to different kinds of systems. In brief, thermodynamics of mixtures are more complicated than for pure compounds and the difficulty in mixture analysis increases with the extent of non-ideality.

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VLE predictions are much more complicated for ternary systems and for the systems exhibiting azeotropic behavior. Artificial neural network, on the other hand, facilitate such predictions and eliminate the need for determining these constants by attempting to map the functional relationship all at once. Artificial neural networks overcome the limitations of the EOS approach in determining the data for highly polar systems including those of azeotropic systems. Also a limited database can be used to train a net to properly make it to learn the possible pattern of the pressure (p), temperature (T), liquid mole fraction (x), and vapor mole fraction (y) for a system.

Artificial neural network

Neural network, inspired by the information processing strategies of the human brain, are proving to be useful in a variety of engineering applications. Artificial Neural Networks (ANN) may be viewed as paralleled computing tools comprising of highly organized processing elements called neurons, which control the entire processing systems by developing association between objects in response to their environment.

ANN has been developed as generalizations of mathematical models of human cognition or neural biology. ANN is generally based on information processing that occurs at many simple elements called neurons and signals that are passed between neurons over connection links. Each connection link has its associated mass that gets multiplied with the signal transmitted and each neuron applies an activation function to determine its output signal.[6]

A neural network is characterized by its pattern of connections between the neurons – called its architecture, the method of determining the mass on the connections known as training or learning algorithm, and its activation function. The researches have proposed many architectures of the network. Two widely used network for non-linear problems are the backpropagation and radial basis function network.

Backpropagation network

In backpropagation, input vectors and the corresponding output vectors are used to train the network until it can approximate a function, associate input vectors with specific output vectors, or classify input vectors in an appropriate way as defined. Networks with biases, a sigmoid layer, and a linear output layer are capable of approximating any function with a finite number of discontinuities.

Limitations of backpropagation

A simple backpropagation is very slow because it requires small learning rates for stable learning. There are many techniques to improve the speed and general performance of backpropagation such as momentum and adaptive learning rates. Multi layered networks are capable of performing just about any linear or non-linear computation, and can approximate any reasonable function arbitrarily well. The error surface of a nonlinear network is more complex than the error surface of a linear network. Unlike linear networks, there is no easy way of picking a good learning rate for non-linear multi-layered networks.

The problem is that non-linear transfer function in multi-layered networks introduces many local minima in the error surface. As gradient descent is performed on the error surface, it is possible for the network solution to become trapped in one of these local minima. Settling in local minima may be good or bad depending on how close local minimum is to the global minimum and how low an error is required. Networks are also sensitive to the number of neurons in their hidden layers. Too few neurons can lead to underfitting. Too many neurons can contribute to overfitting, in which all training points are well fit, but the fitting curve takes wild oscillations between these points.

Radial basis function network

Radial basis function network form one of the essential categories of neural networks. A radial basis function (RBF) network is a two-layer network whose output units form a linear combination of the basis functions computed by the hidden units. A function is radially symmetric (or is an RBF) if its output depends on the distance of the input sample (vector) from another stored vector. Neural networks whose node functions are radially symmetric functions are referred to as radial basis function networks.[8]

RBF neuron model

The transfer function for a radial basis neuron is radbas. The radial basis neuron receives as net input, the vector distance between its weight vector $\boldsymbol{w}$ and the input vector $\boldsymbol{p}$, multiplied by the bias b. The basis functions in the hidden layer produce a localized response to the inputs so that each hidden unit has a localized receptive field. The basis function can be viewed as the activation function in the hidden layer. The outputs of the hidden unit lie between 0 and 1. The closer the input to center of the Gaussian, the larger the response of the node. The node produces an identical output for inputs with equal distance from the center of the Gaussian; it is called a radial basis. The output unit finds a linear combination of

the nonlinear basis functions and thus the network performs a nonlinear transformation of the input. The general model of RBF is as shown in Fig. 1.

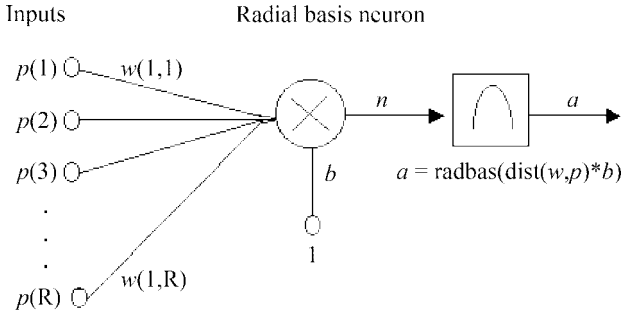


Fig. 1 – *Radial basis neuron model*

RBF network is capable of approximating any arbitrary mapping. The main difference between the RBF network and the backpropagation network is in their basis functions. The radial basis function covers only small regions, whereas the sigmoid function assumes nonzero values over an infinitely large region of the input space. Classification tasks are more amenable to the RBF network than the backpropagation network in the case when the problem is extended to higher dimensions.

