WALAILAK JOURNAL
http://wjst.wu.ac.th Engineering and Physical Sciences

Development of a Neural Fuzzy System for Advanced Prediction of Gas Hydrate Formation Rate in Pipeline

Mohammad Javad JALALNEZHAD[1,2,*], Mohammad RANJBAR[3], Amir SARAFI[4] and Hossein NEZAMABADI-POUR[5]

[1]*Department of Petroleum Engineering, Shahid Bahonar University of Kerman, Kerman, Iran*
[2]*Young Researchers Society, Shahid Bahonar University of Kerman, Kerman, Iran*
[3]*Department of Mining Engineering, Shahid Bahonar University of Kerman, Kerman, Iran*
[4]*Department of Chemical Engineering, Shahid Bahonar University of Kerman, Kerman, Iran*
[5]*Department of Electrical Engineering, Shahid Bahonar University of Kerman, Kerman, Iran*

([*]Corresponding author's e-mail: javad.jalalneghad@yahoo.com)

Received: 8 January 2014, Revised: 6 February 2014, Accepted: 9 July 2014

Abstract

With the development of the natural gas industry in the 20th century, the production, processing and distribution of natural gas under high-pressure conditions has become necessary. Under these conditions, it was found that the production and transmission pipelines were becoming blocked with what looked like to be ice. Hammerschmidt determined that hydrates were the cause of plugged natural gas pipelines. Gas hydrates and difficulties related to their formation in production and transmission pipelines and equipment, are the major concerns of the gas industry. The main objective of this study was to present a novel approach to access more accurate hydrate formation rate predicting models based on a combination of flow loop experimental data with learning power of adaptive neural-fuzzy inference systems and more than 900 data points of the CO_2, C_1, C_3, and i-C_4 hydrate formation rate. Using this data set different predictive models were developed. It was found that such models can be used as powerful tools, with total errors less than 6 % for the developed models, in predicting hydrate formation rate in these cases.

Keywords: Adaptive Neural-Fuzzy Inference System, gas hydrate formation, kinetic inhibitor, rate model

Introduction

Gas hydrates are ice-like crystalline solid compounds formed from water and low molecular diameter non-polar or slightly polar molecules (usually gases) under low temperature, but well above the freezing point of water, and elevated pressure conditions. Based on the crystal structure, hydrates are classified into three well-known types of structures: sI, sII and sH [1]. With the development of the natural gas industry in the 20th century, the production, processing and distribution of natural gas under high-pressure conditions were necessary. Under these conditions, it was found that the production and transmission pipelines were becoming blocked with what looked like ice. Hammerschmidt [2] determined that hydrates were the cause of the plugged natural gas pipelines. Several processes were investigated in order to prevent and/or combat hydrate plugs and ensure regular flow: chemical, hydraulic, thermal and mechanical processes. The chemical method consists in injecting chemicals in the pipeline. These chemicals fall into three classes: Thermodynamic hydrate Inhibitors (THIs), Kinetic Hydrate inhibitors (KHIs) and anti-agglomerants (AAs) [3]. Unlike the THIs, the KHIs (generally polymers) do not alter the thermodynamics of the hydrate formation but instead, modify the kinetics of hydrate formation by preventing nucleation or by hindering or slowing down the crystal growth [1].

Prediction of gas hydrate formation rate (HFR) plays an important role in developing models that can describe and predict the hydrate formation processes and also in studying the mechanisms of nucleation and growth of hydrate plugs in pipelines. Thus research has been performed concerning the measurement and modeling of hydrates formation rate based on the hydrate-former gases consumption values [4-8]. There are also some studies that investigate the effects of different inhibitors, surfactants and additives on the formation rate of different gas hydrates by using high pressure cells and flow mini-loops [9-12]. Talaghat *et al*. [11] proposed a new Rate equation to predict gas consumption rate during hydrate formation in the presence or absence of kinetic inhibitors in a flow mini-loop apparatus (the so called "Talaghat-model").

However, these models are not accurate enough to predict HFR in pipelines and often consider only simple pure gases. Most of them require complex and time consuming computations and also a lot of input information to achieve the answer.

Based on the above discussion, it is obvious that there is a need for developing new models. These models should not have the limitations and complexities of the available models. In other words the new models should be more accurate, robust and less sensitive to noisy input data, adaptive to new input-output information and also should require the least amount of input information. Intelligent models offer all of the above desirable characteristics. Therefore, the main objective of this study was to present (**A**daptive **N**etwork-Based **F**uzzy **I**nference **S**ystem) ANFIS models for predicting the HFR of common hydrate-former gases (C_1, C_3, i-C_4 and CO_2), with or without the presence of KHIs using experimental data obtained from flow mini-loop apparatus. In the next step of this study, the overall performance of the developed models was evaluated by comparison between the rate predictions of the ANFIS models, the experimental data and the rate predictions of the Talaghat-model. As far as we are aware no research paper is available in this field of study of gas hydrates.

Materials and methods

Fuzzy inference system

Fuzzy logic is a superset of conventional Boolean logic that has been extended to handle the concept of partial truth. It was first introduced by Dr. Lotfi Zadeh, a professor at the University of California at Berkley, in the 1960s as a means to model the uncertainty of natural language. Notions like rather tall or very fast can be formulated mathematically and processed by computers, in order to apply a more human-like way of thinking in the programming of computers [13].

