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Predicting the Hydrate Formation Temperature by a New Correlation and Neural Network

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Abstract: *Gas hydrates are a costly problem when they plug oil and gas pipelines. The best way to determine the HFT and pressure is to measure these conditions experimentally for every gas system. Since this is not practical in terms of time and money, correlations are the other alternative tools. There are a small number of correlations for specific gravity method to predict the hydrate formation. As the hydrate formation temperature is a function of pressure and gas gravity, an empirical correlation is presented for predicting the hydrate formation temperature. In order to obtain a new proposed correlation, 356 experimental data points have been collected from gas-gravity curves. This correlation is programmed and assessed with respect to its capabilities to match experimental data published in the literature under varying system conditions (i.e. temperature, pressure, and composition).The SPSS software has been employed for statistical analysis of the data. In order to establish a method to predict the hydrate formation temperature, a new neural network has also been developed with the BP(Back Propagation) method. This neural network model enables the user to accurately predict hydrate formation conditions for a given gas mixture, without having to do costly experimental measurements.*

Keywords: *Hydrate Formation Temperature, AUT Correlation, Artificial Neural Network*

1. Introduction

Gas hydrates (gas clathrates) are solid compounds of natural gas molecules encaged within a crystal structure consisting of water molecules. Hydrates can form anywhere and anytime that hydrocarbons and water are present at low temperatures and high pressures (Sloan, 2000). The

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formation of gas hydrates during hydrocarbon production and transportation is a serious problem in the petroleum industry.

Typical hydrate-forming guests include CH_4, C_2H_6, C_3H_8, i-C_4H_{10}, CO_2, H_2S, $CHCL_3$ and the noble gases (Sloan, 2008). Gas hydrate formation is a concomitant process requiring the presence of both the host and guest molecular species. Conditions under which hydrates will form are determined largely by the nature of the guest, but the most common compounds of natural gas will crystallize at temperatures above the ice point with pressures nearly 10 atm. Such conditions are also common in oil and gas transmission hence gas hydrate formation is a major possible cause of pipeline occlusion. The existence of clathrate hydrate was first documented by Sir Humphery Davy in 1810, who observed that a solution of chlorine gas in water freezes more readily than pure water. Since 1934, when Hummerschmidt concluded that natural gas hydrates were blocking gas transmission lines, the susceptibility of forming solid hydrates in gas transmission under normal operating conditions has led to many investigations aimed at understanding and avoiding hydrate formation, an area of ongoing research (Sloan, 2008).

The first model for predicting hydrate formation conditions is the K-Value method (Wilcox et al, 1941) which utilizes the vapor-solid equilibrium constants for prediction. The hydrate forming conditions are predicted from empirically estimated Vapor-solid equilibrium constants given by:

$$K = \frac{y_i}{x_i} \tag{1}$$

Where y_i is the mole fraction of the i^{th} hydrocarbon component in the gas phase on a water-free basis and x_i is the mole fraction of the same component in the solid phase on a water-free basis. The hydrate formation conditions should satisfy

$$\sum_{i=1}^{n} \frac{y_i}{x_i} = 1 \tag{2}$$

The second model was developed by Katz (Katz, 1945) known as the gas gravity method. The Katz's plots relate the hydrate formation pressure and temperature with gas gravity.

There are a few numbers of correlations in order to calculate the hydrate formation conditions. Common correlations to calculate Hydrate Formation Temperature (HFT) are as below:

1. Kobayashi et al Correlation (Kobayashi et al, 1987).

In 1978 Kobayashi et al presented a correlation to predict HFT based on gas gravity curves where

T is given as follows:

$$\begin{aligned} T=1/[&A_1+A_2(\ln\gamma_g)+A_3(\ln P)+A_4(\ln\gamma_g)^2+A_5(\ln\gamma_g)(\ln P)+A_6(\ln P)^2 \\ &+A_7(\ln\gamma_g)^3+A_8(\ln\gamma_g)^2(\ln P)+A_9(\ln\gamma_g)(\ln P)^2+A_{10}(\ln P)^3+A_{11}(\ln\gamma_g)^4 \\ &+A_{12}(\ln\gamma_g)^3(\ln P)+A_{13}(\ln\gamma_g)^2(\ln P)^2+A_{14}(\ln\gamma_g)(\ln P)^3+A_{15}(\ln P)^4] \end{aligned} \tag{3}$$

