

APPLICATION OF ARTIFICIAL NEURAL NETWORKS IN PREDICTION OF VAPOUR LIQUID EQUILIBRIUM DATA

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ABSTRACT

The associative property of artificial neural networks (ANNs) and their inherent ability to “learn” and “recognize” highly non-linear and complex relationships finds them ideally suited to a wide range of applications in chemical engineering. The present paper deals with the potential applications of ANNs in thermodynamics – particularly, the prediction/estimation of vapour-liquid equilibrium (VLE) data. The prediction of VLE data by conventional thermodynamic methods is tedious and requires determination of “constants” which is arbitrary in many ways. Also, the use of conventional thermodynamics for predicting VLE data for highly polar substances introduces a large number of inaccuracies. The possibility of applying ANNs for VLE data prediction/estimation has been explored using the back propagation algorithm. Application of ANNs to the VLE predictions of $\text{NH}_3 - \text{H}_2\text{O}$ and $\text{CH}_4 - \text{C}_2\text{H}_6$ system is investigated. The results of neural equation of state (NEOS) are compared with popular thermodynamic approaches. The inputs to the net consist of Temperature and Pressure. Liquid and vapour phase compositions are obtained as outputs. These outputs are compared with the predictions obtained by using Peng Robinson equation of state and Wilson activity coefficients. For $\text{NH}_3 - \text{H}_2\text{O}$ system vapour phase compositions are well predicted by all three approaches but thermodynamic approaches are unable to predict liquid phase compositions. ANNs or NEOS gives good results. For $\text{CH}_4 - \text{C}_2\text{H}_6$ system, both Peng Robinson EOS and NEOS give good results. From the present work it is concluded that though for simple systems ANNs do not offer additional advantage, they are certainly superior when complex and polar systems are encountered. An heuristic approach to reduce the trial and error process for selecting the “optimum” net architecture is discussed.

INTRODUCTION

Man has been fascinated by the capabilities of human brain and has tried to make a computer mimic the way the human brain sorts through the information. ‘Neurocomputing’ is such an attempt, to understand and simulate its functioning. It has been touted as the first known alternative to the programming paradigm that has dominated computing for the past fifty years.

Artificial Neural Network (ANN) can thus be cited as ‘algorithmic equivalent’ of the human learning process and information processing scheme at a modest scale. They are pattern recognition architecture which can identify patterns between complex sets of input and output data. These patterns are then used to predict outcomes for fresh inputs. They do not require the specification of correlations which govern process, but are trained on real life data.

The chief advantage of ANNs lies in the fact that ANN uses a generic model which covers a wide class of problems. It does not require a fundamental understanding of the process or phenomena being studied and can handle complex and non linear models. Thus they are gaining a rapid interest within engineering, medical, financial and various other fields. They have thus made strong advances in area of continuous speech recognition, classification of noisy data, market forecasting, process modelling, fault detection and control.

In present work, application of neurocomputing for estimating VLE data has been explored. Conventional thermodynamic techniques for VLE data estimation of mixtures are tedious and have a certain amount of empiricism by way of determining mixture “constants” using arbitrary mixing rules. ANNs, on the other hand, help such predictions and eliminate the need for determining these constants by finding the functional relationship all at once. ANNs also offer the potential to overcome the limitations of existing equations-of-state (EOS) in determining VLE data for highly polar systems. The ammonia-water ($\text{NH}_3 - \text{H}_2\text{O}$) and the methane-ethane ($\text{CH}_4 - \text{C}_2\text{H}_6$) systems were studied and the results are presented. The methodology used and the advantages and the limitations of this approach have also been discussed.

Various types of Nets are formed by combination of different algorithms and activation functions. The Multilayer Perceptron (MLP) model is capable of mapping data that are non linear and complex. Thus, an MLP model using Back Propagation Algorithm (BPA) based on sigmoidal activation function has been found to be most suited for chemical engineering applications and is used in present work.

COMPARISON OF ANN WITH EQUATION OF STATE (EOS) FOR VLE DATA PREDICTION

Separation of a fluid mixture into pure components is an essential operation in chemical process industries. These mixtures are often separated by diffusional operations such as distillation, absorption and extraction. This requires a quantitative knowledge of the equilibrium composition of coexisting vapour and liquid phases at a given temperature and pressure.

