

Use of Artificial Neural Network for Correlation of Vapour-Liquid Equilibrium Data for Hydrocarbon Ternary System (Ethane-n-Butane-n-Pentane)

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Abstract

In this study, existing experimental vapour-liquid equilibrium (VLE) data covering a wide range of temperature, phase composition and pressure for ethane-n-butane-n-pentane was correlated using MATLAB (Matrix Laboratory) software. To increase the reliability of correlations, neural network was trained using existing vapour-liquid equilibrium data with the aid of Levenberg Marquardt algorithm. Network parameters are fine-tuned until the output generated by simulation are checked and observed to match with pre-determined experimental V L E data. It was found that there is high degree of coherence between the chosen targets from experimental data and predicted values. This confirms that correlations and predictions of V L E data using neural network is efficient and significant.

Key words: Correlation, Levenberg Marquardt algorithm, MATLAB, Network parameters, Neural network, Simulation, Vapour-liquid equilibrium (VLE)

Word count: 126

INTRODUCTION

Recently, many methods and approaches have been used to determine vapour-liquid equilibrium data for hydrocarbon binary, ternary and multi-components because of its relevance and great importance in chemical process industries, refining and petroleum industries. A few of these approaches are: graphical, analytical and statistical approaches. However, it was found that the correlation with these approaches were not so accurate as expected and in some cases it proved quite difficult to correlate unfamiliar data and operating conditions. Due to these frustrating efforts, artificial neural network (ANN) or machine learning algorithm (embedded in the MATLAB) comprising of a large number of highly interconnected processing elements (neurons) was developed for correlating vapour-liquid equilibrium data because it can reason more dynamically like the human brain, follow trend, capable of self training and can organize both enormous, familiar and unfamiliar data.

Vapour liquid equilibrium (abbreviation VLE) is a state where liquid and its vapour (gas phase) are in equilibrium: a condition or a state where the rate of evaporation (liquid becoming vapour) is equal to the rate of condensation (vapour becoming liquid), under specific conditions where no net vapour liquid inter-conversion intervenes [1]. When heat is added slowly to a closed vessel that contains liquid mixture of several components at constant pressure, the

temperature increases until a point is reached when the first bubble forms, at this point, vapour and liquid is said to be in a state of equilibrium [2]. As vapourization proceeds, the composition of the remaining liquid continually changes and hence does its vapourization temperature. A similar phenomenon occurs when a mixture of vapour is subjected to a condensation process at constant pressure, at some temperature. The first droplets of liquid form, thereafter the compositions of vapour and the condensation temperature both changes. When a liquid is heated slowly, at constant pressure, the temperature at which the first vapour bubble forms is the bubble point temperature of the liquid at a given pressure and when a gas (vapour) is cooled slowly at constant pressure, the temperature at which the first liquid droplet forms is the dew temperature [2] [3].

When two phases are in equilibrium, vapour-liquid equilibrium data is useful in relating the composition of the liquid phase to that of the vapour phase [4].

The modeling and control of equipment for different chemical processes require the vapour-liquid equilibrium data [5] [6].

The thermodynamics of ternary phase equilibrium is based on the concept of the chemical potential. The chemical potential was introduced by Gibbs while G. N. Lewis

proposed the idea of the quantity called fugacity, which has a unit pressure.

Since, in a mixture of ideal gases, the fugacity of each component is equal to its partial pressure. Physically, it is convenient to think of the fugacity as thermodynamic pressure, in real mixtures, the fugacity can be considered as a partial pressure which is corrected for non-ideal behavior. Fugacity F_i is related to partial pressure of a system when the system approaches or behaves like an ideal situation. Thus,

$$\frac{F_i}{y_i P} \quad \text{as } p \rightarrow 0 \quad (1)$$

Pressure is low and approaches zero

$y_i = \text{mole fraction of component } i \text{ in the vapour phase}$

$P = \text{total pressure}$

For two phases which are at the same temperature, the equation of equilibrium for each component is expressed in terms of fugacity F_i as

$$F_i = F_L \quad (2)$$

Thus, fugacity coefficient Φ relates the vapour phase fugacity F_i^V to vapour phase mole fraction y_i and total pressure P by

$$\Phi = \frac{F_i^V}{y_i P} \quad (3)$$

The activity coefficient γ on the other hand relates the liquid phase fugacity F_i^L to the liquid phase mole fraction x_i and to the standard state fugacity F_i^{01} as

$$\gamma_i = \frac{F_i^L}{x_i F_i^{01}} \quad (4)$$

Combining equations 3 and 4, the equation for any component i becomes:

$$F_i^V = F_i^L \quad (5); \text{ and}$$

$$\Phi_i y_i P = \gamma_i x_i F_i^{01} \quad (6)$$

The relation between fugacity coefficient Φ and activity coefficient γ is may be given as

$$\Phi_i y_i P = \gamma_i x_i P_i \quad (7)$$

For non ideal gas and liquid solution

$$\Phi_i y_i P = \gamma_i x_i P_i^{Sat} \Phi^{Sat} \quad (8)$$

Equation 7 reduces to

$$y_i P = x_i P_i \quad (9)$$

$P_i = \text{vapour pressure}$

$P_p = \text{Partial pressure}$

P^{Sat} is the saturation vapour pressure of pure component i

Φ^{Sat} is the fugacity coefficient at saturation of pure component i

but

$$y_i P = P_p \tag{10}$$

Equation 9 becomes

$$P_p = x_i P_1 \tag{11}$$

Activity coefficient is related to molar Gibb's energy by:

$$n_T G^E = RT \sum_i n_i \ln \gamma_i \tag{12}$$

$$RT \ln \gamma_i = \left(\frac{\partial(nG^E)}{\partial n_i} \right)_{T,P,n_j \neq i} \tag{13}$$

$$\ln \gamma_i = \frac{G^E}{RT} = \frac{1}{RT} \left(\frac{\partial(nG^E)}{\partial n_i} \right)_{T,P,n_j \neq i} \tag{14}$$

where

R is the ideal gas constant and T is temperature

[7], [8], [9], [10], [11], and [12], have all reported the correlation of vapour-liquid equilibrium data for binary systems while [13] have shown how NRTL equation can be used to correlate vapour-liquid equilibrium data for ternary systems. In this work, MATLAB, software with neural network conditions was used to correlate experimental vapour-liquid equilibrium data with the aid of Levenberg Marquardt algorithm employed to train the set of data. The Levenberg Marquardt algorithm is one of the best training rules designed to approach network speed [14], cited by [8]. Neural network is an interconnected group akin to the vast network of neuron in the human brain. It is an information processing paradigm that is inspired by the way biological system neurons such as the brain process information [15]. The artificial neural network is a model based on some experimental results that is proposed to predict the required data because of avoiding more experiments. [8]. In the course of data correlation, neural network provides a network which organizes and provides good pattern for vapour- liquid equilibrium data.

Table 1.0: Experimental vapour-liquid equilibrium data for the ethane-n-butane-pentane ternary system

Source: [16], [17]

Pressure (kPa)	Ethane vapour mole	Butane vapour mole	Pentane vapour mole		Ethane liquid mole	Butane liquid mole	Pentane liquid mole

	fraction (y ₂)	fraction (y ₄)	fraction (y ₅)		fraction (x ₂)	fraction (x ₄)	fraction (x ₅)
2444.3	0.846	0.102	0.052		0.517	0.274	0.209
2855.2	0.875	0.086	0.039		0.602	0.229	0.169
3026.2	0.860	0.077	0.037		0.713	0.175	0.112
3271.5	0.882	0.081	0.037		0.765	0.144	0.091
3386.0	0.872	0.085	0.043		0.794	0.128	0.078
2111.7	0.828	0.149	0.023		0.510	0.378	0.112
2507.8	0.852	0.122	0.020		0.599	0.312	0.089
2903.0	0.871	0.111	0.018		0.690	0.245	0.065
3146.4	0.871	0.110	0.019		0.740	0.208	0.052
3349.46	0.865	0.115	0.020		0.788	0.170	0.042
2164.2	0.893	0.046	0.061		0.560	0.131	0.309
2254.2	0.898	0.043	0.059		0.064	0.113	0.246
2923.7	0.904	0.041	0.055		0.718	0.093	0.246
3316.6	0.911	0.041	0.051		0.793	0.076	0.131
3439.6	0.910	0.038	0.052		0.823	0.068	0.109
3517.5	0.904	0.041	0.055		0.836	0.061	0.103