2. Towler and Mokhatab correlation (Carroll, 2009). In their correlation T is given by:

$$T=A_1\ln(P)+A_2\ln(\gamma_g)-A_3\ln(P)\ln(\gamma_g)-A_4 \quad (4)$$

3. Motiee correlation (Motiee, 1991). Equation 5 provides a relationship to estimate T as a function of pressure and gas gravity:

$$T = b_1+b_2\log (P) +b_3 (\log (P))^2+b_4\gamma_g+b_5 \gamma_g^2+b_6 \gamma_g (\log (P)) \quad (5)$$

4. Hammerschmidt correlation (Hammerschmidt, 1934).
This correlation describes T as a function of pressure:

$$T = 8.9\times P^{0.285} \quad (6)$$

Van der Waals and Platteeuw derived the basic statistical thermodynamic equations for gas hydrates (Van der Waals and Platteeuw, 1959). It has been modified by many investigators. Parrish and Prausnitz first generalized this model for prediction of the conditions under which hydrates form by using the Kihara potential with modified Kihara parameters (Parrish and Prausnitz, 1972). Chen et al developed a hydrate model based on the proposed two-step hydrate formation mechanism, in which the evaluation of the thermodynamic properties of the unstable empty hydrate lattice is not required (Chen and Guo, 1998). Qing et al proposed a model in which the PT Equation of State was coupled with the hydrate model proposed by Chen and Gue. It was then applied to predict the hydrate formation conditions of various systems containing polar inhibitors (Qing et al, 2003). Bahadori and Vuthaluru formulated a novel empirical correlation for rapid estimation of hydrate formation condition of sweet gases (Bahadori and Vuthaluru, 2009). Ameripour and Barrufet proposed two new correlations that can calculate the hydrate formation temperature or pressure (Ameripour and Barrufet, 2009). The main objectives of the present study are to develop a new correlation for predicting the HFT for natural gases with various gravities as well as its prediction using the IPS (Intelligent Proxy Simulator) neural network model. At last, the IPS model will be compared with our correlation and experimental data.

2. Artificial neural networks

Artificial neural networks represent a type of computation that is based on the way the brain performs computations. Neural networks are good at fitting non-linear functions and recognizing patterns. One of the well- known types of neural networks is the MLP (Multi Layer Perceptron) which is utilized to classify and estimate neural problems. This type of network contains a layer of input neurons, a layer of output neurons, and one or several hidden layers of intermediate neurons. The training algorithm used in this work is the back propagation method. The back-propagation neural network is applicable to a wide variety of problems and is considered to be the predominant supervised training algorithm. The term ‘back-propagation’ indicates the method by which the network is trained, and the term ‘supervised learning’ implies that the network is trained through a set of available input–output patterns. The network is repeatedly exposed to these input–output relationships and the weights among the neurons are continuously adjusted until the network ‘learns’ the correct input–output behavior. Initially, all the weights are set randomly and the difference or error between the desired and calculated outputs is calculated. This error is propagated back through the network and is iteratively used to update the weights among neurons. This update process is repeated for all patterns many times until the network becomes capable of reproducing the input–output relationships within an acceptably small tolerance (Elgibal and Elkamel, 1998).

In this case, the training algorithm is back propagation algorithm and among 356 data, the network is being taught by 300 data and the 56 remaining data is used to test the generalization capacity of the network. Neural network variables contain temperature (°F), pressure (psi) and specific gravity of gas, the domains of which are illustrated in Table 1. The optimal number for hidden layers is 3 and each layer contains 6-4-6 neurons, respectively. The network image is illustrated in Figure 1.

Table 1. The domains and Domain

Variable	Minimum	Maximum
Temperature (°F)	31.95	78.8
Pressure (Psi)	50.98	3874.1
Specific gravity	0.6	1.0

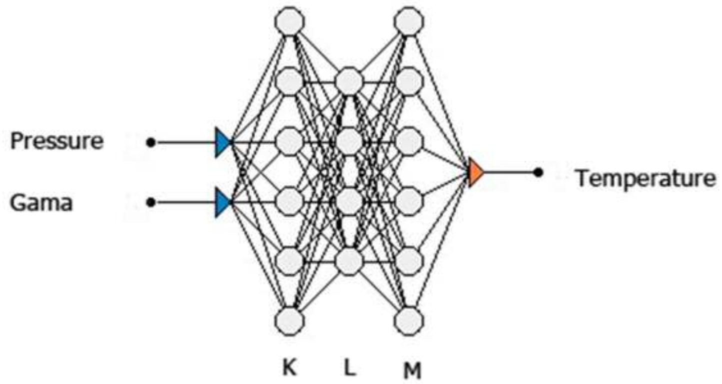

Fig. 1. The network image (Pressure and gama are input layers; K, L and M are hidden layers and Temperature is output layer)

