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PREDICTION OF PVT PROPERTIES OF AMMONIA BY USING ARTIFICIAL NEURAL NETWORK AND EQUATIONS OF STATE

Abdolreza Moghadassi[1], Fahime Parvizian[1], Sayed Mohsen Hosseini[1] and Amir Sharifi[2]
[1]Department of Chemical Engineering, Faculty of Engineering, Arak University, Arak, Iran
[2]Department of Chemical Engineering, Faculty of Engineering, Farahan branch, Azad University, Arak, Iran
E-Mail: Fahime_Parvizian@yahoo.com

ABSTRACT

Ammonia is an important gas that plays significant role in many processes. Consequently, knowledge of the thermodynamic properties of Ammonia is necessary for the interpretation of physical and chemical processes. A new method based on Artificial Neural Networks (ANN) for prediction of thermodynamic properties has been proposed for both superheated and saturated region of Ammonia. For this development, the data sets that collected from Ammonia thermodynamic table [Perry's Chemical Engineering Handbook] were used. After training the networks, the models were tested by unseen data to evaluate their accuracy and trend stability. Among this training the back-propagation learning algorithm with various training such as Scaled Conjugate Gradient (SCG), Levenberg-Marquardt (LM) and Resilient Back propagation (RP) methods were used. The best suitable algorithm with appropriate number of seven neurons in the hidden layer which provides the minimum Mean Square Error (MSE), 0.0000900135, is found to be the SCG algorithm. Then ANN's results were compared with results of some equations of state such as Lee Kesler, NRTL, Soave-Redlich-Kwong and Peng Robinson. Comparisons showed the ANN capability for prediction of the thermodynamic properties of Ammonia.

Keywords: model, ammonia, artificial neural network, thermodynamic, equation of state.

1. INTRODUCTION

Ammonia (NH_3) is normally encountered as a gas with a characteristic pungent odor. Ammonia contributes significantly to the nutritional needs of terrestrial organisms by serving as a precursor to foodstuffs and fertilizers. Ammonia, either directly or indirectly, also is a building block for the synthesis of many pharmaceuticals. Consequently, knowledge of the thermodynamic properties of Ammonia is important for the interpretation of physical and chemical processes. In physics and thermodynamics, an equation of state is a relation between state variables such as its temperature, pressure, volume, or internal energy. Equations of state are useful in describing the properties of fluids, mixtures of fluids, solids, and even the interior of stars [1]. A number of much more accurate equations of state such as Vander Waals, Soave-Redlich-Kwong, Peng Robinson, Lee-Kesler and NRTL have been developed for gases and liquids. However, this equation becomes increasingly inaccurate at higher pressures and lower temperatures, and fails to predict condensation from a gas to a liquid. The most prominent use of an equation of state is to predict the state of gases and liquids. Besides the high costs of the experimental work it is difficult if not impossible, to get a clear picture of the condition and possible problems of the work. Therefore a model based on some experimental results is proposed to predict the required data instead of doing more experiments. The major processes in the chemical engineering are unfortunately nonlinear. ANN is a model that attempts to mimic simple biological learning processes and simulate specific functions of human nervous system. This model creates a connection between input and output variables and keeps the underlying complexity of the process inside the system. The ability to learn the behavior of the data generated by a system is the neural network's versatility and privilege [3]. Fast response, simplicity, and capacity to learn are the advantages of ANN compared to classical methods. This model has been widely applied to predict the physical and thermodynamic properties of chemical compounds. ANN has recently been used to predict some pure substances and petroleum fraction's properties [4], activity coefficients of isobaric binary systems [5], thermodynamic properties of refrigerants [6,7,8] and activity coefficient ratio of electrolytes in amino acid's solutions [9] etc. To the best of our knowledge no attempt has been made to model the thermodynamic properties of water by artificial neural network. Defining the ANN and selecting the best ANN predictor to predict the thermodynamic properties of saturated and superheated water instead of approximate and complex analytical equations are the main focus of this work. In the following sections after ANN introduction, the best ANN predictor is chosen. Finally results of the ANN model is evaluated against with the unseen data and then compared with the experimental work.

2. ARTIFICIAL NEURAL NETWORKS

In order to find relationship between the input and output data derived from experimental work, a more powerful method than the traditional ones are necessary. ANN is an especially efficient algorithm to approximate any function with finite number of discontinuities by learning the relationships between input and output vectors [4, 10]. These algorithms can learn from the experiments, and also are fault tolerant in the sense that they are able to handle noisy and incomplete data. The ANNs are able to deal with non-linear problems, and once trained can

perform prediction and generalization rapidly [11]. They have been used to solve complex problems that are difficult to be solved if not impossible by the conventional approaches, such as control, optimization, pattern recognition, classification, and so on. Especially it is desired to have the minimum difference between the predicted and observed (actual) outputs [12]. Artificial neural networks are biological inspirations based on the various brain functionality characteristics. They are composed of many simple elements called neurons that are interconnected by links and act like axons to determine an empirical relationship between the inputs and outputs of a given system. Multiple layers arrangement of a typical interconnected neural network is shown in Figure-1. It consists of an input layer, an output layer, and one hidden layer with different roles. Each connecting line has an associated weight. Artificial neural networks are trained by adjusting these input weights (connection weights), so that the calculated outputs may be approximated by the desired values. The output from a given neuron is calculated by applying a transfer function to a weighted summation of its input to give an output, which can serve as input to other neurons, as follows [13]:

$$\alpha_{jk} = F_k\left(\sum_{i=1}^{N_{k-1}} w_{ijk}\alpha_{i(k-1)} + \beta_{jk}\right) \quad (1)$$

Where α_{jk} is neuron j's output from k's layer β_{jk} is the bias weight for neuron j in layer k. The model fitting parameters w_{ijk} are the connection weights. The nonlinear activation transfer functions F_k may have many different forms.