To numerically define uncertainty, membership functions are employed. The role of a membership function is to allocate a degree of membership to each element of a fuzzy set. The degree of membership of a fuzzy set indicates the certainty or uncertainty that the element belongs to that set [14]. For fuzzy systems in general, the dynamic behavior of the system is characterized by a set of linguistic fuzzy rules. These rules are based on the knowledge and experience of a human expert within that domain. Basically, a fuzzy inference system (FIS) is composed of a knowledge base, which includes the information given by the expert in the form of linguistic fuzzy rules; a fuzzifier, which transforms the crisp inputs into degree of match with linguistic values; an inference system (engine), which uses them together with the knowledge base to make inference by means of a reasoning method; and a defuzzifier, which transforms the fuzzy results of the inference into a crisp output using a defuzzification method. The basic structure of a fuzzy inference system is shown in **Figure 1**.

There are 2 major fuzzy inference methods: the first model was proposed by Mamdani in an attempt to control a steam engine by a set of linguistic control rules obtained from experienced human operators [15]. Eq. (1) shows the general form of a Mamdani model rule, where x, y, and z are the linguistic terms;

$$\text{If X is x and Y is y then Z is z} \tag{1}$$

The Sugeno fuzzy model, which is also known as the TSK fuzzy model, was proposed by Tagaki and Sugeno [16] in an effort to develop a systematic approach to generate fuzzy rules from a given input-output data set [16]. A typical fuzzy rule in a Sugeno fuzzy model has the following structure;

$$\text{if x is A and y is B then Z} = f(x, y) \tag{2}$$

The main difference between these 2 methods is that Mamdani used fuzzy sets as the rule consequent, whereas Sugeno employed linear functions of input variables as the rule consequent [17]. In this article, both types of inference models are employed. Generally, there are 2 different approaches to construct the knowledge base of a fuzzy inference system. The first method is based on the knowledge of a human expert about the problem, which helps the expert to initially select the membership functions and rules. The second approach is to tune the membership function parameters using evolutionary algorithms or neural nets. The latter, which was first introduced by Jang [18,19] is called an adaptive network-based fuzzy inference system (ANFIS).

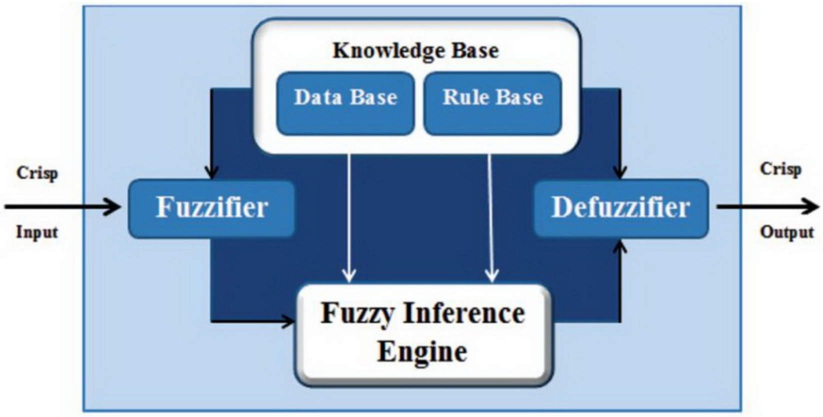

Figure 1 Basic structure of a fuzzy inference system.

Adaptive network-based fuzzy inference system

An adaptive network is a multilayer feed forward network in which each node performs a particular function (node function) on incoming signals as well as a set of parameters pertaining to this node [18]. Like an ANN, ANFIS is capable of mapping unseen inputs to their outputs by learning the rules from the previously seen data. A simple structure of this type of network having just 2 inputs, x and y, and one output, f is shown in **Figure 2**. ANFIS contains five layers in its architecture, including the fuzzy layer, product layer, normalized layer, defuzzification layer, and total output layer. It should be noted that assuming just 2 membership functions for each of the input data x and y, the general form of a first-order TSK type of fuzzy if-then rule would be;

If x is A_i and y is B_i THEN $f_i = p_i x + q_i y + r_i$, I = 1,2,..,n (3)

where n is the number of rules and p_i, q_i, and r_i are the parameters that are determined during the training process. Through the learning process, at the first stage, the membership degree (μ) of each of the linguistic labels A_i and B_i is calculated;

$$O_i^1=\mu_{Ai}(x),\ i=1,2,..,n \quad (4)$$

$$O_i^1=\mu_{Bi}(x),\ i=1,2,..,n \quad (5)$$

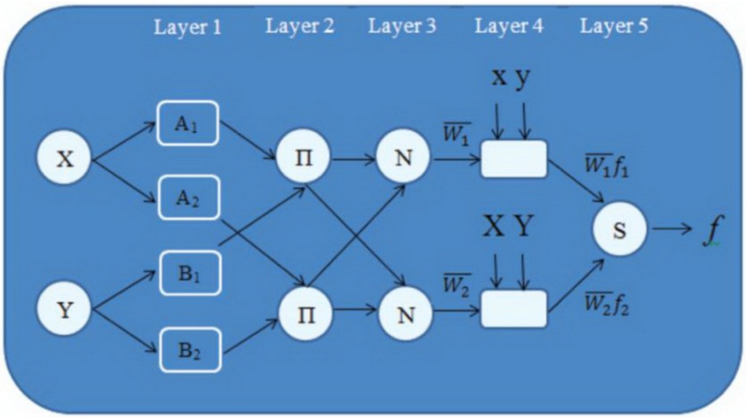

Figure 2 ANFIS structure.