3. AUT correlation

In this research, 356 data points were gathered at the three-phase equilibrium from gas–gravity curves (Katz, 1945) The SPSS software was used as a regression model for 300 experimental data points in order to find the best correlation among variables. The correlation (Equation7) has 15 unknown parameters. In order to obtain these parameters, the SPSS software has been employed. The following equation is the result of regression which predicts the hydrate formation temperature when pressure and gravity are given.

$$\begin{aligned}T = {} & -A_1+A_2(\ln P)+A_3(\ln\gamma_g)+A_4(P)(\gamma_g)^2+A_5(P)^2(\gamma_g)^3-A_6(\ln P)(\ln\gamma_g) \\ & -A_7(P)(\gamma_g)-A_8(P)^6(\gamma_g)^8+A_9(\ln P)^5+A_{10}(\ln\gamma_g)^7+A_{11}(P)^8(\gamma_g)^8 \\ & -A_{12}(P)^8(\gamma_g)^7+A_{13}(\ln P)^6(\ln\gamma_g)^8-A_{14}(\ln P)^7(\ln\gamma_g)^4+A_{15}(\ln P)(\ln\gamma_g)^3\end{aligned} \quad (7)$$

Where T is the temperature of the system in °F, P is the pressure of the system in psia and $A_1 \dots A_{15}$ are obtained from Table 2. In the second step, 56 unseen data were employed for validating the equation. Corresponding coefficient (R^2) for the equation and parameters values of correlation have been obtained in Table 2. To check the accuracy of the correlation and to compare the predicted results with the experimental data, a statistical error analysis was applied.

Table 2. Statistical results and value of parameters for the AUT correlation

Coefficient	Value	Std,error	Coefficient	Value	Std,error
A1	-20.928	0.245	A9	0.001	1.03E-04
A2	13.623	0.059	A10	281.743	19.122
A3	29.67	1.342	A11	1.25E-27	1.78E-04
A4	-0.006	1.02E-04	A12	-7.24E-28	1.12E-04
A5	4.14E-06	2.10E-04	A13	0.002	1.40E-04
A6	-0.979	0.308	A14	-1.84E-05	3.02E-04
A7	-0.19	1.09E-03	A15	0.792	0.345
A8	-1.25E-20	1.41E-04			

No.point = 300 MSE=1.9017084E-2 R^2 = 0.992

4. Results and discussion

The most common type of hydrate data taken are temperatures and pressures; which are of great importance in natural gas applications. Most available experimental data of this type has been compiled and compared with predictions from AUT correlation and IPS neural network. Several neural networks architectures have been investigated to achieve the highest accuracy. Three hidden layers network, with 16 neurons in the hidden layers showed reasonably accurate results for this model. This section deals with the HFT prediction in different pressures and special gravities using the IPS neural network and the presented model. The Results of the comparison with experimental data are illustrated in Figures 2 to 11.