The classical ones are threshold, sigmoid, Gaussian and linear function, etc… [8], for more details of various activation functions see Bulsari [13]. The training process requires a proper set of data i.e. input (I_i) and target output (t_i). During training the weights and biases of the network are iteratively adjusted to minimize the network performance function [16]. The typical performance function that is used for training feed forward neural networks is the network Mean Squares Errors (MSE) Eq. (2).

$$MSE = \frac{1}{N}\sum_{i=1}^{N}(e_i)^2 = \frac{1}{N}\sum_{i=1}^{N}(t_i - \alpha_i)^2 \quad (2)$$

There are many different types of neural networks, differing by their network topology and/or learning algorithm.

In this paper the back propagation learning algorithm, which is one of the most commonly used algorithms is designed to predict the thermodynamic properties of water. Back propagation is

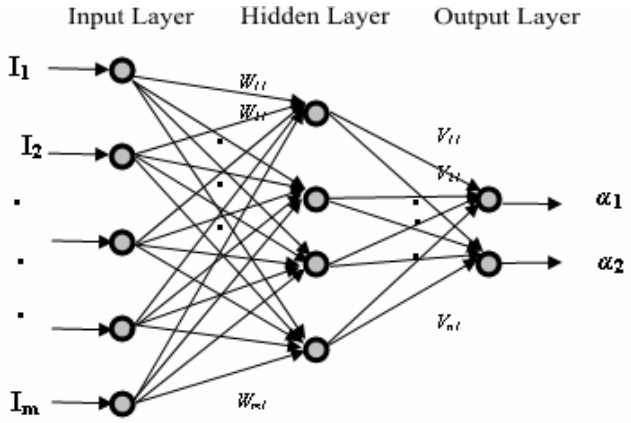

Figure-1. Schematic of typical multi-layer neural network model.

a multilayer feed-forward network with hidden layers between the input and output [6]. The simplest implementation of back propagation learning is the network weights and biases updates in the direction of the negative gradient that the performance function decreases most rapidly. An iteration of this algorithm can be written as follows [13]:

$$x_{k+1} = x_k - l_k g_k \quad (3)$$

Where x_k is vector of weights, g_k is gradient and l_k is learning rate. There are various back propagation algorithms Such as Scaled Conjugate Gradient (SCG), Levenberg-Marquardt (LM), Gradient Descent with Momentum (GDM), variable learning rate Back propagation (GDA) and Resilient back Propagation (RP). LM is the fastest training algorithm for networks of moderate size and it has the memory reduction feature to be used when the training set is large. One of the most important general purpose back propagation training algorithms is SCG [10, 11].

The neural nets learn to recognize the patterns of the data sets during the training process. Neural nets teach themselves the patterns of the data set letting the analyst to perform more interesting flexible work in a changing environment .Although neural network may take some time to learn a sudden drastic change, but it is excellent to adapt constantly changing information. However the programmed systems are constrained by the designed situation and they are not valid otherwise. Neural networks build informative models whereas the more conventional models fail to do so. Because of handling very complex interactions, the neural networks can easily model data, which are too difficult to model traditionally (inferential statistics or programming logic). Performance of neural networks is at least as good as classical statistical modeling, and even better in most cases [16]. The neural networks built models are more reflective of the data structure and are significantly faster.

Neural networks now operate well with modest computer hardware. Although neural networks are

computationally intensive, the routines have been optimized to the point that they can now run in reasonable time on personal computers. They do not require supercomputers as they did in the early days of neural network research.

3. THERMODYNAMIC MODELS

3.1. Soave-Redlich-Kwong (SRK) equation of state

Introduced in 1949 the Redlich-Kwong equation of state was a considerable improvement over other equations of the time. It is still of interest primarily due to its relatively simple form. While superior to the Van der Waals equation of state, it performs poorly with respect to the liquid phase and thus cannot be used for accurately calculating vapor-liquid equilibria. However, it can be used in conjunction with separate liquid-phase correlations for this purpose [14].

$$p = \frac{RT}{V_m - b} - \frac{a}{\sqrt{T}V_m(V_m + b)} \quad (4)$$

$$a = \frac{0.42748R^2T_c^{2.5}}{p_c} \quad (5)$$

$$b = \frac{0.08662RT_c}{p_c} \quad (6)$$

The Redlich-Kwong equation is adequate for calculation of gas phase properties when the ratio of the pressure to the critical pressure (reduced pressure) is less than about one-half of the ratio of the temperature to the critical temperature (reduced temperature):

$$\frac{p}{p_c} \prec \frac{T}{2T_c} \quad (7)$$

In 1972 Soave replaced the a/√ (T) term of the Redlich-Kwong equation with a function α (T,ω) involving the temperature and the acentric factor. The function was devised to fit the vapor pressure data of hydrocarbons and the equation does fairly well for these materials.

$$p = \frac{RT}{V_m - b} - \frac{\alpha a}{V_m(V_m + b)} \quad (8)$$

$$\alpha = (1 + (0.48508 + 1.5517\omega - 0.15613\omega^2)(1 - T_r^{0.5}))^2 \quad (9)$$

$$T_r = \frac{T}{T_c} \quad (10)$$

Ω is the acentric factor for the species. Note especially that this replacement changes the definition of *a* slightly, as the T_c is now to the second power.