Then, at the second layer, which is the product layer, the previously calculated membership degrees of linguistic variables are multiplied;

$$O_i^2 = W_i = \mu_{Ai}(x)\, \mu_{Bi}(y)\ , \quad i=1,2,\ldots,n \tag{6}$$

The third layer is the normalized layer, in which the ratio of each weight to the total weights is calculated;

$$O_i^3 = \overline{w}_i = \frac{w_i}{\sum_{i=1}^{n} w_i}\ , \quad i=1,2,..,n \tag{7}$$

The fourth layer is the defuzzification layer with adaptive nodes, which means that their outputs depend on the parameter(s) pertaining to these nodes and the learning rule specifies how these parameters should be changed to minimize a prescribed error measure [18]. The relationship for these nodes is as follows;

$$O_i^4 = \overline{w}_i f_i = \overline{w}_i(p_i x + q_i y + r_i)\ , \quad i=1,2,..,n \tag{8}$$

Finally, in the fifth layer, the summation of all of the incoming signals is performed where the result would be the output of the system;

$$O_i^5 = \sum_{i=1}^{n} \overline{w}_i f_i\ , \quad i=1,2,..,n \tag{9}$$

Data acquisition and processing

In this study, there are 946 data points to measure gas hydrate formation rate. Available data have been extracted and collected from articles which investigate the phenomenon of gas hydrate formation in laboratory-scale [11]. Using a linear transformation first, data is normalized between (0 1), in order to data rate reduction, noise suppression and avoiding ill conditioning;

$$V_{norm} = (v - x_{min}) / (x_{max} - x_{min}) \tag{10}$$

where v is a current value of the variable, x_{min} is the minimum value for this variable, and, x_{max} is the maximum value for that variable x in the data set.

Fuzzy models input data

To develop a Fuzzy model, the most important physical skill required is to make a decision as to what the principal inputs and output(s) of the system are. In this study, the inputs to the Fuzzy models were temperature, pressure, molecular weight of hydrate-former, time and concentrations of the KHIs. The desirable output of the models was the hydrate formation rate (gas consumption amount). With this motivation in mind 2 Fuzzy models, named HPNFS1 and HPNFS2 (**H**ydrate-formation-rate **P**rediction using **N**eural **F**uzzy **S**ystem) were considered. In the first model, HPNFS1, a system without the presence of the KHIs was considered. In this model, HFR was a function of temperature (T), pressure (P), molecular weight of hydrate-former (MW_{hf}) and time(t);

$$\text{HFR} = f_{\text{HPNFS1}}\,(\text{T, P}, MW_{hf}, \text{t}) \tag{11}$$

The model HPNFS2 was a HFR predicting model with six input variables. In this model the concentrations of 2 KHIs ($PVP_{CONC}, L - Tyrosine_{CONC}$) were added to the set of the input variables. So, the HPNFS2 model was expressed in the form of;

$$\text{HFR} = f_{\text{HPNFS2}}(\text{T, P}, MW_{hf}, \text{t}, PVP_{CONC}, L - Tyrosine_{CONC}) \tag{12}$$

Neural -fuzzy modeling

An effective method developed by Dr. Roger Jang (Fuzzy logic toolbox, 1995) for neuro-fuzzy modeling is called ANFIS (Adaptive Neuro-Fuzzy Inference System), which has been used in this study. The fuzzy HFR modeling systems used in this study is a multi-input single output (MISO) Takagi-Sugeno system. The first available data in the presence and absence of kinetic inhibitors were 467 and 479 respectively which were divided into 2 parts of training data with the number of 350 and 359 and testing data with the number of 120 and 117. Choosing this configuration was done based on a trial and error procedure to achieve best results. Because of the large number of input variables, scatter partitioning was used to avoid the “curse of dimensionality” problem instead of grid partitioning.

The HPNFS1 model was developed to predict the HFR of C_1, C_3, i-C_4 and CO_2 gases. To develop this model 467 data (**Table 1**) was used and with a random selection, 350 of the data was used as train set data and the remaining 117 data was used as test set data. The HPNFS2 model was developed to take into consideration the effect of the KHIs ($PVP_{CONC}, L - Tyrosine_{CONC}$) on simple gas hydrate formation of the mentioned gases in the absence of the KHIs. To develop this model 479 data (**Table 2**) was used and with a random selection, 359 of data was used as train set data and the remaining 120 data was used as test set data. **Table 3** shows the details of the optimal fuzzy model designed for the models, HPNFS1 and HPNFS2. This arrangement resulted by a trial and error procedure. The best parameters of obtained fuzzy clustering designed for models of HPNFS1 and HPNFS2 are respectively shown in **Tables 4** and **5**. Hybrid optimization methods were used to optimize generated fuzzy inference systems (FIS) and the best models were selected according to minimum total average absolute deviation percent (TAAD%). The structure of the designed models for HPNFS1 and HPNFS2 are respectively shown in **Figures 3** and **4**.

Table 1 Ranges of the input variables used in developing the HPNFS1 model.

Parameter	Hydrate-Former	Minimum	Maximum
Temperature (K)	C_1, C_3	277.15	277.15
	i-C_4	275.15	275.15
	CO_2	280.15	280.15
Pressure (MPa)	C_1	5	8
	C_3	1	2
	i-C_4	1	2
	CO_2	4	7
Molecular Weight (gr/mol)	C_1,C_3, i-C_4, CO_2	16.043 (C_1)	58.123 (i-C_4)
Time (min)	C_1,C_3, i-C_4, CO_2	0	185

Table 2 Ranges of the input variables used in developing the HPNFS2 model.

Parameter	Hydrate-Former	Minimum	Maximum
Temperature (K)	C_1, C_3	277.15	277.15
	i-C_4	275.15	275.15
	CO_2	280.15	280.15
Pressure (MPa)	C_1	5	8
	C_3	1	2
	i-C_4	1	2
	CO_2	4	7
Molecular Weight (gr/mol)	C_1,C_3, i-C_4, CO_2	16.043 (C_1)	58.123 (i-C_4)
Time (min)	C_1,C_3, i-C_4, CO_2	0[a]	185[a]
		0[b]	485[b]
PVP concentration (ppm)	C_1,C_3, i-C_4, CO_2	0	200
L-Tyrosin concentration (ppm)	C_1,C_3, i-C_4, CO_2	0	200

[a] In the absence of KHIs
[b] In the presence of KHIs

Table 3 Characteristics of fuzzy model for models of HPNFS1 and HPNFS2.