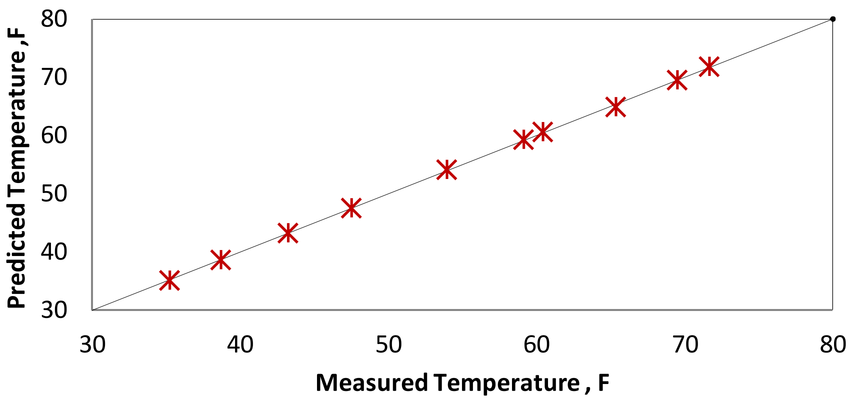

Fig. 2. Cross plot of the AUT Correlation for Specific gravity = 0.6

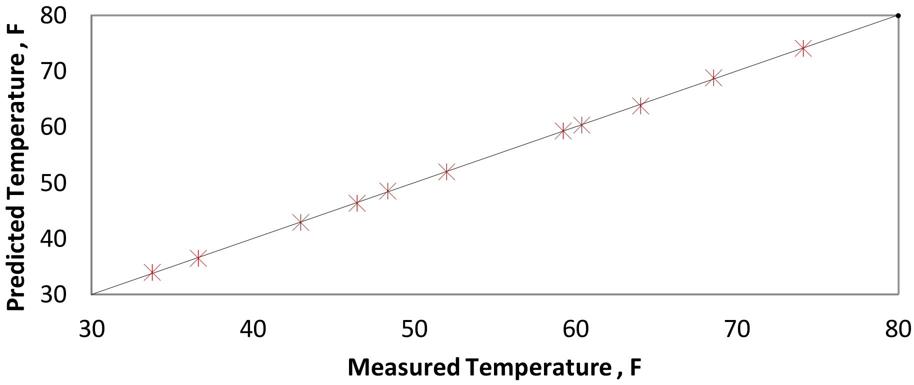

Fig. 3. Cross plot of the AUT Correlation for Specific gravity = 0.7

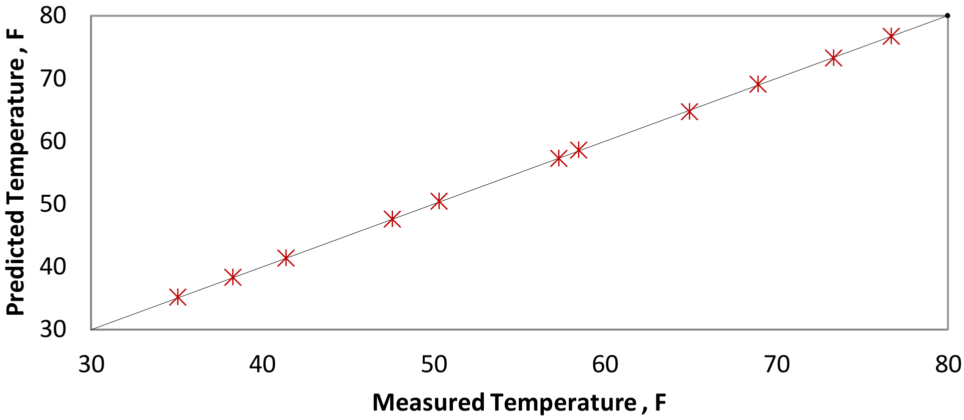

Fig. 4. Cross plot of the AUT Correlation for Specific gravity = 0.8

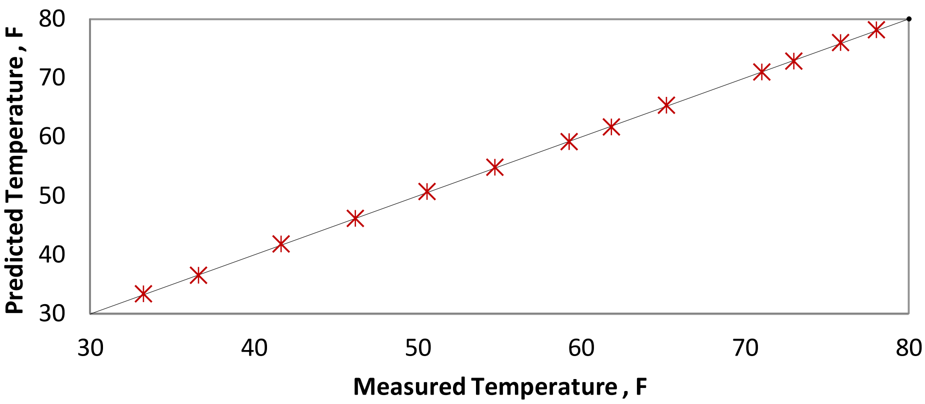

Fig. 5. Cross plot of the AUT Correlation for Specific gravity = 0.9

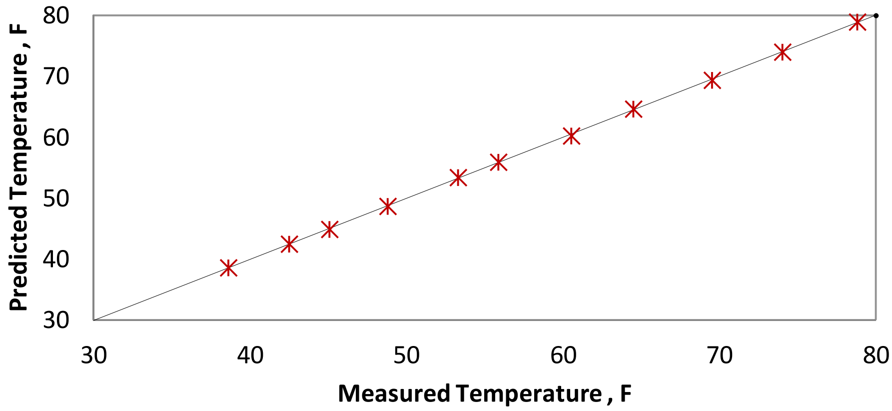

Fig. 6. Cross plot of the AUT Correlation for Specific gravity = 1.0

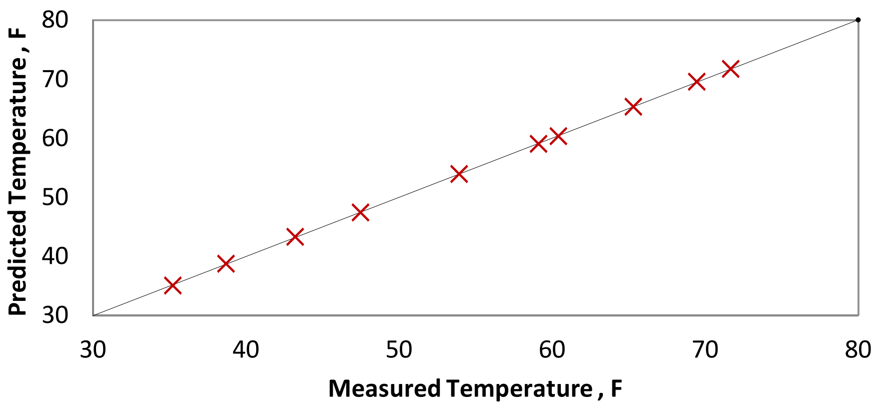

Fig. 7. Cross plot of the ANN (IPS) for Specific gravity = 0.6

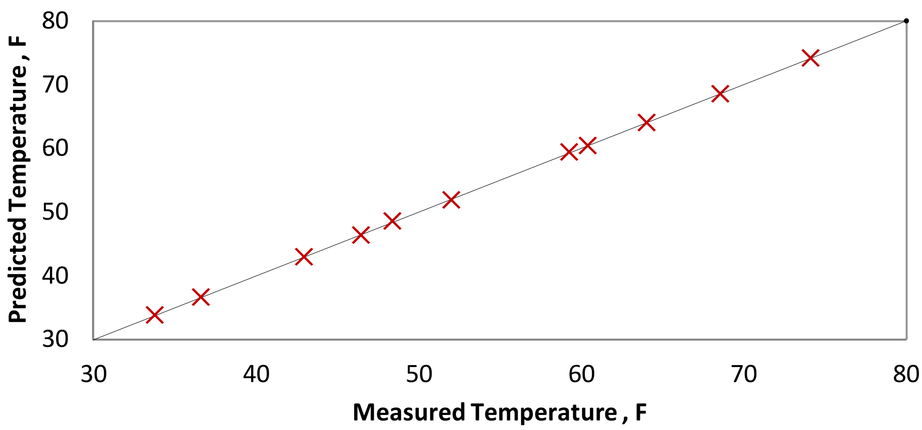

Fig. 8. Cross plot of the ANN (IPS) for Specific gravity = 0.7

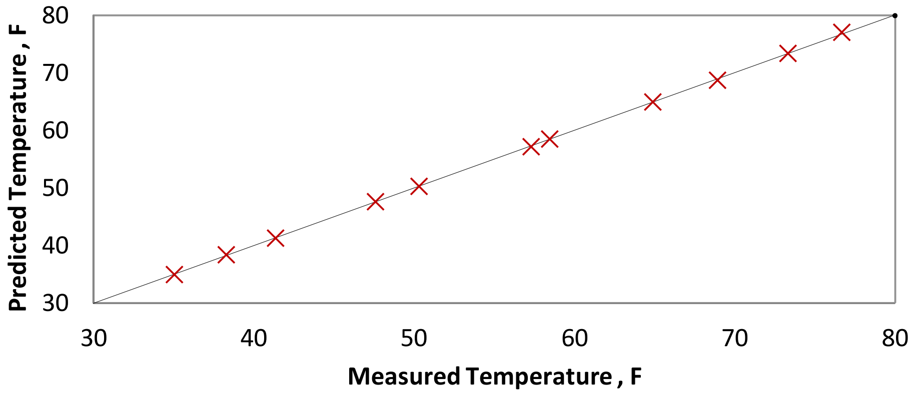

Fig. 9. Cross plot of the ANN (IPS) for Specific gravity = 0.8

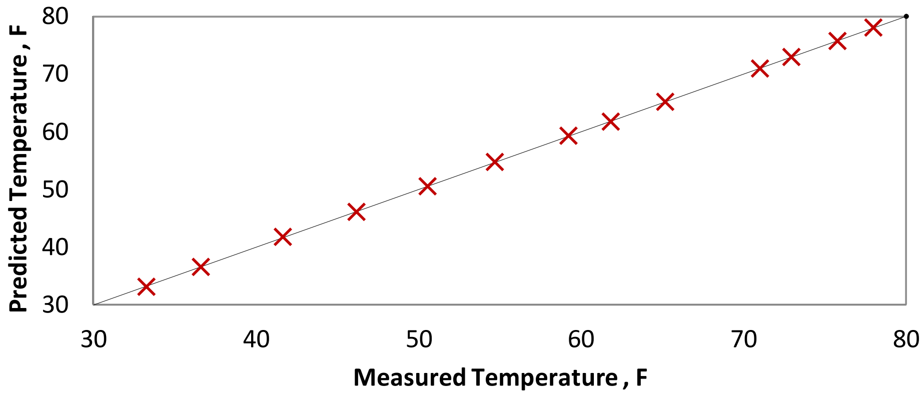