3.2. Peng-Robinson (PR) equation of state

The Peng-Robinson equation was developed in 1976 in order to satisfy the following goals:

- The parameters should be expressible in terms of the critical properties and the acentric factor;
- The model should provide reasonable accuracy near the critical point, particularly for calculations of the compressibility factor and liquid density;
- The mixing rules should not employ more than a single binary interaction parameter, which should be independent of temperature pressure and composition; and
- The equation should be applicable to all calculations of all fluid properties in natural gas processes.

$$p = \frac{RT}{V_m - b} - \frac{a\alpha}{V_m^2 + 2bV_m - b^2} \quad (11)$$

$$a = \frac{0.45724R^2T_c^2}{p_c} \quad (12)$$

$$b = \frac{0.07780RT_c}{p_c} \quad (13)$$

$$\alpha = (1 + (0.37465 + 1.54226\omega - 0.26992\omega^2)(1 - T_r^{0.5}))^2 \quad (14)$$

$$T_r = \frac{T}{T_c} \quad (15)$$

In polynomial form:

$$Z^3 - (1 - B)Z^2 + (A - 3B^2 - 2B)Z - (AB - B^2 - B^3) = 0 \quad (16)$$

$$A = \frac{a\alpha P}{R^2T^2} \quad (17)$$

$$B = \frac{bP}{RT} \quad (18)$$

Where, ω is the acentric factor of the species and *R* is the universal gas constant. For the most part the Peng-Robinson equation exhibits performance similar to the Soave equation, although it is generally superior in predicting the liquid densities of many materials, especially nonpolar ones [14].

3.3. NRTL equation of state

The NRTL (Non-Random-Two-Liquid) equation, proposed by Renon and Prausnitz in 1968, is an extension of the original Wilson equation. It uses statistical mechanics and the liquid cell theory to represent the liquid structure. These concepts, combined with Wilson's local composition model, produce an equation capable of representing VLE, LLE, and VLLE phase behavior. Like the Wilson equation, the NRTL model is thermodynamically consistent and can be applied to ternary and higher order systems using parameters regressed from binary equilibrium data. The NRTL model has accuracy comparable to the Wilson equation for VLE

systems. The NRTL combines the advantages of the Wilson and van Laar equations. Also it is an activity coefficient model that correlates the activity coefficients γ with the composition of a mixture of chemical compounds, expressed by mole fractions *x*. For a binary mixture the following equations are used [14]:

$$\ln \gamma_1 = x_2^2 \left[\tau_{21} \left(\frac{G_{21}}{x_1 + x_2 G_{21}} \right)^2 + \frac{\tau_{12} G_{12}}{(x_2 + x_1 G_{12})^2} \right] \quad (19)$$

$$\ln \gamma_2 = x_1^2 \left[\tau_{12} \left(\frac{G_{12}}{x_2 + x_1 G_{12}} \right)^2 + \frac{\tau_{21} G_{21}}{(x_1 + x_2 G_{21})^2} \right] \quad (20)$$

with

$$\ln G_{12} = -\alpha_{12} \tau_{12} \quad (21)$$

and

$$\ln G_{21} = -\alpha_{12} \tau_{21} \quad (22)$$

τ_{12} and τ_{21} as well as α_{12} are suitable parameters. In most cases the parameters τ

$$\tau_{12} = \frac{\Delta g_{12}}{RT} \quad (23)$$

and

$$\tau_{21} = \frac{\Delta g_{21}}{RT} \quad (24)$$

Are scaled with the gas constant and the temperature and then the parameters Δg_{12} and Δg_{21} are fitted. The NRTL parameters are fitted to activity coefficients that have been derived from experimentally determined phase equilibrium data (vapor-liquid, liquid-liquid, and solid-liquid) as well as from heats of mixing.

3.4. Lee Kesler (LK) equation of state

The Lee-Kesler correlation is a three parameter corresponding state method for estimating thermodynamic properties of pure, nonpolar fluids. For the compressibility factor Z, it takes the form

$$Z = Z_0 + \omega Z_1 \quad (25)$$

Where Z_0 is the compressibility factor for fluids of nearly spherical molecules ω is Pitzer's Acentric factor and Z_1 corrects for nonspherical intermolecular forces. Table and charts provide values of Z_0 and Z_1, from which Z and, hence, the molar volume can compute. At subcritical temperatures, Z_1 is typically negative ($Z_1<0$), indicating that attractive forces dominate the nonspherical contribution to Z. At supercritical temperature, Z_1 is typically positive ($Z_1>0$), indicating the dominance of repulsive forces that arise when molecules collide. Note that simple fluids have $\omega = 0$ [14].