Such data are generally not available and in some cases only partial data are present. Thus it is necessary to reduce and correlate the limited data to the best possible interpolations and extrapolation. This is the incentive for the application of various correlations to the calculation of phase equilibrium relationships.

The EOS fairly predicts the VLE data of hydrocarbon systems but is quite handicapped for systems containing polar compounds. Further, the EOSs are neither able to describe the critical region satisfactorily for mixtures nor estimate the liquid properties accurately. Activity coefficients are generally used for determining liquid properties and several estimation techniques exist in the literature (Smith and van Ness, 1995; Fredenslund et al., 1977). However, each has its limitations in its applicability to different systems; for instance, even though the UNIFAC method (Fredenslund et al., 1977) is applicable even to systems containing water, it has its limitations with regard to hydrocarbon mixtures. So, thermodynamics of mixtures are more complicated than for pure compounds and the difficulty in mixture analysis increases with the extent of non-ideality.

ANNs being purely "numeric" in nature do not require thermodynamic modelling; and hence, are convenient for VLE data prediction. A limited database can be used to train a net properly for it to learn the possible pattern of the pressure (P) – temperature (T) – liquid mole fraction (x) – vapour mole fraction (y) surface for a system in a small range of P and T.

CONCEPT OF NEOS FOR A BINARY SYSTEM

According to Gibbs phase rule, two intensive properties are required to completely describe a binary two phase system at equilibrium. Thus two intensive properties can be selected to describe all the properties

of the system irrespective of the methodology used. Pressure and Temperature are two convenient intensive properties, since they can be easily measured and controlled. Therefore, the net developed NEOS uses P and T as the inputs and gives x and y as the outputs.

Two widely different systems, the ammonia-water and the methane-ethane systems, were selected to explore the use of ANNs as an alternative to VLE data prediction. EOS fails to give accurate prediction for the NH₃-H₂O system due to presence of H₂O which is a polar compound. CH₄-C₂H₆ being a simple system can be described using relevant EOS. These two systems represent the "extremes" of thermodynamic behaviour to highlight the application of ANN to represent a wider range of systems than an EOS. The predictions of the ANN for the above two systems compare well with literature values, thereby demonstrating that the NEOS indeed has the potential to be at par with, if not better than, the existing EOSs.

METHODOLOGY FOR DEVELOPING NEOS

NH₃ – H₂O System

ANN was trained for the NH₃ – H₂O system using values taken from the literature (Macriss et al., 1964). The training data used for each approach consisted of:

- Data for only a small range of pressure (at 20-50 psia),
- Data points at extreme pressures only (at 25, 50, 275 and 300 psia),
- All the available data points.

All the above data was within the temperature range of 0-417.43°F. The optimum net architecture – the 2-13-2 net – for which the "best" results were obtained was arrived at by an heuristic approach discussed below.

CH₄ – C₂H₆ System

Only limited VLE data is available for the CH₄ – C₂H₆ system so the net was trained for all the points (a total of 75 points in the pressure and temperature range of 28-719 psia and 234.7-359.9°F, respectively). The optimum net architecture was again determined to be 2-13-2 by the heuristic approach described below.

THE HEURISTICS

A number of techniques have been given for network architecture selection (Kung and Hwang), however it still remains an iterative trial and error procedure. The following heuristic approach was adopted to overcome the trial and error process. One hidden layer with an appropriate number of hidden units is capable of mapping any input presentation (Sartori and Panos, 1991); hence, the study is restricted to one layer only (training with two layers was also attempted without any gain in accuracy). For the purpose of determining the optimum number of hidden units, the learning rate

and the momentum term were assigned arbitrary but constant values the gain term was fixed at a value of unity.

With all the parameters thus fixed, various net topologies exhibited the same trends relative to each other vis-à-vis the overall absolute error as a function of the number of iterations in the training mode (Dwivedi, 1992). Thus, it was found possible to determine the optimum net architecture within 50-100 iterations without traversing the entire graph of the absolute error as a function of the number of iterations for each topology. The number of iterations was used as the criterion for determining the overall absolute error rather than the time-steps. The time-steps are generally used as the criterion whenever on-line predictions are to be made such as for chemical process control where computation time is of importance.

Once the net parameters and the net architecture were fixed, the minimum number of training data sets required for adequate mapping was determined by a trial and error procedure. The net was then ready to learn the data presented using the BPA.

