

## **ANALYSES OF GAS HYDRATE AND A DETECTION METHODE**

*Ildikó Bölkény<sup>1</sup>, Viktor Füvesi<sup>2</sup>, József Konyha<sup>1</sup>*

<sup>1</sup> Assistant Research Scientist

<sup>2</sup> Research Scientist, PhD

<sup>1,2</sup> *University of Miskolc, Research Institute of Applied Earth Sciences*

### **ABSTRACT**

In this article a model of gas hydrate and a machine learning based predictive detection system are introduced. To develop a preventive, model-based, inhibitor dosing system, some measurement data from width parameter range is needed. Using the data models can be created, which highly influence the quality and stability of control system. In last decade several measurements, which connected to formation of gas hydrate, were performed on our department. These raw data was used for model development and analyses.

### **1. INTRODUCTION**

Appearance of gas hydrate is very similar to ordinary snow. This material can cause serious problem during the production of gas, when the ingredients present and the conditions are enabled, hydrate crystals are formed in the pipeline. In contretemps' case the number of hydrate molecules can be increased, which can agglomerate with each other, it can cause plug in the section of pipeline. In worst case the hydrate plug can effect production outages which results loss of money for the maintainer or in other cases "just" decrease in production [1].

There are more preventive technologies to apply against the formation of hydrate. In practice of gas industry one of the most popular solutions is the usage of thermodynamic inhibitors (THI). The dosage of THI shifts the hydrate curve to region where the conditions are not corresponding for a stable hydrate formation [2]. The methanol (MeOH) is a common compound beside monoethylene glycol (MEG). These inhibitors have to inject in high volume to the gas to be effective against hydrate formation. This technology is not a modern solution, because it has several drawbacks like: a) cost of additional pipe to the gas wells; b) the cost of the methanol regeneration [3]; c) methanol contaminates also intensively the environment.

The injection of low-dosage hydrate inhibitors is a newer alternative technology, where kinetic hydrate inhibitors (KHI) are used to prevent the growth of hydrate molecules [4]. In this group belongs even the antiagglomerants (AA) [6] products, which allow the formation of gas hydrates but keep the hydrate crystals small and dispersed [5]. These modern, low-dosing inhibitors enable the usage and noticeably dynamically spread of locally installed injection systems in the field, at site of the gas wells. Thus injection unit systems are needed for this purpose [7].

During the last decades the staff of the Research Institute of Applied Earth Sciences takes part in severely project, where hydrate prevention and inhibitor injection were in the centre. This paper has two object: a) to show the determination

process of hydrate phase curve; b) show a neural network (NN) based solution for preventive gas hydrate detection alarm system.

## 2. DETERMINATION OF HYDRATE PHASE LIMIT CURVE

On one hand hydrate phase limit curve can be determined by measuring. Practical measurements have been performed by a hydrate forming tester machine at the Research Institute of Applied Earth Sciences. The equipment developed by the Department of Research Instrumentation and Informatics which is shown in Figure 1.



Figure 1  
Hydrate forming tester equipment

This modeling equipment is suitable for simulation of gas pipeline flow. The equipment using field conditions such as  $-20 \dots +30$  °C temperature range, and original gas pipeline pressure range. The flow values in accordance to modeling principles, the flow rate range is 1-10 nl/min. The hydrate forming in capillary cell which placed in a thermostat. To promote hydrate formation water is added which comes from gas well.

Important to note that the practical measurements are only forecasts hydrate limit temperature as a function of time at predefined pressure. The hydrate formation in gas sample is beginning at approximately 16 °C temperature and 60 bar pressure. Results of practical measurements are shown in Table 1 and Figure 2/A.

Table 1  
Results of measurements

Measurements	Start time of hydrate forming [sec]	Temperature [°C]
1.	868.22	16.1
2.	2044.14	16.2
3.	1172.67	17.9

On one hand hydrate phase limit curve can be determined by calculations. Simulation program, called PVTP, was used to perform the calculations of hydrate curve. The program has been patented by the Petroleum Experts. The software supports Windows 32 bit systems. The shape of the limit curve depends on the composition of gas well (Table 2).

Table 2  
Composition of gas

No.	Component	Type	Molar percent [%]	Molecular Weight [mg]
1.	N2	Pure Non Hyd	5.683	28.01
2.	CO2	Pure Non Hyd	22.655	44.01
3.	C1	Pure Hyd	61.941	16.04
4.	C2	Pure Hyd	5.322	30.1
5.	C3	Pure Hyd	2.278	44.1
6.	iC4	Pure Hyd	0.615	58.1
7.	nC4	Pure Hyd	0.768	58.1
8.	iC5	Pure Hyd	0.274	72.2
9.	nC5	Pure Hyd	0.222	72.2
10.	C6	Pure Hyd	0.163	86.2
11.	C7	Pure Hyd	0.063	99.5
12.	C8	Pure Hyd	0.016	112

Therefore the composition of the gas is needed in order to calculate the curve. After the measurements of the components, the hydrate phase limit curve can be calculated by the software.