4. EXPERIMENTAL DATA

Knowledge of the pressure/volume/temperature (PVT) behavior of natural gases is necessary to solve many petroleum engineering problems. Gas reserves, gas metering, gas pressure gradients, pipeline flow and compression of gases are some of the problems requiring precise calculation of gas density [15].

A set of data containing pressure, temperature and molar volume was collected from Chemical engineers' Handbook [14]. Table-1 lists samples of these data which were used for training and testing the neural network.

Table-1. Minimum and maximum of data used to train the Neural Network [14].

Properties	Min	Max
Pressure (kPa)	50	10000
Temperature (°C)	-30	440
Volume (m^3)	0.20351	78.605

5. NEURAL NETWORK MODEL DEVELOPMENT

Developing the neural network model to accurately predict the PVT properties of different gases requires its exposure to a large its exposure to a large data set during the training phase.

The back propagation method with SCG, LM and RP learning algorithm has been used in feed forward, single hidden layer network. Input layer neurons have no transfer functions. Similarly, inputs are the reduced temperature and reduced pressure while output is the compressibility factor. The neurons in the hidden layer perform two tasks: summing the weighted inputs connected to them and passing the result through a non linear activation function to the output or adjacent neurons of the corresponding hidden layer. The computer program has been developed under MATLAB [13]. Two thirds of data set is used to train each ANN and the rest have been used to evaluate their accuracy and trend stability. The number of the hidden layer neurons is systematically varied to obtain a good estimate of the trained data [12]. The selection criterion is the net output MSE. The MSE of various hidden layer neurons are shown in Figure-2. As it can be seen the optimum number of hidden layer neurons is determined to be 60 for minimum MSE.

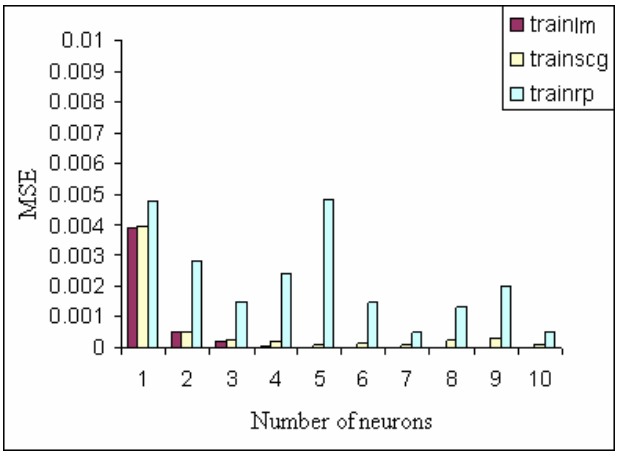

Figure-2. Determining the optimum number of neurons for the training some algorithms.

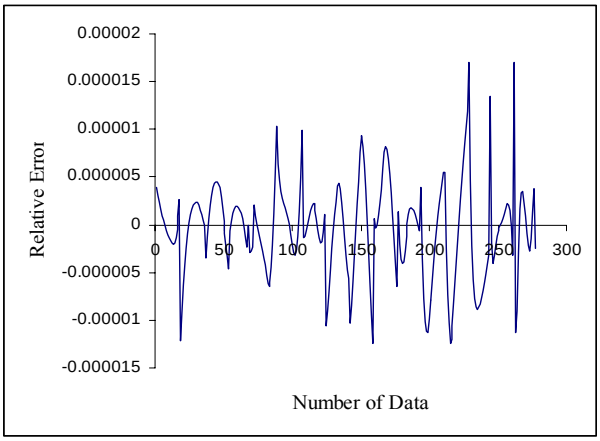

Figure-3. The relative errors between predicted data by ANN and experimental data.

Similarly the MSE of various training algorithms are calculated and listed in Table-2 for the obtained nineteen hidden layer neurons. As Table-2 shows the Levenberg-Marquardt (LM) and Scaled Conjugate Gradient (SCG) algorithms have the minimum MSE.

Table-2. MSE Comparison between different algorithms to train ANN.

Algorithm	MSE of network training
Trainscg	0.0000900135
Trainlm	0.0002428
Trainrp	0.0005799

Now the trained ANN models are ready to be tested and evaluated against the new data. Table-3 lists the various MSE of the network testing. According to these tables the Scaled Conjugate Gradient (SCG) algorithm is the most suitable algorithm with the minimum MSE.

Table-3. MSE Comparison between different algorithms to test ANN.

Algorithm	MSE of network training
Trainscg	0.000083526
Trainlm	0.00257
Trainrp	0.00318

Consequently, SCG provides the best minimum error average for both training and testing of the network. Figure-4 shows the SCG algorithm relative error fluctuations.

6. RESULTS AND DISCUSSIONS

The results have been exhibited that the ANN predictions are very close to the measurements. Figure-4 shows the scatter diagrams that compare the experimental data versus the neural network computed data. As seen, a tight cloud of points about the 45° line was obtained for these additional data points. This indicates an excellent agreement between the experimental and the calculated data.