RBF training procedure

The radial basis neural networks have been designed by using the function newrb available in the neural network toolbox supported by MATLAB.[9] The function newrb iteratively creates a radial basis network by including one neuron at a time. Neurons are added to the network until the sum of squared error is found to be very small or the maximum numbers of neurons are reached. At each iteration, the input vector, which will result in lowering the network error most, is used to create a radial basis neuron.

During the training, each of the connecting weights of the individual neuron is compared with input signals. The distance between the connecting weights determines the output of hidden neurons and input vector, which is further, multiplied by bias, an additional scalar quantity being added between neuron and fictitious neuron.

The output is propagated in a feed forward direction to output layer neuron, which will give output if the connection weights are close to input signal. This output is compared with target vector. If the error reaches the error goal, then training is terminated, otherwise the next neuron will be added. The connecting weights are modified each time by changing maximum neurons and the spread constant. The value of maximum neuron and spread constant keeps on changing till the network is trained properly. Radial basis networks can be used to approximate functions. newrb adds neurons to the hidden layer of a radial basis network until it meets the specified mean squared error goal.

Advantages of radial basis network

The time taken in designing a radial basis network is often less when compared to the training a sigmoid/linear networks.

The number of neurons required for designing the network is considerably less when compared to standard backpropagation network.

Radial basis neural network in VLE data predictions

Cyclohexane – trimethylpentane system

The RBF network was trained for the cyclohexane-trimethylpentane system[10] having 26 VLE data points at different isothermal conditions such as 308.15, 318.15, 338.15 and 348.15 K. The trained network was used to predict the VLE data at 328.15 K and the results are compared with the experimental values as shown in Fig. 2.

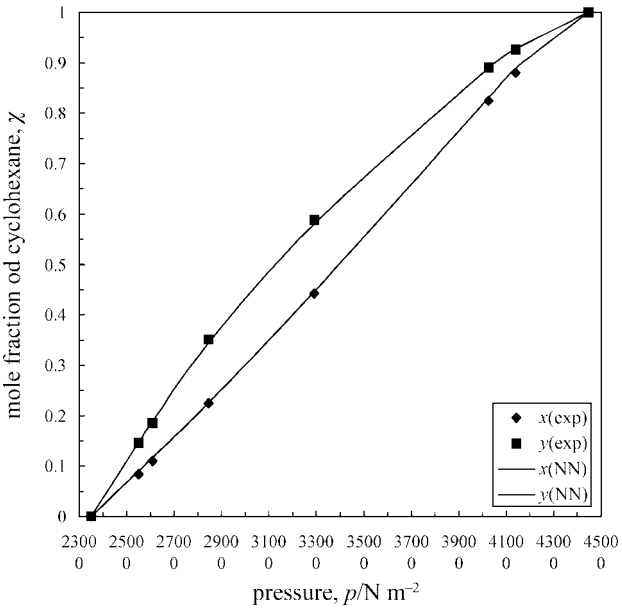


Fig. 2 – *Experimental and neural network VLE data for cyclohexane – trimethylpentane at 328.15 K*

Diethylamine – triethylamine system

The RBF network was trained for the diethylamine – triethylamine system[10] having 85 VLE data points at different isobaric conditions such as 46662.84, 53328.96, 59995.08, 66661.2, 79993.44, 86659.56, 93325.68, 106657.92 and 113 324.04 N m^{-2}. The trained network was used to predict the VLE data at