There are three different structures of gas hydrates, the first (S1) and second (S2) structure are very frequent in industry. The calculation was made in two ways, according to the above mentioned two main hydrate structure types. Both hydrate phase limit curves are shown in Figure 2/B. (S1 is red; S2 is blue). As we can see low temperature and high pressure are needed for the formation of gas hydrates [8].

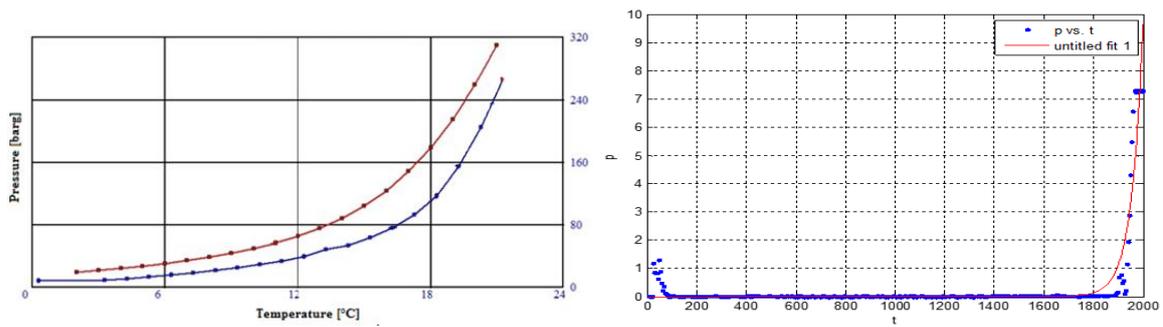


Figure 2

A, Calculated diagram of structure 1 (red) and 2 (blue);

B, Result of a practical measurement

Generally in industrial conditions the operating gas pressure is at 60 bar, therefore this point of this diagram is very important. As we can see the hydrate forming limit temperature of this point is approximately at 12-13 °C in case of structure 1, and at 16 °C in case of structure 2.

As we can see in case of structure 2 the simulated result is meets the measured result, namely 16 °C at 60 bar.

### 3. MEASUREMENTS FOR DETECTION METHODE

High number of measurements were performed with the previously detailed hydrate forming test equipment (Figure 1), using different inhibitor materials and gases from all over Hungary. In last decade more than thousands of analyses were done using that test bench. From this huge database of measurement 50 pieces were randomly selected and used for the investigation. During the measurements mainly the values of differential pressure, inlet pressure, temperature of gas were saved for later investigation.

The appearance of gas hydrate molecules in gas flow is occurred the rises of the pressure in pipe section. It is important to sign in time the appearance of gas hydrate. From practical view the differential pressure gives the most valuable information about the processes in the tube. Thus, in our investigation the value of that parameter was used mainly as input of the alarm system.

### 4. GENERATION OF DATASETS

Three main datasets were generated for the training of alarm system. First a longer training dataset is needed to train the features of prediction and configuration the weights of the network during the training process. One of the most important parameter during the training process is to stop it on time. The early stop occurs that the network does not learn the main features of the training data. In other case, when the training process of the neural networks (NN) takes longer than the optimum, the network can be overtrained. It means that the network can give good estimation when unknown independent dataset is used. So an independent dataset, called validation dataset, is used to stop the training process correct time. When

mean squared error (MSE) value of validation dataset is lowest than it is optimum to stop the training process of the network. In generally the mean squared error can be calculated in the next form, (1).

$$MSE = \frac{1}{n} \sum_i [y_{req}(i) - y_{est}(i)]^2 \quad (1)$$

Where  $y_{req}(i)$  is the required output of the network in the  $i$ th time step,  $y_{est}(i)$  is the estimation of actual network in  $i$ th time step,  $n$  is for the number of samples.

The third generated dataset is the test dataset, which is also independent from the training and validation sets. This dataset is used to compare the result of different network structures. The main parameters of datasets can be found in the next Table 3.