RESULTS AND DISCUSSION

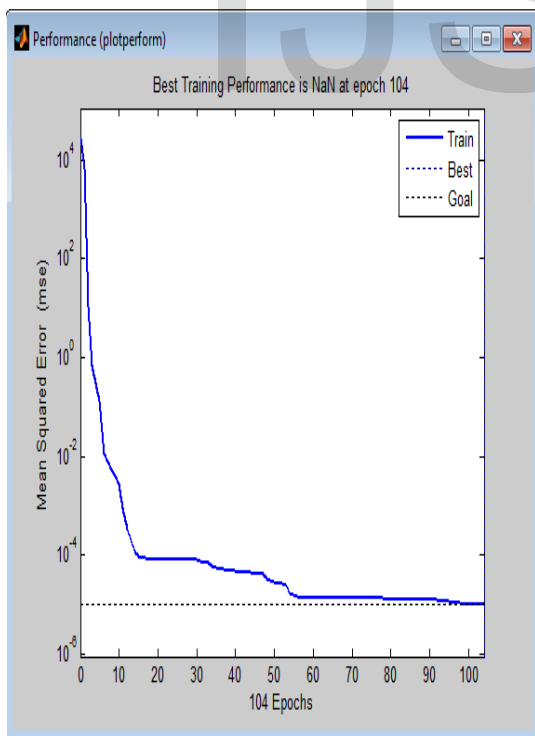


Fig.1.0: Performance Trainlm Graph

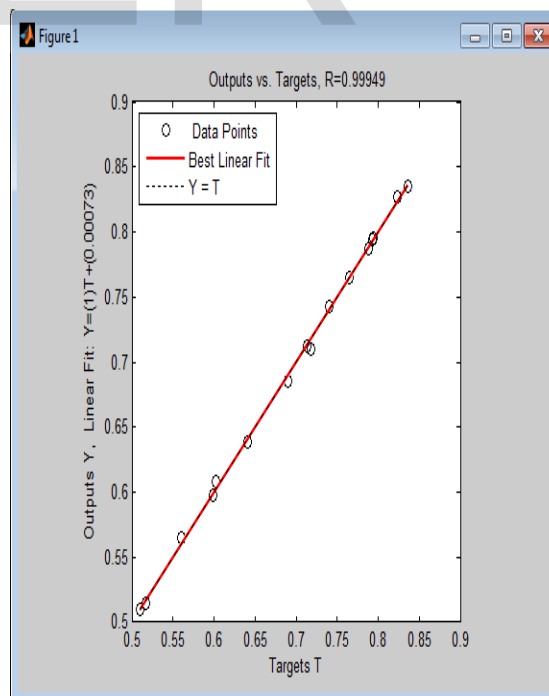


Fig.3.0: Output-target Graph (Ethane)

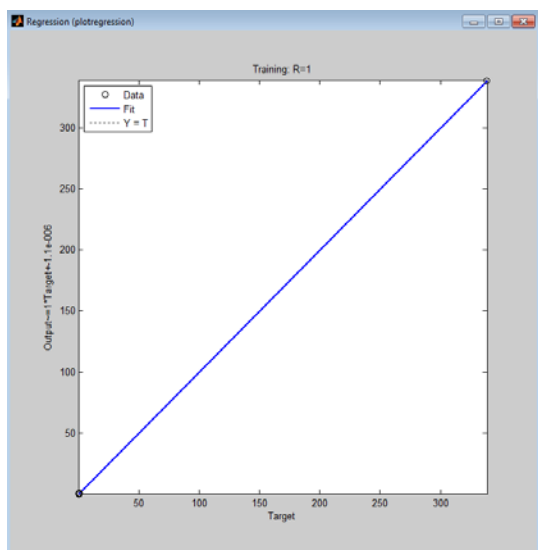


Fig. 2.0: Regression Graph

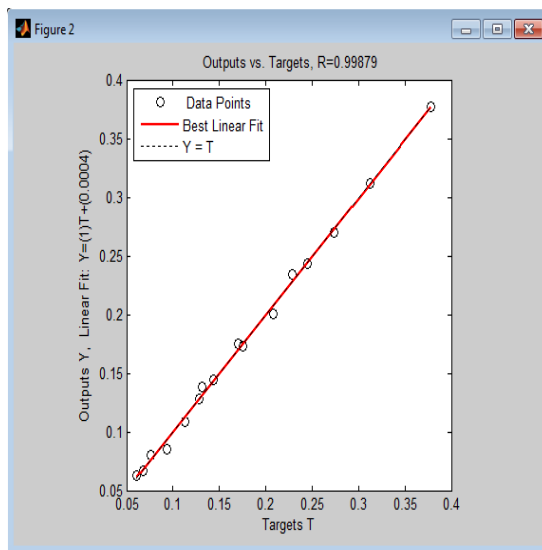


Fig. 4.0: Output-target Graph (Butane)