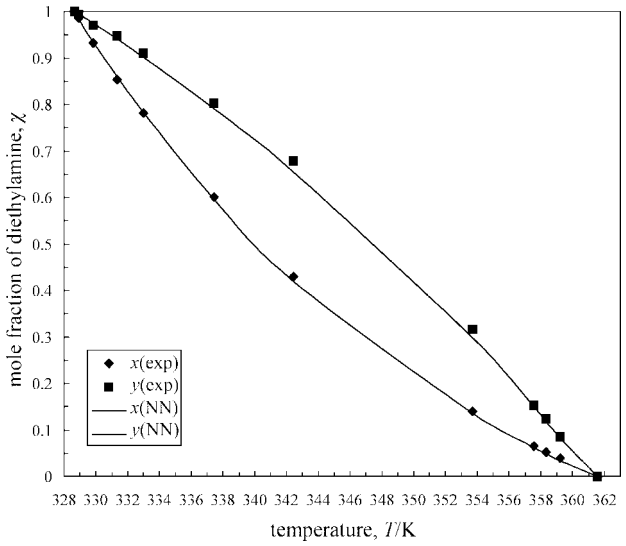


Fig. 3 – *Experimental and neural network VLE data for diethylamine – triethylamine at 101 325 N m^{-2}*

101 325 N m^{-2} and the results were compared with the experimental values and as shown in Fig. 3.

Ethanol – toluene

The RBF network was trained for the ethanol – toluene (minimum boiling azeotropic) system[10] having 100 VLE data points at various isothermal conditions such as 308.15, 318.15, 323.15, 328.15, 338.15, 343.15, 348.15, 353.15 and 358.15 K. The trained network was used to predict the VLE data at 333.15 K and the results are compared with the experimental values and are shown in Fig. 4.

Acetone – chloroform

The RBF network was trained for the acetone – chloroform (maximum boiling azeotropic) system[10] having 38 VLE data points at various isobaric conditions such as 97 592, 99 992 and 101 325 N m^{-2}. The trained network was used to predict the VLE data at 98 392 N m^{-2} and the results are compared with the experimental values and are shown in Fig. 5.

Hexane – benzene – butanol system

The RBF network was trained for the hexane-benzene-butanol ternary system[11] having 72 VLE data points at a pressure of 101 325 N m^{-2}. The trained network was used to predict those VLE data points that were excluded in the training set and the results have been compared with experimental values and are as shown in Fig. 6.

Results and discussion

The neural network based equation of state used in this paper to predict liquid phase composition (x) and vapor phase composition (y) at given conditions of temperature and pressure has been found to be satisfactory. In this paper, two binary systems, two azeotropic systems, and a ternary system were studied to predict the VLE data using Radial Basis Network. The data used in the study consisted of pressure and temperature as inputs to the network. The

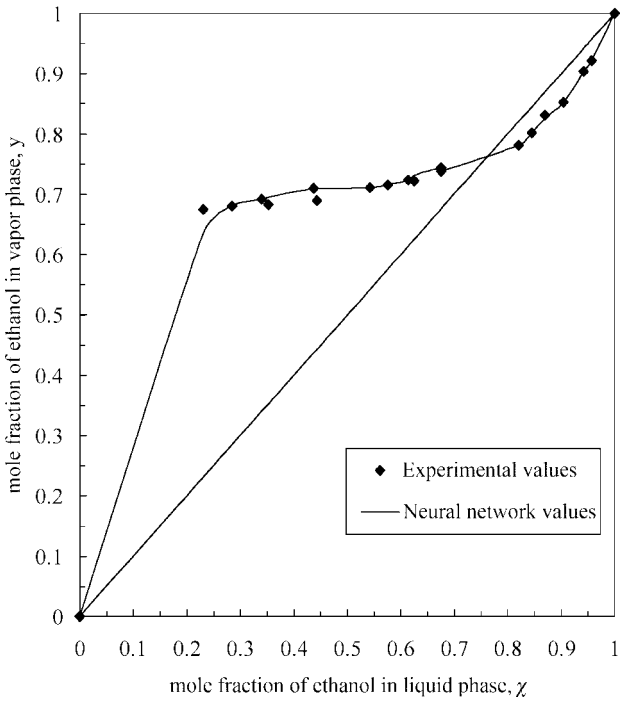


Fig. 4 – *Experimental and neural network values for ethanol – toluene at 313.15 K*