Table 3  
Main parameters of the datasets

Dataset	Number of used measurements [pcs]	Number of data points [pcs]
Training dataset	26	2576
Validation dataset	10	1077
Test dataset	10	1698

As input, the scaled, normalized differential pressure values were used from the measurements in datasets. The required output was an artificially generated alarm signal, which was created from the differential pressure values. A 75% limit, relative to maximal value of the dataset, separates the zero and alarmed levels. Until the actual differential pressure values were under the limit, the datapoints of alarm signal were also zero. When pressure value reaches the limit, the value of alarm signal changed to 1. An example for alarm generation can be seen on Figure 3.

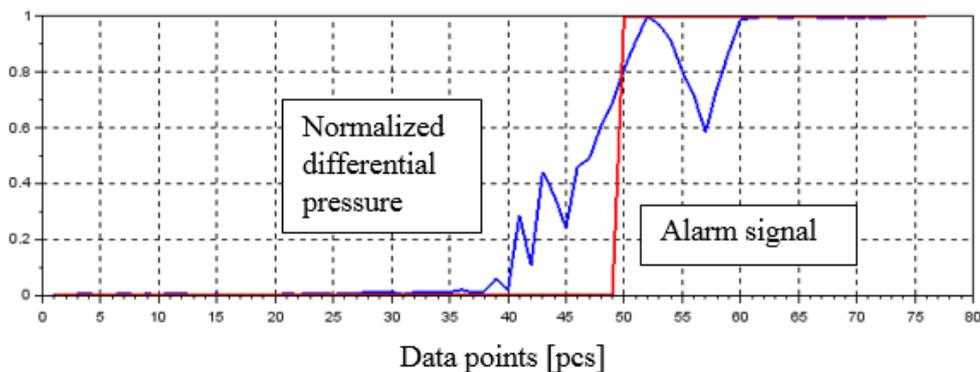


Figure 3  
An example for alarm generation

## 5. ARCHITECTURE OF USED NEURAL NETWORKS AND TRAINING PROCESS

In this paper two network families are compared. First is the neural network auto-regressive model with exogenous input (NNARX), which uses the required outputs as inputs in regressor, so it is not real recurrent network type. Other network type is a neural network output error model (NNOE), which use its earlier outputs as input. These networks realize a nonlinear model using their inputs. The used regressor of that models and the mapping function can be found in (2) for NNARX and (3) for NNOE models.

$$y_{est}(t)=f[x(t-1),x(t-2),\dots,x(t-n_i),y_{req}(t-1),\dots,y_{req}(t-n_{ro})] \quad (2)$$

$$y_{est}(t)=f[x(t-1),x(t-2),\dots,x(t-n_i),y_{est}(t-1),\dots,y_{est}(t-n_o)] \quad (3)$$

Here the estimation of the neural network in  $t$ th timestamp is  $y_{est}(t)$ ,  $x(t-1)$  is the used input of the network in  $(t-1)$ th timestep,  $y_{req}(t-1)$  is the required output of the neural network in  $(t-1)$ th timestamp.  $n_i$  is the size of used tapped delay line of the inputs,  $n_{ro}$  is the size of used tapped delay line of the required outputs,  $n_o$  is the size of used tapped delay line of the outputs of the network. One-one examples for networks can be seen on Figure 4A and 4B.

During the model selection the size of the regressor and the number of hidden neurons in hidden layer of networks were varied.

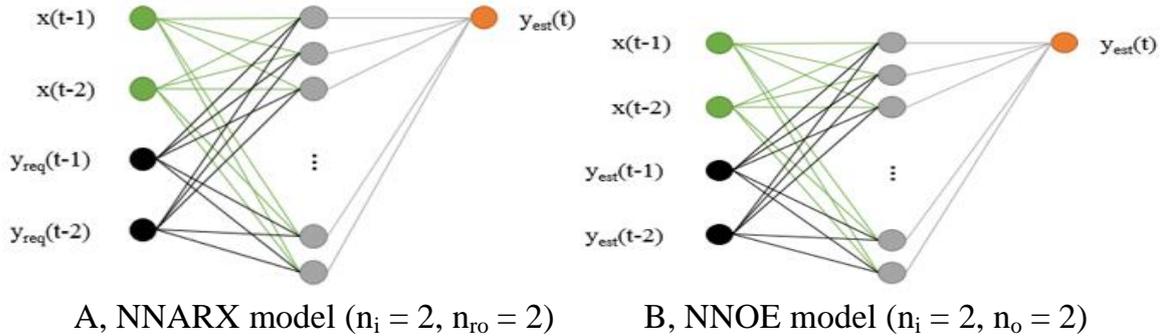


Figure 4  
Example for the used neural network model structure

Twelve neural networks were trained using the generated datasets. Every network was trained for 1000 iterations but just the validated network was saved, which produced the smallest MSE value during the training process. For training the Levenberg-Marquard algorithm was used in Matlab environment with NNSYSID toolbox [10].