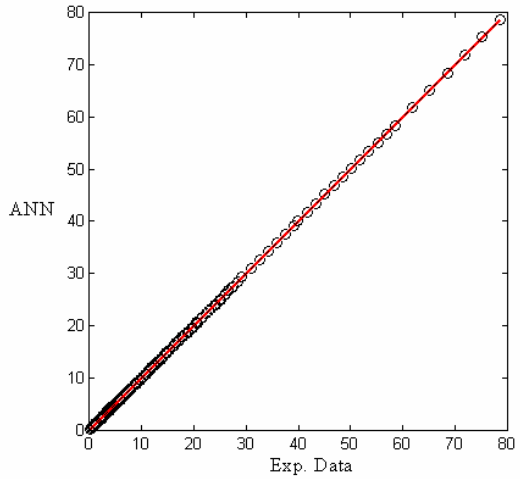

Figure-4. A typical comparison between experimental data and ANN model.

Also the amounts of volume were calculated by some equations of state such as Lee Kesler, NRTL, Soave-Redlich-Kwong and Peng Robinson and the comparisons between results of ANN and equations of state are shown in Figures (5-20).

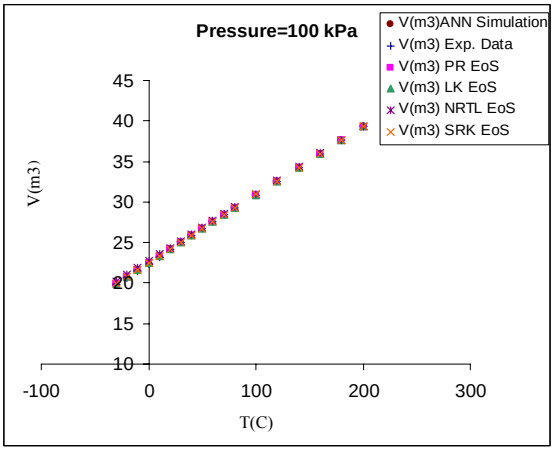

Figure-5. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 100 kPa).

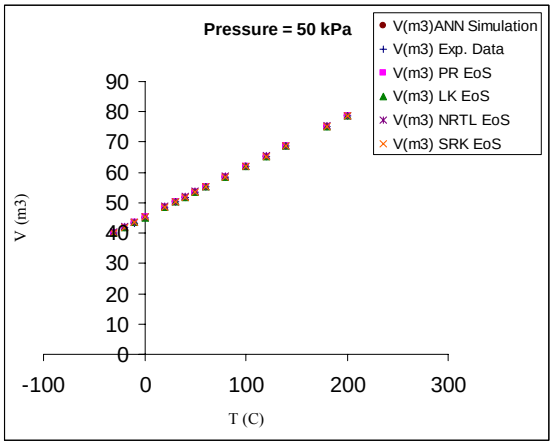

Figure-6. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 50 kPa).

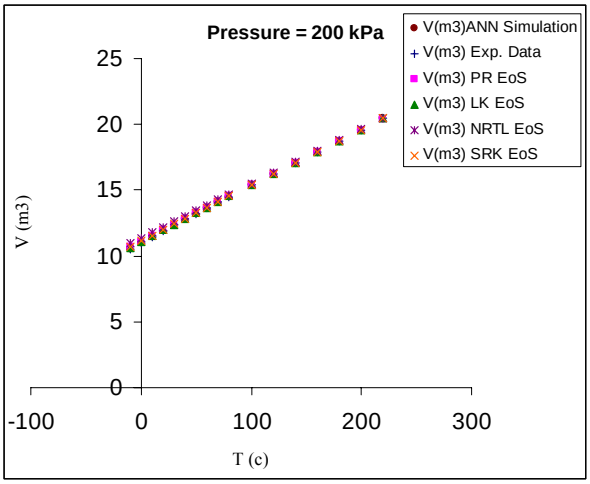

Figure-7. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 200 kPa).

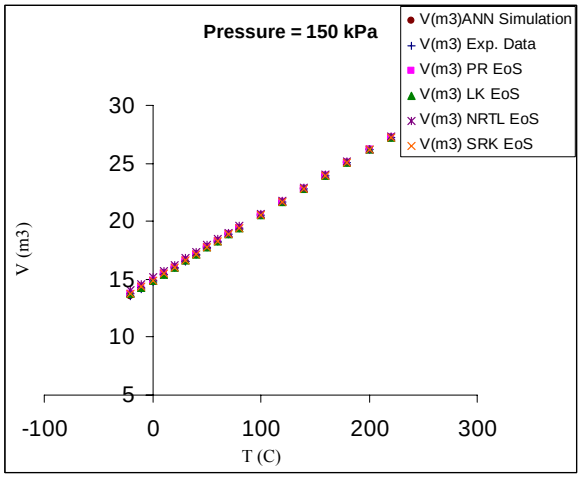

Figure-8. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 150 kPa).