RESULTS AND DISCUSSION

Software was developed to predict the VLE data using the Peng Robinson EOS. This software can be used for any binary system and requires the critical properties and Antoine's coefficients of pure components. It takes ideal compositions as first guesses and then iterates till convergence. The performance of this software was evaluated by predicting the equilibrium data for $\text{CH}_4 - \text{C}_2\text{H}_6$ system. This is a simple system which obeys the Peng Robinson EOS.

NEOS approach to predict phase compositions (x and y) at a given P and T conditions was found to be satisfactory. As explained earlier, the $\text{NH}_3 - \text{H}_2\text{O}$ and the $\text{CH}_4 - \text{C}_2\text{H}_6$ systems were studied for this application. The data used in the present work consisted of pressure (psia) and temperature ($^{\circ}\text{F}$) as inputs to the net. Liquid and vapour weight fractions (x and y respectively) were predicted by the net. Several data sets were used for training and the net architecture of 2-13-2 was found suitable for all the cases presented. The performance of the net was evaluated on the basis of an overall absolute error specified by the difference in the desired and actual outputs as defined below:

$$\text{Overall absolute error} = \sum (\text{actual output} - \text{desired output})/n,$$

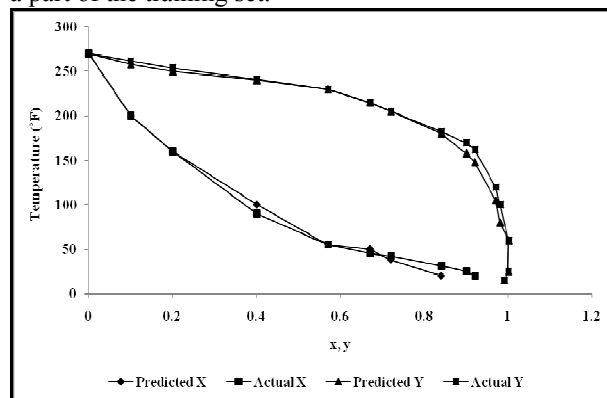
Where, n equals the number of data sets.

$\text{NH}_3 - \text{H}_2\text{O}$ System

Training of Net

One of the approaches to train the net was to use only a small range of training data; that is data at pressures of 15, 25, 30 and 50 psia, respectively and the

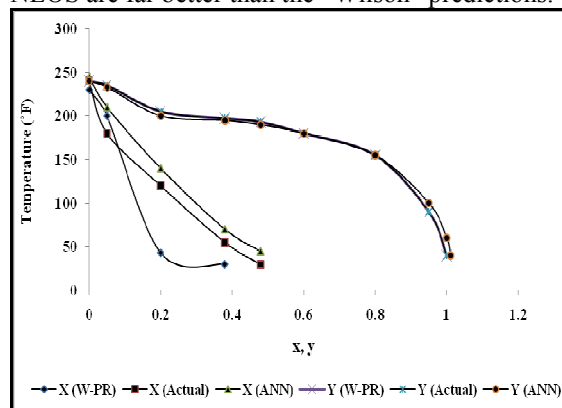
corresponding temperatures (a total of 96 data points). The net was then used to predict VLE data at 40 psia (which was not included in the training data) and the results are presented in Fig. 1. A comparison of the predicted and the "actual" curves shows that the net was able to predict the VLE data with an overall absolute error of only 0.006 for a pressure that was not a part of the training set.



Figures 1: VLE predictions for $\text{NH}_3/\text{H}_2\text{O}$ system at 40 psia (not a training set).

Comparison of NEOS and EOS data

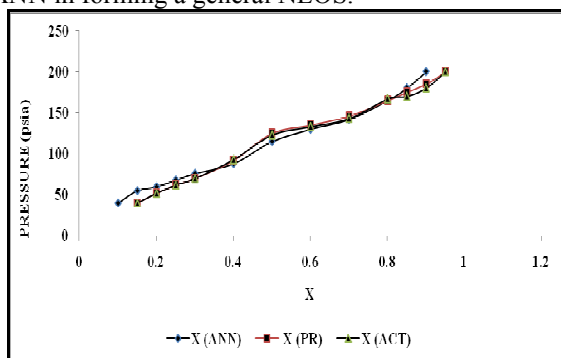
In order to establish NEOS as a plausible alternative to the thermodynamic methods for VLE data prediction of a highly polar system ($\text{NH}_3 - \text{H}_2\text{O}$), the results obtained by NEOS were compared with those obtained using the best available activity coefficient approach (Gupta and Vashishtha, 1997). Wilson parameters were found to be best suited for this purpose (Reid et al., 1977). Wilson's parameters were estimated by reducing the VLE data for the system at 100°F . The vapour phase fugacity coefficients were calculated using the Peng-Robinson EOS (1976). The results of such predictions are plotted along with the predictions of NEOS and actual values at 15 psia in Fig. 2. The vapour phase predictions of both the approaches appear to be good but liquid phase predictions of NEOS are far better than the "Wilson" predictions.