## 6. SELECTION PROCESS OF NETWORK AND RESULTS

To investigate the performance of a developed system, the MSE value does not give adequate information about the efficiency of the networks, thus some other index value usage is suggested. The characteristic of the output of the alarm system is similar to shorter or longer impulse shapes. The appearance of rising edge (RE) of these impulses in output of the network can be investigated and use for comparison.

There are several methods, which can be used to find edges in one dimension. One traditional method is, where after some filtering the signal derivatives are analyzed to find the step like changes in the signal [11]. Drawback of the method is the sensitivity to the noises. The other used method is the Canny edge detection method, which use the first derivative of Gaussian to approximate the optimal finite length filter [12]. This method gives good result in our case.

Result of twelve networks were compared, using the MSE and the relative error of found rising edges in the simulated output of the network and the required alarm signal. The comparison of networks can be found in Table 4. The most efficient networks were the networks with smaller hidden layer and with simple regressor. The complicated input configuration do not gave so good results.

Table 4  
Main parameters of the datasets

Network struct.	Reg. of network	No. of hidden neurons [pcs]	Training dataset		Validation dataset		Test dataset	
			MSE	Rel. error of found RE [%]	MSE	Rel. error of found RE [%]	MSE	Rel. error of found RE [%]
NNARX	$n_i = 1;$ $n_{ro} = 1$	10	0,0083	96,2	0,0065	100,0	0,0146	90,0
		12	0,0081	96,2	0,0064	100,0	0,0146	90,0
	$n_i = 1;$ $n_{ro} = 2$	10	0,0209	73,1	0,0184	70,0	0,0227	70,0
		12	0,0202	73,1	0,0177	80,0	0,0203	90,0
	$n_i = 2;$ $n_{ro} = 2$	10	0,0223	73,1	0,0186	90,0	0,0259	70,0
		12	0,0388	69,2	0,0350	50,0	0,0316	60,0
NNOE	$n_i = 1;$ $n_o = 1$	10	0,0086	100,0	0,0062	100,0	0,0157	90,0
		12	0,0058	96,2	0,0048	100,0	0,0127	90,0
	$n_i = 1;$ $n_o = 2$	10	0,0284	76,9	0,0253	60,0	0,0326	60,0
		12	0,0265	65,4	0,0236	70,0	0,0278	50,0
	$n_i = 2;$ $n_o = 2$	10	0,0269	69,2	0,0252	70,0	0,0183	60,0
		12	0,0347	53,8	0,0325	50,0	0,0321	30,0

Most efficient network was a NNOE network with small regressor numbers. The test dataset is showed on Figure 5.