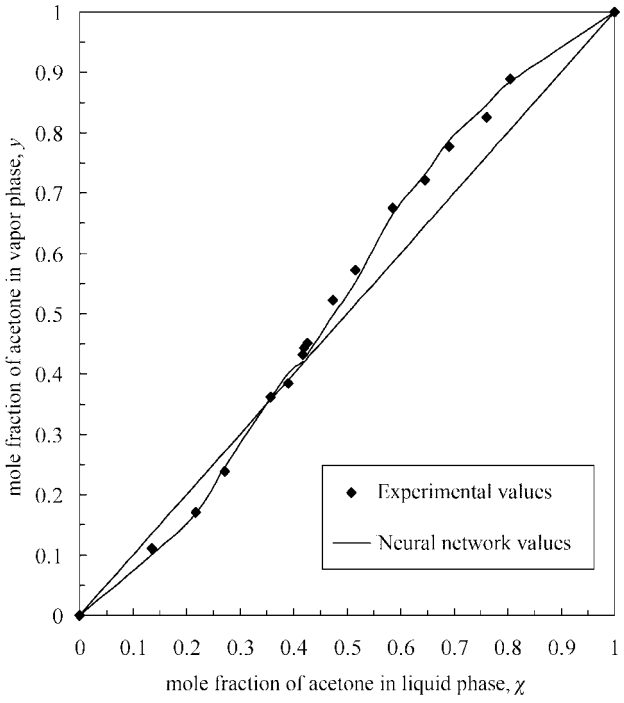
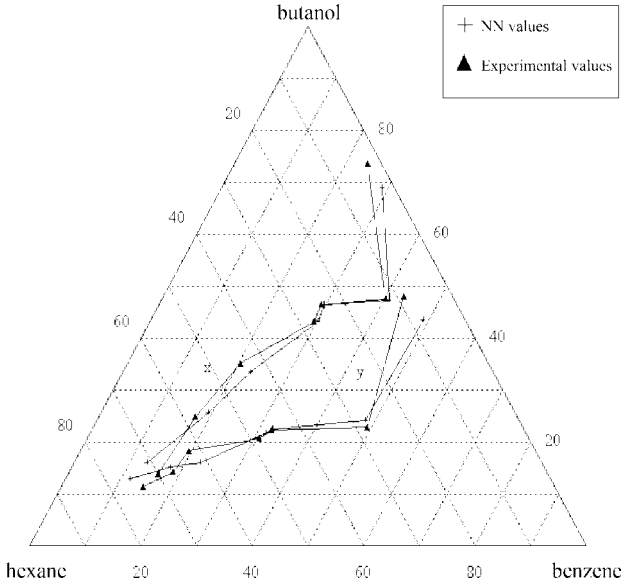


Fig. 5 – *Experimental and neural network values for acetone – chloroform at 98 392 N m^{-2}*



F i g . 6 – *Neural network predicted and experimental values of mole fractions % for hexane – benzene – butanol system at 101 325 N m^{-2}*

designed network is used to predict the liquid and vapor phase mole fraction. For the ternary system, the network was designed with pressure, temperature, and the liquid phase mole fraction as inputs and the vapor phase mole fraction as the output of the network. The performance of the network has been evaluated on the basis of an overall absolute error and root mean square error specified by the difference in the desired and actual outputs.

Mehmet Bilgin et al had employed the neural network to predict the vapor-liquid equilibrium (VLE) data for six different binary systems having different chemical structures and solution types (azeotrope-nonazeotrope) in various conditions (isothermal or isobaric). A feed-forward back-propagation neural network was employed in the study.[12] The present study overcomes the limitations back-propagation technique. The computational time required by radial basis function network is 0.2 s in a Pentium III processor based personal computer system, whereas the time required by back propagation network is about 3 s.

Absolute error

$$\mathrm{ABSD} = \frac{\sum |(\text{NN value} - \text{exp. value})|}{\text{number of data points}}$$

Root mean square error

$$\mathrm{RMSD} = \sqrt{\frac{\left(\sum |(\text{NN value} - \text{exp. value})|^2\right)}{\text{number of data points}}}$$

Conclusion

The radial basis network approach used in this work for vapor liquid equilibrium data prediction gives favorable results for the isothermal system of cyclohexane – trimethyl pentane and isobaric system of diethylamine – triethylamine. In this paper, the RBF network was successfully implemented for the VLE data prediction of minimum boiling azeotropic system of ethanol – toluene and for the maximum boiling azeotropic system of acetone – chloroform. A successful attempt has also been made to predict the VLE data for the *n*-hexane – benzene – sec-butanol ternary system. In this method fewer number of experimental data point were used to predict the VLE data for the systems exhibiting complex behavior. In this approach it was found that the network gives better results with an uneven spread of data point which is not the case with the backpropagation network which requires an even spread of data for faster convergence and less error. The RBF network can be used for better VLE predictions of isothermal, isobaric, azeotropic, and non-azeotropic binary systems, and also for ternary systems. Hence radial basis function network used to predict vapor liquid equilibrium data for thermodynamic systems is proved to be an efficient, reliable, and robust technique.

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