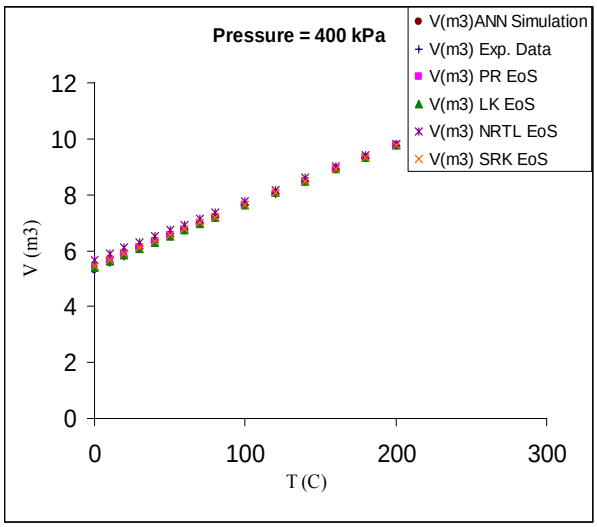

Figure-9. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 400 kPa).

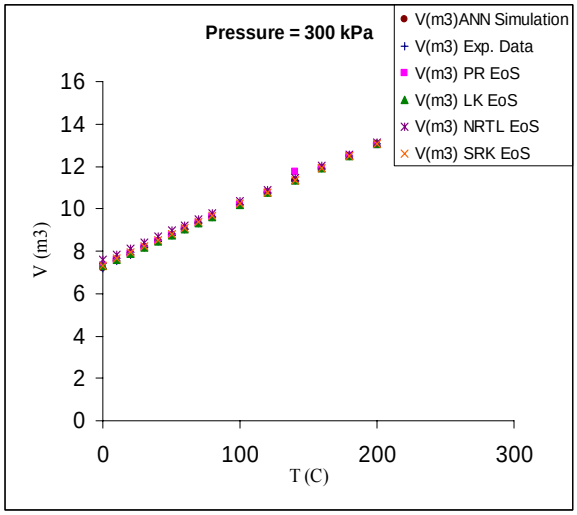

Figure-10. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 300 kPa).

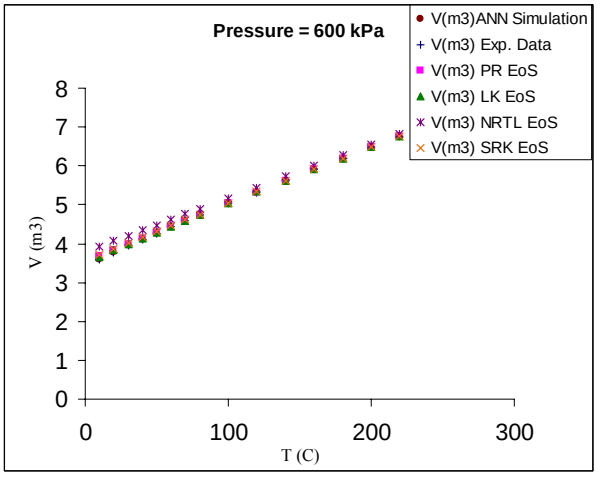

Figure-11. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 600 kPa).

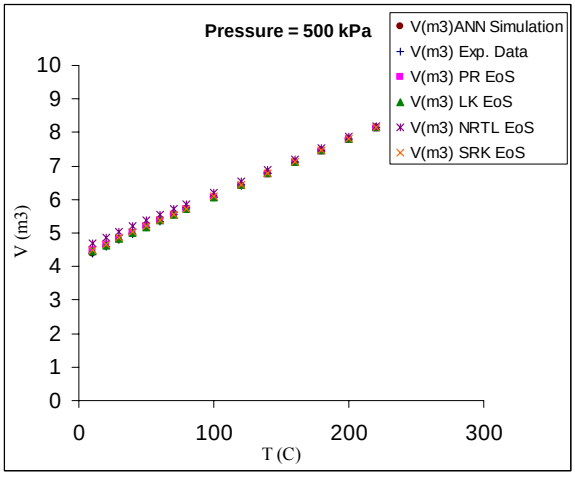

Figure-12. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 500 kPa).

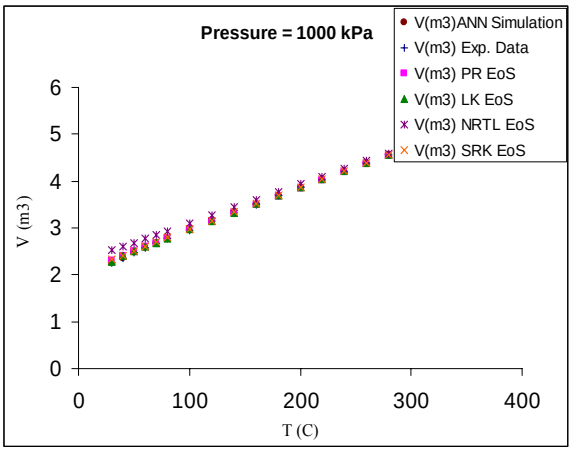

Figure-13. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 1000 kPa).

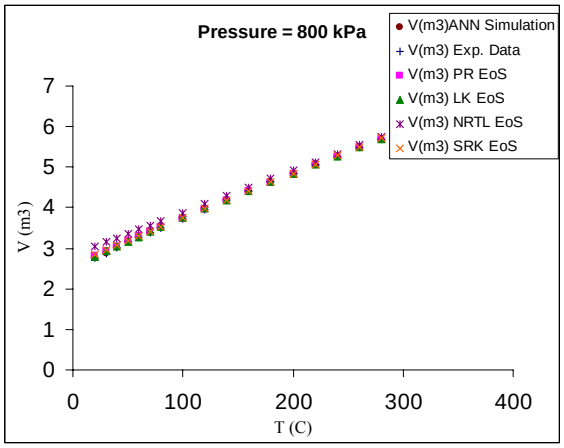

Figure-14. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 800 kPa).