Parameter	Operator
AND	prod
OR	probor
Implication	prod
Aggregation	max
Difuzzification	wtaver

Table 4 The best parameters set parameters for the ANFIS (Genfis2) in the absence of inhibitors Kinetic.

Parameter	Value
Range of influence	0.23
Squash factor	1.25
Squash factor	0.5
Reject ratio	0.15

Table 5 The best parameters set parameters for the ANFIS (Genfis2) in the presence of inhibitors Kinetic.

Parameter	Value
Range of influence	0.18
Squash factor	1.25
Squash factor	0.5
Reject ratio	0.15

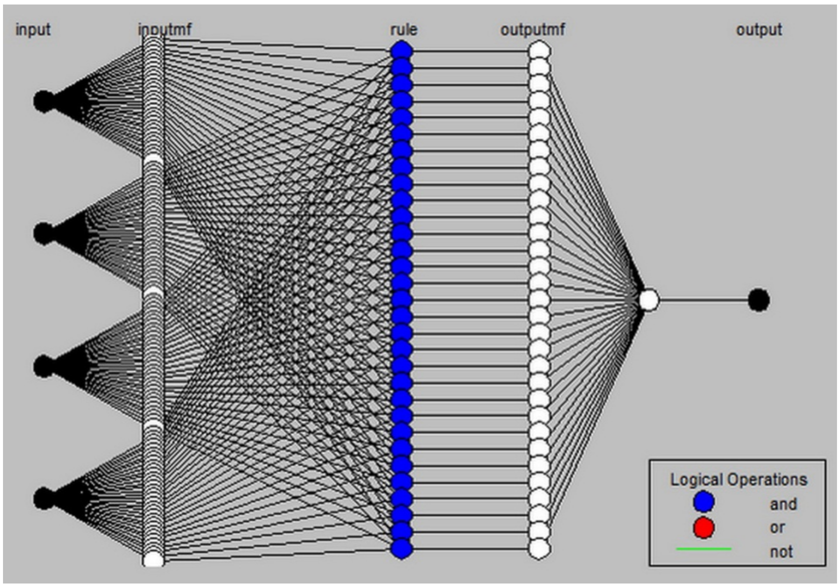

Figure 3 ANFIS model structure of the HFR prediction in the absence of kinetic inhibitors.

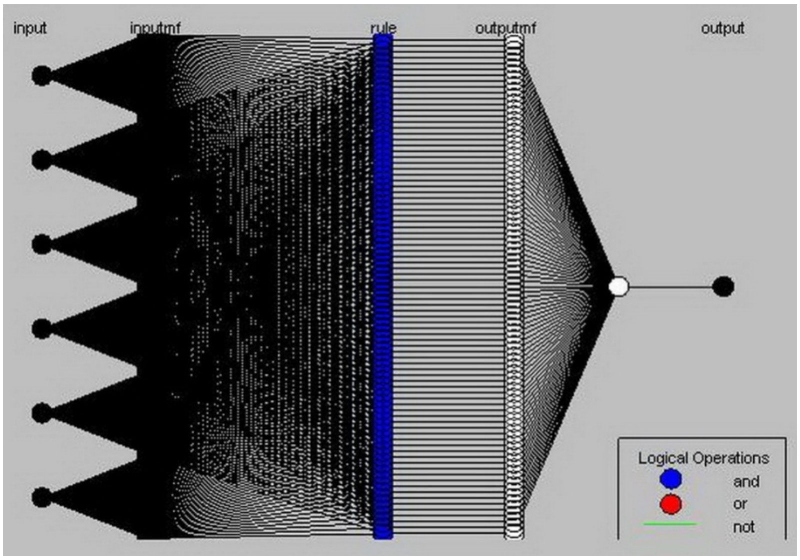

Figure 4 ANFIS model structure of the HFR prediction in the presence of inhibitors.

Results and discussion

Figures 5 to **8** show the results of the testing model for HPNFS1, along with the Talaghat experimental model compared with the experimental results in this study. Moreover, 4 different types of fuzz component gas hydrates, including CO_2, C_1, C_3, and i-C_4 at different pressures are illustrated. **Figures 9** and **13** show the results of the testing model for HPNFS2, along with the Talaghat experimental model compared with the experimental results in this study. In addition, 4 different types of component gas hydrates, including CO_2, C_1, C_3, and i-C_4 at different pressures are shown.

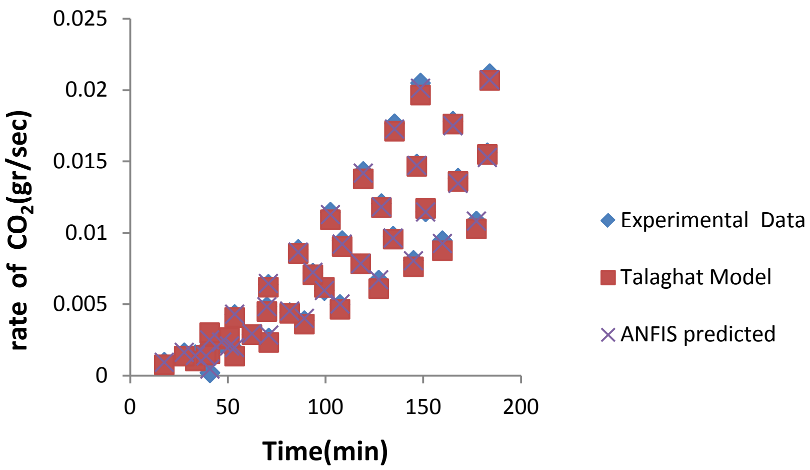

Figure 5 Results of testing the HPNFS1 model for the rate of CO_2 hydrate formation as a function of time at 280.15 K.