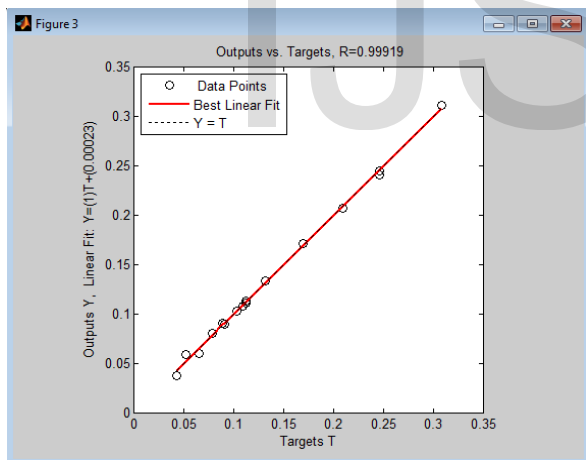


Fig. 5.0: Output-target Graph (Pentane)

Table 2.0: Table showing liquid mole fraction and temperature values from experimental data (target)

Ethane (x ₂)	Butane (x ₄)	Pentane (x ₅)	Temperature (K)
0.517	0.274	0.209	338.555
0.602	0.229	0.169	338.555
0.713	0.175	0.112	338.555
0.765	0.144	0.091	338.555
0.794	0.128	0.078	338.555
0.510	0.378	0.112	338.555
0.599	0.312	0.089	338.555
0.690	0.245	0.065	338.555
0.740	0.208	0.052	338.555
0.788	0.170	0.042	338.555
0.560	0.131	0.309	338.555
0.641	0.113	0.246	338.555
0.718	0.093	0.246	338.555
0.793	0.076	0.131	338.555
0.823	0.068	0.109	338.555
0.836	0.061	0.103	338.555

Table 3.0: Table showing predicted values of liquid mole fraction and pressure with the use of Artificial Neural Network (A N N)

Ethane (x ₂)	Butane (x ₄)	Pentane (x ₅)	Temperature (K)
0.5173	0.2738	0.2091	338.5500
0.6029	0.2290	0.1690	338.5550
0.7127	0.1734	0.1122	338.5551
0.7649	0.1450	0.0900	338.5551
0.7957	0.1284	0.0780	338.5549
0.5104	0.3776	0.1121	338.5550
0.5980	0.3121	0.0890	338.5552
0.6857	0.2443	0.0652	338.5551
0.7426	0.2060	0.0520	338.5550
0.7866	0.1708	0.0422	338.5550
0.5653	0.1310	0.3090	338.5551
0.6400	0.1131	0.2455	338.5500
0.7105	0.0920	0.2460	338.5500
0.7942	0.0762	0.1314	338.5500
0.8266	0.0667	0.1090	338.5550
0.8349	0.0617	0.1033	338.5549

Simulation takes the network input P and the network object and returns the network output a. In this work, the simulation is repeated as a loop. In the loop, the model is run for specific time, parameter changed and model run again. This was repeated until the correlation was completed and the simulation was completed when the target was met after 104 iterative processes. This is shown in Fig. 1.0, with the trainlm line (performance goal line) touching the target line.

The small value of the mean square error (M S E) as shown in Fig. 1.0 is an indication that the developed network has efficiently trained the data and that the output (predicted values) are not far from the target. Comparing table 2.0 and 3.0, the output (predicted values) of liquid phase compositions and temperature gives set of data that is close to the target (experimental liquid phase compositions and temperature data).

From Fig. 2.0, 3.0, 4.0 and 5.0 (regression and output-target) graphs, deviations of data points from the line of best fit is negligible which indicates that there is high degree of correlation between predicted values and experimental data. Correlation coefficient R will be significant when the indices given by [i, j] in the P matrix is less or equal to 0.05. Correlation coefficient R in Fig. 2.0 (Regression graph) is 1, which means there is 100% correlation between the predicted and experimental vapour-liquid equilibrium data: an indication that the developed neural network is highly efficient. The value of R in Fig. 3.0, 4.0 and 5.0 is given as 0.99949, 0.99879 and 0.99919 respectively meaning that there is 99.949%, 99.879% and 99.919% correlation between output and the set target for ethane-butane-pentane ternary system. It also confirms that there is good correlation between the input, target and output parameter.

From the experimental data, the values of pressure and vapour phase composition of the ternary system were given as input to the network while the target was taken to be the

respective temperatures and liquid phase composition.

Comparing table 2.0 and 3.0, the output (predicted values) of liquid phase compositions and temperature gives set of data that is close to the target (experimental liquid phase compositions and temperature data).

Vapour mole fraction of ethane, butane and pentane picked at random at a particular pressure, [0.865; 0.115; 0.020; 3349.46], were given as input to the trained network which predicted its equivalent composition of the liquid phase and temperature as [0.7881; 0.1699; 0.0420; 3380.55]. These values agree with the experimental data.