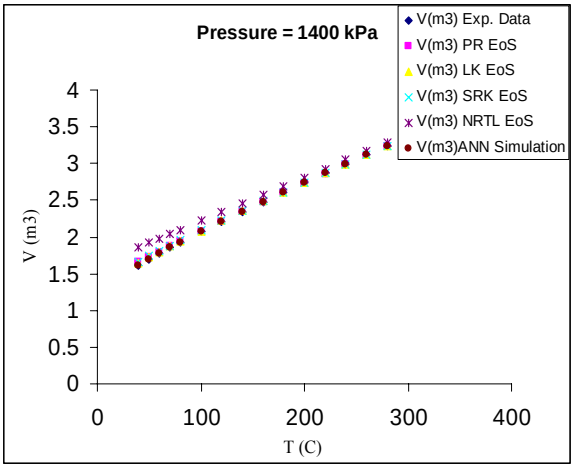

Figure-15. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 1400 kPa).

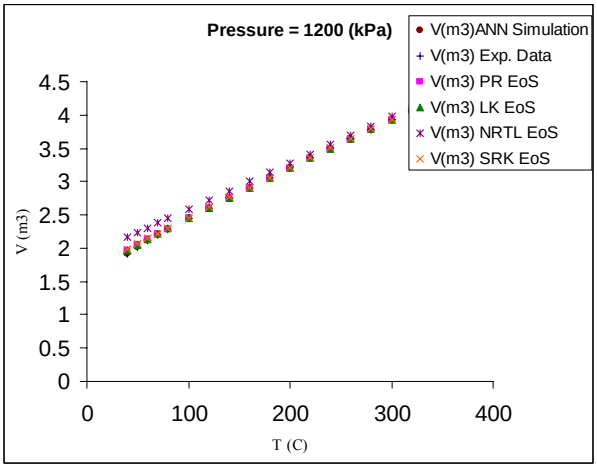

Figure-16. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 1200 kPa).

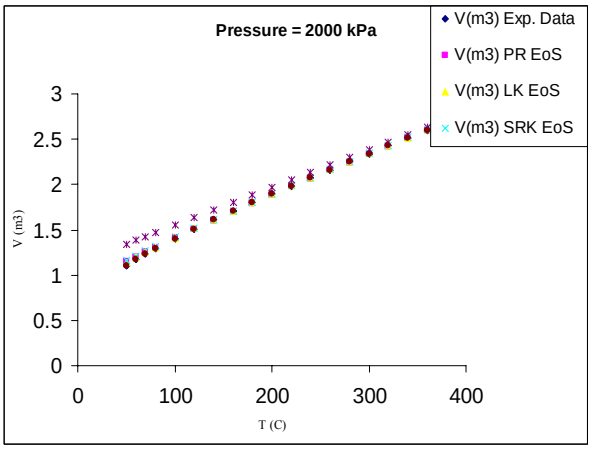

Figure-17. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 2000 kPa).

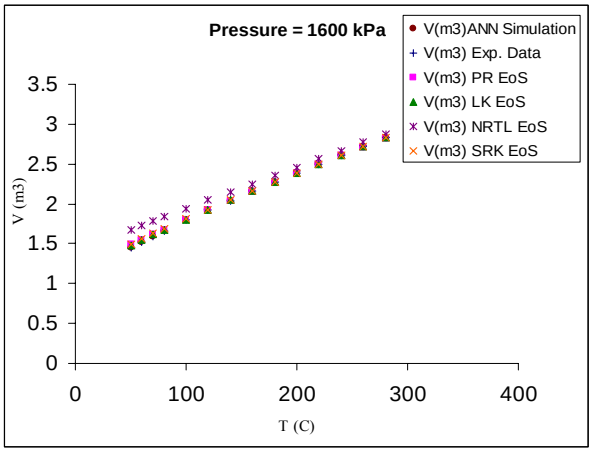

Figure-18. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 1600 kPa).

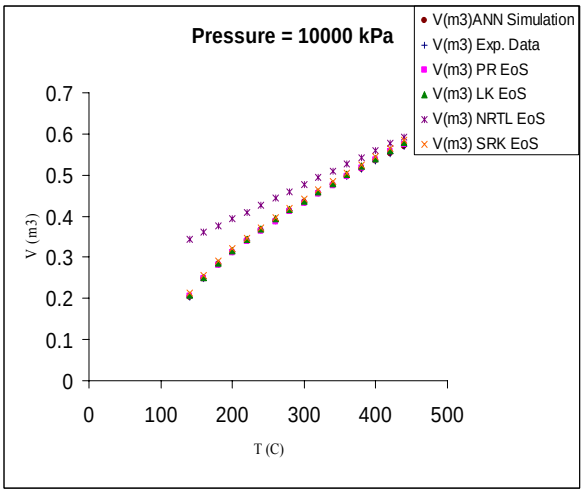

Figure-19. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 10000 kPa).