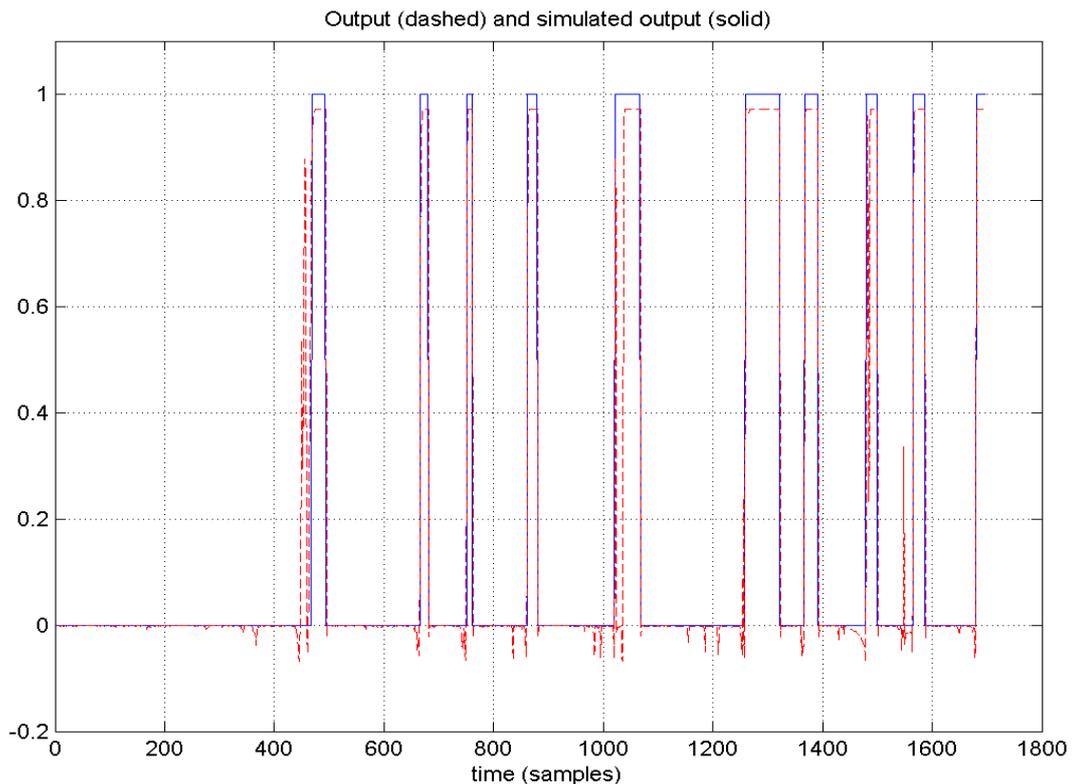


Figure 5  
Example for the used neural network model structure

## 7. CONCLUSION

In the first part of the paper the analyses of gas hydrate measurements is showed. In the second part of the paper a prediction method of appearance of gas hydrate was detailed based on NN. The on time sign of appearance of hydrate is a hard task during the gas production process. In this paper a method is showed which is capable to sign the hydrate on time. The method is based on neural network with recurrent architecture. Using the results of some experiments datasets were generated for training, validation and test purpose of neural networks. Twelve networks were trained and their results compare to get accurate, usable alarm signal on wide parameter range. A simple NNOE network served the most accurate results. For comparison the found rising edges of the signal were investigated with success beside the well-known MSE value.

## 8. REFERENCES

- [1] Gao, S. Q.: **Hydrate risk management at high watercuts with anti-agglomerant hydrate inhibitors**, Energy Fuels, 2009, 23, pp. 2118–2121.
- [2] Kelland, M. A.: **History of the development of low dosage hydrate inhibitors**, Energy Fuels, 2006, 20, pp. 825–847.

- [3] Lederhos, J. P, Long J. P., Sum A., Christiansen R. L., Sloan, JR. E.D.: **Effective Kinetic Inhibitors for Natural Gas Hydrates**, Chemical Engineering Science, 1996, 51 (8), pp. 1221–1229.
- [4] Boxall, J.; May E.: **Formation of gas hydrate blockages in under-inhibited conditions**, 7th International Conference on Gas Hydrates (ICGH 2011), Edinburgh, Proceeding, 2011
- [5] Yana, K. L., Suna, C. Y., Chena, J., Chenb, L. T., Shena, D., Jiaa, M. L., Niua, M., Lva, Y. N., Lia, N., Songa, Z. Y., Niua, S. S., Chena, G. J.: **Flow characteristics and rheological properties of natural gas hydrate slurry in the presence of anti-agglomerant in a flow loop apparatus**, Chemical Engineering Science, 2014, 106, pp. 99–108.
- [6] Hammerschmidt, E. G.: **Formation of gas hydrates in natural gas transmission lines**, Ind. Eng. Chem, 1934, 26 (8), pp. 851-855.
- [7] Jónap, K., Füvesi, V., Vörös, Cs., Vágó, Á., Tornyai, L.: **Analyses and Field Applications of Gas Hydrate Inhibitors**, Journal “Nafta I Plin” (Oil & Gas), 2015
- [8] ME-AFKI: **Nagy inert tartalmú földgázok kitermelését támogató komplex hidrát-gátló technológia kutatása és fejlesztése GOP 1.1.1-09/1-2010-0114**, Megvalósíthatósági tanulmány, 2013
- [9] Rónai, L., Bölkény I.: **Regulation of an Inhibitor Dosing System**, XXVIII. International Multidisciplinary Scientific Conference, Innovative Engineering Design and Development, Miskolc, Hungary, 2015 Paper CD. 6 p. (ISBN: 978-963-358-051-6)
- [10] M. Nørgaard, O. Ravn, N. K. Poulsen, L. K. Hansen: **Neural networks for Modelling and Control of Dynamic Systems**, Springer-Verlag, London, UK, 2000.
- [11] Füvesi, V.; Kovács, E.: **Separation of Faults of Electromechanical Drive Chain using Artificial Intelligence Methods**, 18th “Building Services, Mechanical and Building Industry days” Int. Conf., Debrecen, Hungary, 2012, pp. 19-27.
- [12] Canny, J.: **A computational approach to edge detection**, IEEE Trans. Pattern Anal. Mach. Intell. 8, 6 (Nov. 1986), pp. 679-698.