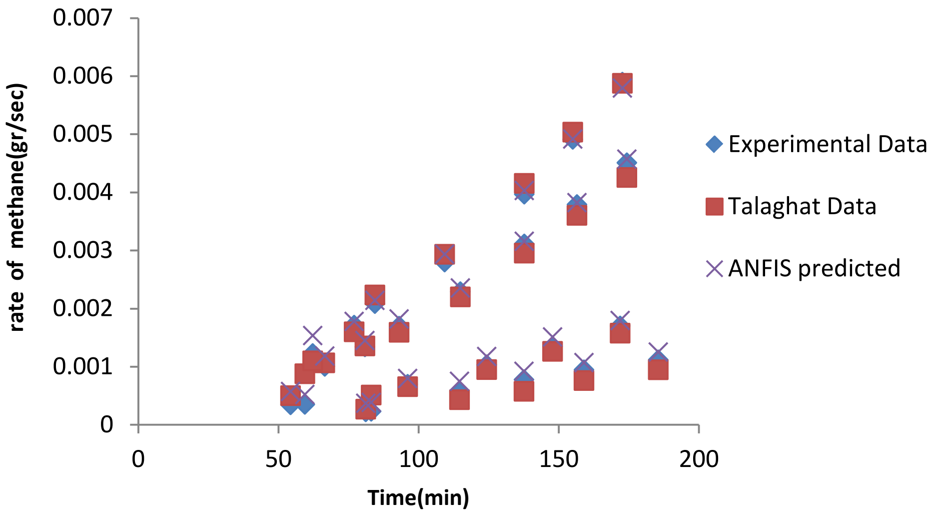

Figure 6 Results of testing the HPNFS1 model for the rate of methane hydrate formation as a function of time at 277.15 K.

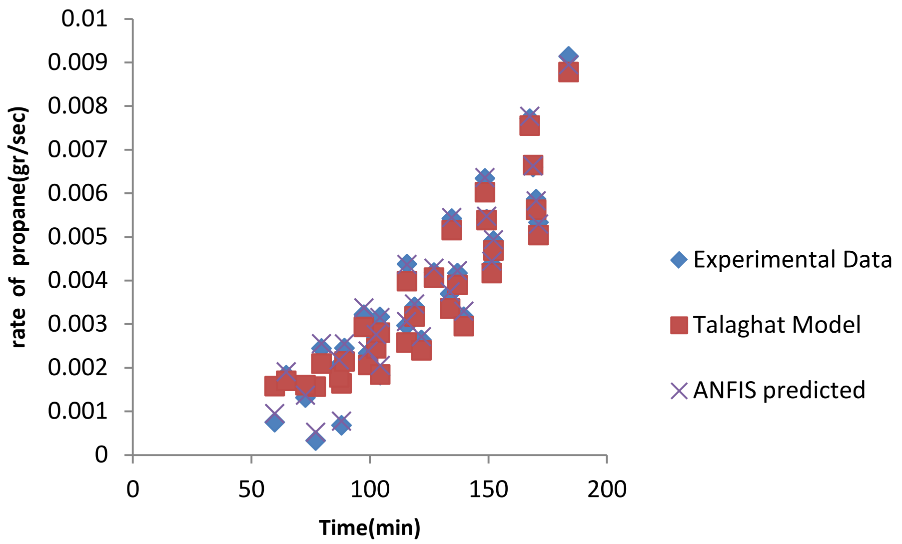

Figure 7 Results of testing the HPNFS1 model for the rate of propane hydrate formation as a function of time at 277.15 K.

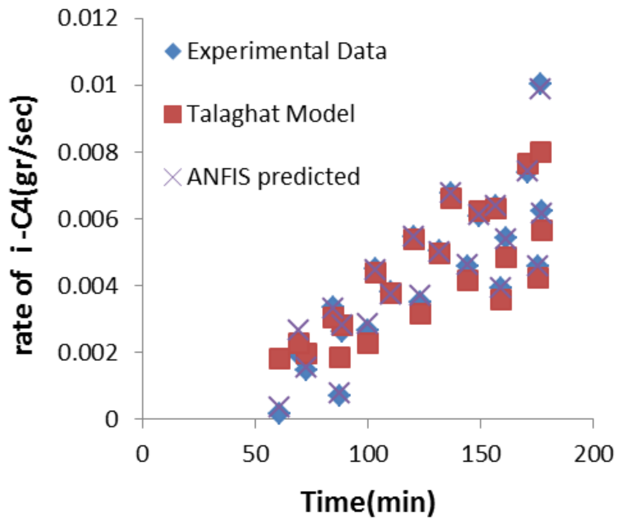

Figure 8 Results of testing the HPNFS1 model for the rate of iso-butane hydrate formation as a function of time at 275.15 K.

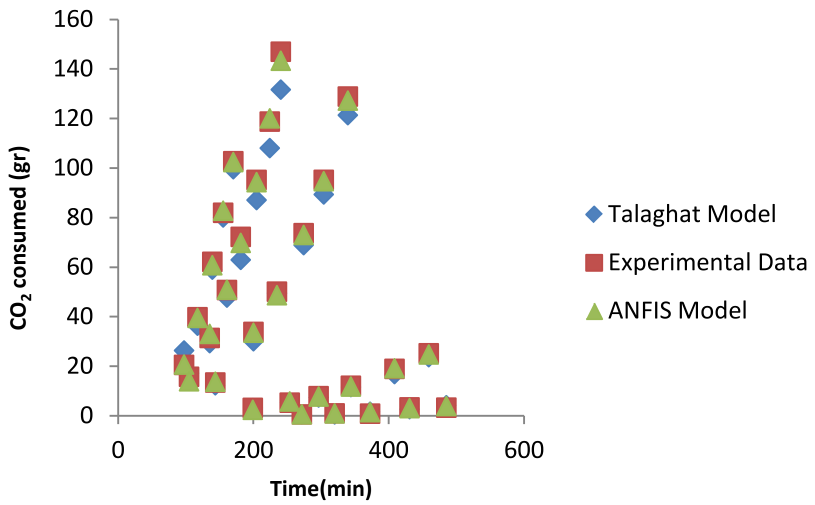

Figure 9 Results of testing the HPNFS2 model for CO_2consumed during hydrate formation as a function of time at 280.15 K and 4 MPa.