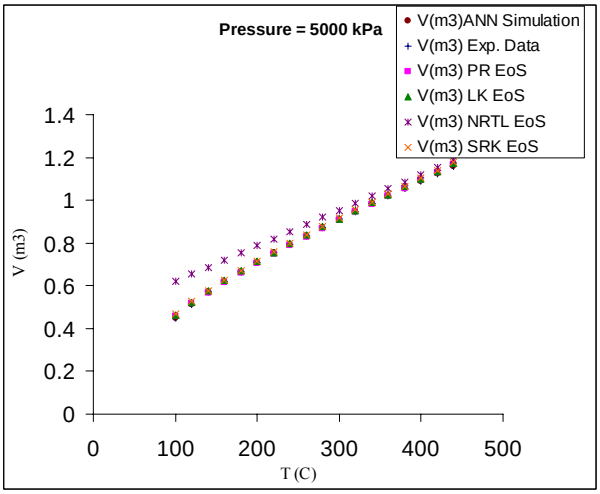

Figure-20. Comparison between ANN, Exp. data and EOS results for volume of NH_3 (P = 5000 kPa).

The results show that NRTL equation of state in high pressure has variation than experimental data. The relative errors are calculated by the following equation:

$$\text{Relative Error} = \frac{\text{Cal.} - \text{Exp.}}{\text{Exp.}} \qquad (26)$$

Table-4. listed the average relative errors of ANN simulations and EoS calculations. The results show the best estimation performance of ANN for prediction the thermodynamic properties of ammonia. Artificial neural network is a new method instead of applying the equations of state. As shown in Figure-21 artificial neural network has the best performance with minimum error that can be used for prediction of the thermodynamic properties of material. Also this figure shows the LK and PR equations of state have a good performance with comparison with SRK and NRTL.

Table-4. Comparison between average relative errors of different equations of state and neural network for different pressures.

	Methods				
Pressure	**PR** (EOS)	**LK** (EOS)	**SRK** (EOS)	**NRTL** (EOS)	**ANN**
50	0.001216	0.00083	0.001382	0.004755	4.11438E-06
100	0.002301	0.001574	0.002624	0.00925	1.1385E-05
150	0.002639	0.001823	0.003079	0.012017	1.0777E-05
200	0.002801	0.001971	0.003366	0.014554	3.04134E-05
300	0.004979	0.002228	0.003795	0.018283	6.46712E-06
400	0.004274	0.003077	0.005238	0.024854	3.28618E-06
500	0.00427	0.003327	0.005406	0.028393	2.53874E-06
600	0.005351	0.00396	0.006731	0.034702	3.76738E-06
800	0.005301	0.004046	0.006877	0.039027	6.0686E-06
1000	0.005353	0.004245	0.007206	0.044926	8.85729E-06
1200	0.004879	0.004055	0.006865	0.04711	7.05511E-06
1400	0.006058	0.005027	0.008447	0.05642	3.33179E-06
1600	0.004863	0.004415	0.007374	0.0533	5.25758E-06
2000	0.006832	0.006104	0.010112	0.069954	1.66692E-05
5000	0.005793	0.007266	0.012312	0.099682	6.72505E-06
10000	0.005771	0.010098	0.022193	0.176897	4.89748E-05

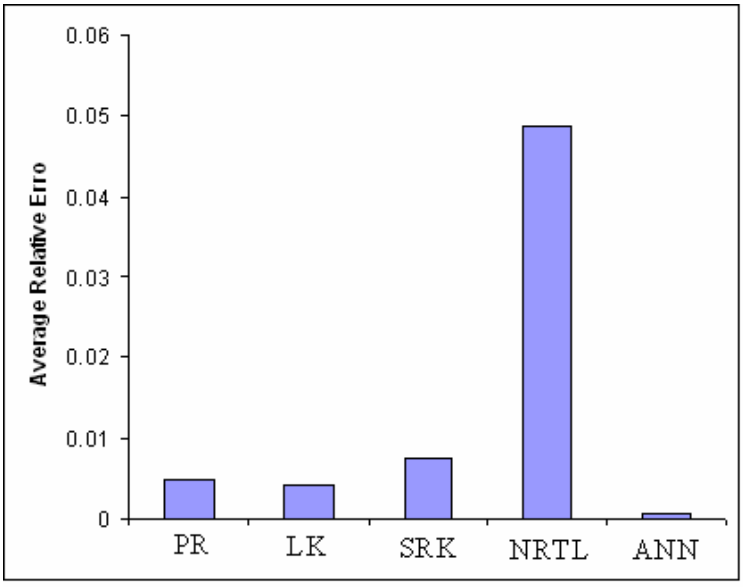

Figure-21. A comparison between thermodynamic models and ANN performance by using average relative error.

7. CONCLUSIONS

The ability of ANN with MLP neural network for modeling and prediction of Ammonia properties have been investigated. The MSE analysis based results are used for verification of the suggested approach. Results show, a good agreement between experimental data and the predicted by ANN. An important feature of the model is it doesn't require any theoretical knowledge or human experience during the training process. This work clearly shows the ability of ANN for the calculation of the thermodynamic properties of Ammonia only based on the experimental data, instead of using equations of state.

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