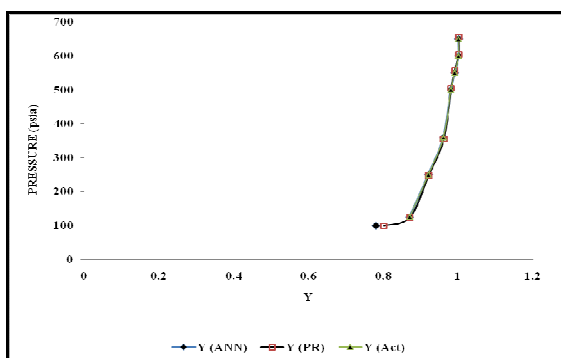
Figures 2: VLE predictions for $\text{NH}_3/\text{H}_2\text{O}$ system at 15 psia.

CH₄ – C₂H₆ System

The VLE curves for this system using NEOS show that the mapping of the P-T-y surface is better than that of the P-T-x surface. Typical results are presented in Figs. 3 and 4. The figures also present the predicted values using the Peng-Robinson EOS. It is clearly visible that the NEOS predictions are as good as the EOS predictions for such a system even with a limited database. Although the results are not exhaustive for a general “NEOS” applicable to all systems, they seem to be conclusive, in principle, to the capability of an ANN in forming a general NEOS.



Figures 3: VLE data for CH₄/C₂H₆ system at 284.7 R.



Figures 4: VLE data for CH₄/C₂H₆ system at 343.6 R.

CONCLUSIONS

The neural network approach used in this work for VLE data prediction gave favourable results for the NH₃ – H₂O and the CH₄ – C₂H₆ systems to within an error of $\pm 1\%$. The net gave better results when generating only one surface – the P-T-x surface or the P-T-y surface – as compared to the simultaneous prediction of both. An even spread of the data seemed to be essential for better predictions. This, in principle, neural networks hold promise as a strategy for solving the tedious VLE data generation problem and the development of a “general NEOS” widely applicable to different systems. The ANNs should particularly be useful for mapping highly polar systems which fall outside the range of applicability of the conventional thermodynamic methods. Applications of ANNs for predicting other thermodynamic properties may also be explored. An heuristic approach has also been

developed which reduces the number of iterations required in the net architecture selection process.

REFERENCES

- Dwivedi, A. (1992). Neural Networks – applications in prediction of VLE data. B.Tech. Senior Project Report, Department of Chemical Engineering, MREC, Jaipur, India.
- Fredenslund, Aa, Gmehling, J., & Rasmussen, P. (1977). Vapor-liquid equilibria using UNIFAC. New York: Elsevier.
- Gupta, N., & Vashishtha, M. (1997). Neural networks : Applications and performance evaluation in prediction of VLE data. B.Tech. Senior Project Report, Department of Chemical Engineering, MREC, Jaipur, India.
- Kung, S. Y., & Hwang, J. N. Research supported by NSF and Informative Science and Technology Office of the Strategic Defence Initiative Organisation, Contact No. N00014-55-k-9469 and N0014-85-k-0599.
- Macriss, R. A. et al. (1964). Research Buletin No. 34, Institute of gas Technology, Chicago.
- Rummelhart, D. E., & McClelland, J. L. (1986). Parallel distributed processing (Vol. 1), Cambridge, MA: MIT Press.
- Reid, R. C., Prausnitz, J. M., & Sherwood, T. K. (1997). The properties of gases and liquids (3rd ed.). New York, McGraw-Hill.
- Sartori, M. A., & Panos, J. A. (1991). IEEE Trans. Neural Networks, 2, 4.
- Smith, J. M., & Van Ness, H. C. (1995). Thermodynamics for chemical engineers (5th ed.). New York: McGraw Hill.

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