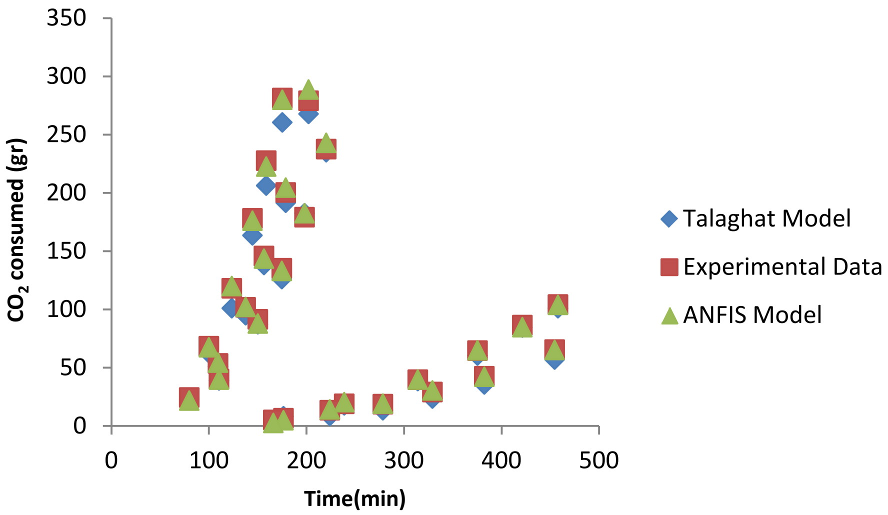

Figure 10 Results of testing the HPNFS2 model for CO_2 consumed during hydrate formation as a function of time at 280.15 K and 7 MPa.

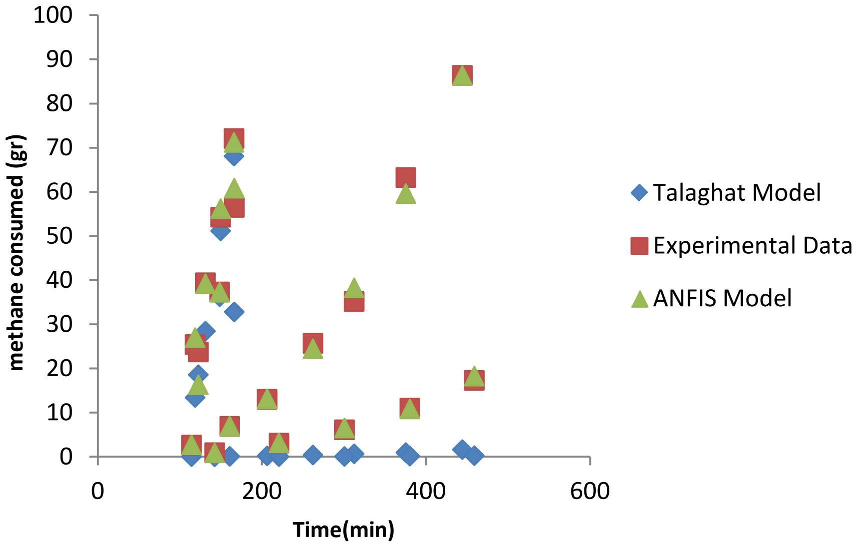

Figure 11 Results of testing the HPNFS2 model for Methane consumed during hydrate formation as a function of time at 277.15 K and 8 MPa.

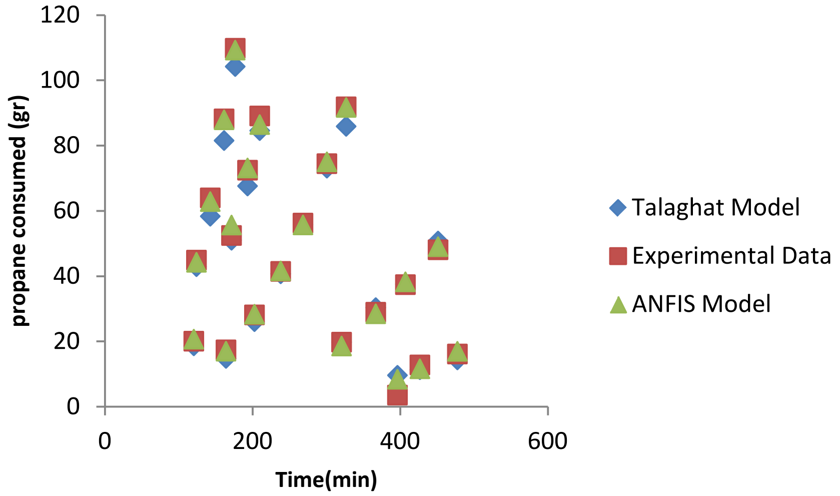

Figure 12 Results of testing the HPNFS2 model for Propane consumed during hydrate formation as a function of time at 277.15 K and 1.5 MPa.