CONCLUSIONS AND RECOMMENDATIONS

Neural network was successfully used to correlate vapour-liquid equilibrium data for hydrocarbon ternary systems (ethane-n-butane-n-pentane). The network was highly efficient and suitable for training the experimental data because the set target and performance goal was met. The accuracy of prediction of neural network increases when the network is iteratively trained. For the ternary systems, mean square errors are negligible and predicted values were very close to the targets. In addition, good correlation results for hydrocarbon ternary systems were achieved as correlation coefficient is close to 1 for each of the system. A successful

validation test evidenced in the corresponding predicted values of liquid phase compositions and its equivalent vapour phase compositions also indicates that the predictive capability of the network is highly efficient. All these confirmed conclusively that neural network is a viable tool for the correlation of vapour-liquid equilibrium data for hydrocarbon ternary system.

Research work can however be extended to other ternary systems like alcohol.

References

- [1] Coulson, J. M. and Richardson, J. F. Particles Technology and Separation Process, Vol., 2, 5th Edition, Sydney, Butterworth Heinemann. Pp. 206-264, (2002).
- [2] Processium. Vapour-liquid Equilibrium. <http://en.processium.com/thermodynamics/vapour-liquid-equilibrium.htm> (accessed: 04-02-2013)
- [3] McCabe, W. L. Smith, J. C. and Harriot, P. Unit Operations of Chemical Engineering, 7th Edition, Mc Graw Hil, Milan. Pp 20-64, (2005).
- [4] Philip, C. W. Separation in Chemical Engineering, Oxford Press, New York. Pp 271-291, (1997).
- [5] Yamamoto, H. and Tochigi, K. Prediction of Vapour-Liquid Equilibria Using Reconstruction Learning Neural Network Method. Fluid Phase Equilibria. 257: 169-172, (2007).

- [6] Ernest, E. L. Applied Process Design. Vol., 2, 3rd Edition Plenum Press, New York. Pp 6-17, (2002).
- [7] Mitra, D., Roy, D., Bandyopadhyay, M and Bhattacharya, P. Prediction of Vapour-liquid Pseudo-equilibrium for Pervaporation of Benzene-cyclo-hexane System. Indian Journal of Chemical Technology, Vol.13 pp. 290- 293, (2006).
- [8] Goral M., Skrzecz, A., Bok, A., Maczynski, A. and Oracz . Journal of Phy.Chem. Ref. Data. 33, 959, (2004).
- [9] Kasprzycka-Gutman, T. and Oracz, P. Recommended Vapour-Liquid Equilibrium Data. Part 4. Binary Alkanol-Alkene/Alkyne Systems. Journal of Physical Chem. Ref. Data.Vol., 35 (4) 1577-1595, (2006).
- [10] Rattan, V. K., Gill, B. K. and Kapoor, S. Isobaric Vapour-Liquid Equilibrium Data for Binary Mixture of 2- methyl tetrahydrofuran and Cumene. World Academy of Science, Engineering and Technology. (23) 41-44, (2008).
- [11] Moghadassi, A. R., Nikkholgh, M. R., Hosseini, S. M, Parvizian, F. and Sanaeirad, A. Prediction of Vapour-Liquid Equilibrium (V LE) Data for Binary Systems: Case Study: Methane/tetra fluoro methane. Journal of Engineering and Applied Sciences. Vol., 6 No (8):1-8, (2011).
- [12] Guha, A. and Pal, R. Prediction of Vapour-Liquid Equilibrium (V LE) of Some Binary Liquid Systems by Generalized London Potential Method. Indian Journal of Chemical Technology. Vol., 14, pp. 178-182, (2007).
- [13] Graczova, E., Steltenpohl, P., Balintova, L. and Krsackova, E. Correlation of Vapour-Liquid Equilibrium Data of Ternary Systems. Chem. Pap. 58(6) 442- 446, (2004).
- [14] Demuth, H. and Beale, M. Neural Network Tool box. User's Guide. The Math Works Inc., M. A. (2002).
- [15] Okeyinka, A. E. Introduction to Modelling and Simulation of Physical Systems. Pp. 120-124, (2006).
- [16] Dasturt, S. P. and Thodos G. Chemical Engineering Science. Vol. 19, Oxford Pergaman Press Ltd. Pp. 935-952, (1964).
- [17] Dasturt, S. P. and Thodos G. Vapour-Liquid Studies for Ethane-Butane- Pentane Ternary System at 150⁰C. Journal of Chemical Engineering. Vol., 7, No 3: 1-17, (1964).