Fig. 10. Cross plot of the ANN (IPS) for Specific gravity = 0.9

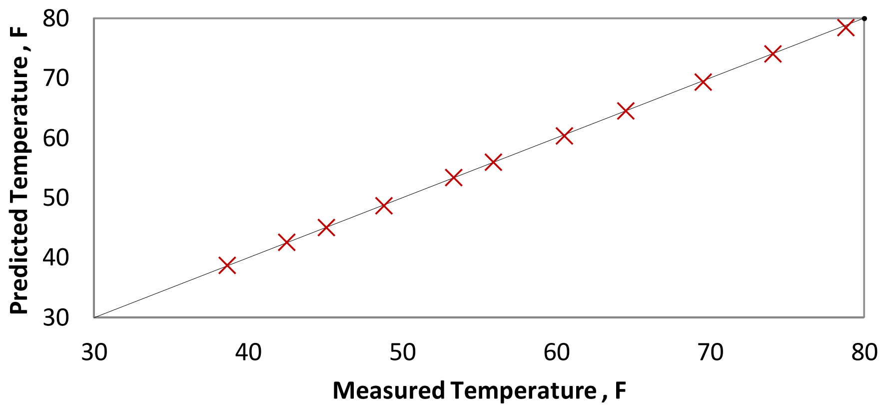

Fig. 11. Cross plot of the ANN (IPS) for Specific gravity = 1.0

The accuracy of the correlation and IPS neural network relative to the experimental data is determined by various statistical means including ARE (Average Absolute Relative Error) and MSE(Mean Square Error).

ARE is defined as: $$E_i = \left| \frac{x_{\exp} - x_{pre}}{x_{\exp}} \right| \times 100$$

$$ARE = \frac{1}{n} \sum_{i=1}^{n} | E_i |$$

MSE is defined as: $$MSE = \frac{1}{n} \sum_{i=1}^{n} (x_{\exp} - x_{pre})^2$$

Where x_{exp}, x_{pre} and n are actual output, predicted output and number of data, respectively. **Table 3** represents MSE and ARE errors resulted from the comparison of experimental data with the IPS neural network and AUT correlation. According to this table, the predicted temperatures by IPS and AUT correlation are in pretty good agreement with experimental data.

Table 3. Error Analysis of IPS neural network and present correlation

	ANN(IPS)	AUT
MSE	0.01069	0.0186690
Min ARE	8.80E-05	0.0001398
Max ARE	0.0045666	0.0059498
Mean ARE	0.0014638	0.0020730

5. Conclusion

In this work, a new correlation for hydrate formation temperature is presented by which HFT is described as a function of pressure and gas gravity. The AUT correlation is in perfect accord with experimental data and can be used for prediction of HFT. In the next step of this study, the ANN model (IPS) has been built for HFT estimation. The ANN model and AUT correlation prediction are compared with experimental data. Based on the results, the MSE and ARE analyses are adopted to verify the suggested approach. The results demonstrate a good agreement between experimental data with AUT correlation and ANN predictions.

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