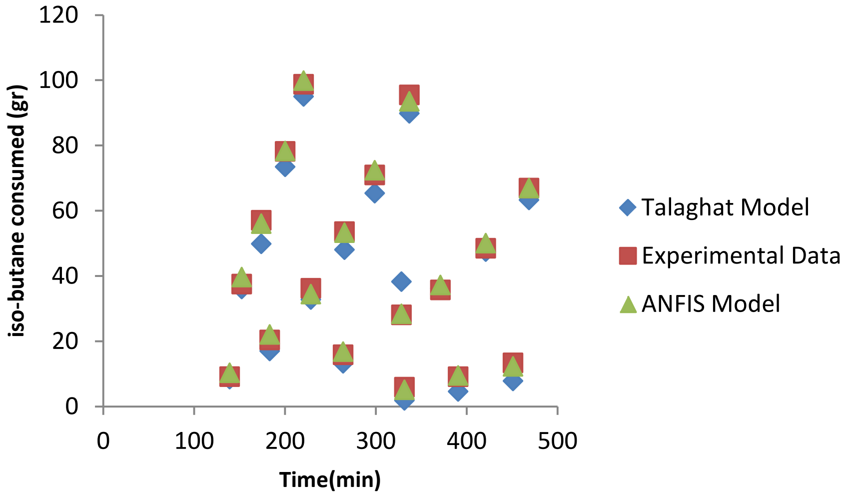

Figure 13 Results of testing the HPNFS2 model for iso-Butane consumed during hydrate formation as a function of time at 275.15 K and 2 MPa.

The performance of the model was evaluated on the basis of the total average absolute deviation percent (TAAD%) defined as;

$$\mathrm{TAAD} = \frac{100}{n} \times \left| \frac{\sum_{i=1}^{N}\left(y_i^{exp} - y_i^{cal}\right)}{y_i^{exp}} \right| \tag{6}$$

The accuracy of the presented models in comparison with the latest models of hydrate formation rate, model of Talaghat to predict gas consumption rate during hydrate formation in an experimental flow loo, is evaluated. Based on **Table 6**, the presented models for HPFNFS1, HPFNFS2 are more accurate than the Talaghat model. TAAD% is the overall average of absolute deviation for normalized data and R is the correlation coefficient for normalized data.

Table 6 Error analysis of different models.

Condition	Model	TAAD%	R
In the absence of KHIs	ANFIS-Model	3.3523	0.9998
	Talaghat-Model	14.9	0.9901
In the presence of KHIs	ANFIS-Model	5.3964	0.9994
	Talaghat-Model	15.8	0.9815

Good performance models for testing data which have no role in training models show their high universality. In **Figures 7** and **8**, actual results against outputs for the HPNFS1 and HPNFS2 models, respectively are shown moreover, they are efficient and consistent.

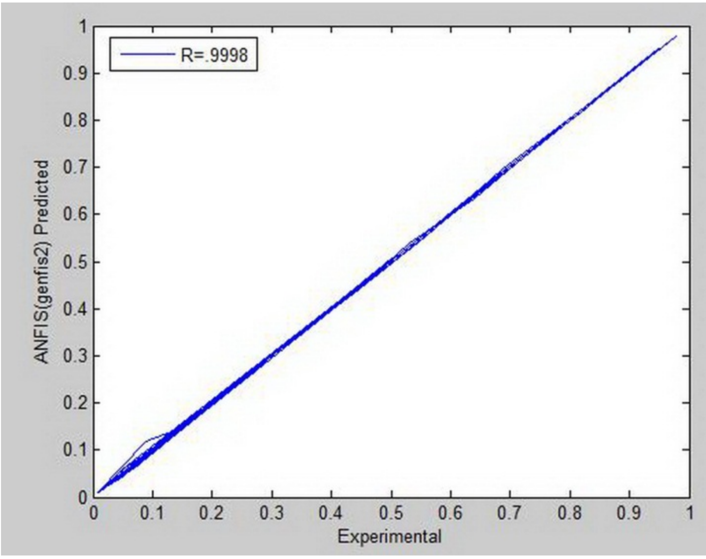

Figure 7 Experimental data versus HPNFS1 model outputs.

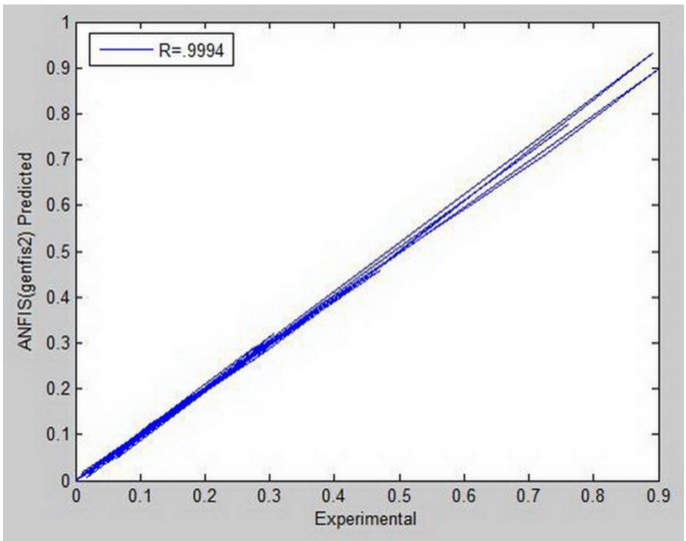

Figure 8 Experimental data versus HPNFS2 model outputs.

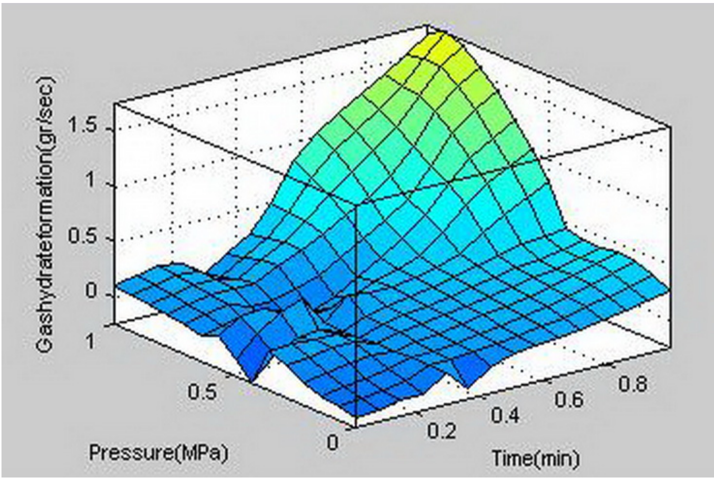

Figure 9 HPNFS2 model generated surface.

The output surfaces built for the HPNFS2 mode is shown in **Figure 9**; the first and second inputs in the figures are pressure and time respectively.

Conclusions

Gas hydrate formation in production and transmission pipelines and consequent plugging of these lines have been a major flow-assurance concern of the oil and gas industry for the last 75 years. The gas hydrate formation rate is one of the most important topics related to the kinetics of the process of gas hydrate crystallization. In this work, utilization of the neural fuzzy technique for predicting the hydrate formation rate has been investigated.

Two ANFIS (adaptive neural fuzzy system) models (HPNFS1and HPNFS2) were developed to predict the HFR in the presence or absence of KHIs (PVP and L-Tyrosine) in the pipeline of a mini-loop apparatus. Based on the results of this study, the following conclusions can be pointed out: intelligent techniques can recognize the possible patterns between input and output spaces. Combination of the explicit knowledge representation of fuzzy logic with the learning power of neural nets can alleviate the problems associated with each of these technologies. Neural fuzzy systems are data driven fundamentally. Thus, more data for training the system, better performance and more generalization will be achieved. Our comparison between experimental results, models of HPNFS1 and HPNFS2 and the Talaghat model show that the predictions of the designed models match well with experimental data so that Top of Form HPFNFS1 model is more than 4 times while the HPFNFS2 model is over 2 times more accurate than the Talaghat model. The HPNFS1and HPNFS2 models can be used to predict the hydrate formation rate of the hydrate-formers C_1, C_3, i-C_4 and CO_2 in different situations in the presence or absence of KHIs when the operational conditions conform to the ranges of the input data used to develop these models.

References

[1] ED Sloan. *Clathrate Hydrates of Natural Gases*. Marcel Dekker, New York, 1997.
[2] EG Hammerschmidt. Formation of gas hydrates in natural gas transmission lines. *Ind. Eng. Chem.* 1934; **26**, 851-5.
[3] S Mokhatab, RJ Wilkens and KJ Leontaritis. A review of strategies for solving gas-hydrate problems in subsea pipelines, recovery utilization and environmental effects. *Energ. Sourc. A* 2007; **29**, 39-45.
[4] JW Vysniauskas and PR Bishnoi. A kinetic study of methane hydrate formation. *Chem. Eng. Sci.* 1983; **38**, 1061-72.
[5] P Englezos, N Kalogerakis, PD Dholabhai and PR Bishnoi. Kinetics of formation of methane and ethane gas hydrates. *Chem. Eng. Sci.* 1987; **42**, 2647-58.
[6] P Skovborg and P Rasmussen. A mass transport limited model for the growth of methane and ethane gas hydrates. *Chem. Eng. Sci.* 1994; **49**, 1131-43.
[7] D Kashchiev and A Firoozabadi. Induction time in crystallization of gas hydrates. *J. Cryst. Growth* 2003; **250**, 499-515.
[8] R Shadravanan, M Schaffie and M Ranjbar. The prediction of hydrate formation rate in the presence of inhibitors. Prediction of hydrate formation rate in the presence of inhibitors. *Energ. Sourc. A* 2014; **36**, 661-72.
[9] CS Zhang, SS Fan, DQ Liang and KH Guo. Effect of additives on formation of natural gas hydrate. *Fuel* 2004; **83**, 2115-21.
[10] H Ganji, M Manteghian, K Sadaghianizadeh, MR Omidkhah and HR Mofrad. Effect of different surfactants on methane hydrate formation rate, stability and storage capacity. *Fuel* 2007; **86**, 434-41.
[11] MR Talaghat, F Esmaeilzadeh and J Fathikalajahi. Experimental and theoretical investigation of simple gas hydrate formation with or without presence of kinetic inhibitors in a flow mini-loop apparatus. *Fluid Phase Equil.* 2009; **279**, 28-40.
[12] MR Talaghat. Intensification of the performance of kinetic inhibitors in the presence of polyethylene oxide and polypropylene oxide for simple gas hydrate formation in a flow mini-loop apparatus. *Fluid Phase Equil.* 2010; **289**, 129-34.
[13] LA Zadeh. Making computers think like people. *IEEE Spectrum* 1984; **8**, 26-32.
[14] AP Engelbrecht. *Computational Intelligence: An Introduction*. 2nd ed. West Sussex, Wiley, UK, 2007.
[15] E Mamdani and S Assilian. An experiment in linguistic synthesis with a fuzzy logic controller. *Int. J. Man Mach. Stud*. 1975; **7**, 1-13.
[16] M Sugeno and G Kang. Structure identification of fuzzy model. *Fuzzy Sets Syst*. 1988; **28**, 15-33.
[17] SN Sivanandam, S Sumathi and SN Deepa. *Introduction to Fuzzy Logic Using MATLAB*. Springer, New York, 2007.

[18] JSR Jang. ANFIS: Adaptive-network-based fuzzy inference system. *IEEE Trans. Syst. Man Cyber*. 1993; **23**, 665-85.
[19] N Gulley, JSR Jang and WC Wang. *Fuzzy Logic Toolbox for use with MATLAB user Guide*. The MathWorks, United States, 1995.