

## **A NOVEL TECHNIQUE FOR MONITORING HYDRATE SAFETY MARGIN**

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### **ABSTRACT**

Great concerns with flow assurance issues have been raised by the oil and gas industry, while the industry is increasingly moving to deepwater reservoirs. Gas hydrate blockages are one of the most common risks for the long distance offshore gas and oil production transport pipelines. Various types of hydrate inhibitors are usually deployed to ensure unimpeded flow of hydrocarbons. At present hydrate inhibitors are injected at the upstream of the pipelines according to approximate assessment of the flowing conditions including the produced water cut and the hydrate phase boundary that is determined based on the worst temperature and pressure conditions, without any means of monitoring the actual degree of inhibition along the pipeline.

A novel technique has been developed to optimize the injection of hydrate inhibitors by monitoring the actual hydrate safety margin (i.e., degree of inhibition), which makes it possible to reduce unnecessary cost and potential impact on the environment. It measures the acoustic velocity and electrical conductivity of downstream aqueous samples and then determines both the inhibitor concentration and salt concentration through a trained artificial neural network. The hydrate phase boundary, hence the hydrate safety margin, are finally determined by an integrated in-house thermodynamic model using the determined salt and inhibitor concentrations. Its performance has been intensively evaluated using synthetic samples and real produced water samples by the authors and some oil & gas and service companies. This communication reports the success in development of the hydrate inhibition monitoring system. Results of the evaluation demonstrate that the system can be used for different inhibition systems including methanol-salt systems, mono ethylene glycol-salt systems, and kinetic hydrate inhibitor-salt systems with an acceptable measurement accuracy.

*Keywords:* gas hydrate; flow assurance; hydrate inhibition; acoustic velocity; electrical conductivity; artificial neural network.

### **INTRODUCTION**

Gas hydrate blockages commonly cause serious problems to oil & gas transport pipelines, processing facilities, and deepwater drilling. It impedes hydrocarbon flow and even exposes personnel safety at danger [1, 2]. A variety of chemical additives are used as hydrate inhibitors to prevent hydrate blockage. In terms of inhibition mechanisms, there are three kinds of hydrate inhibitors, including thermodynamic hydrate inhibitors (THIs), kinetic hydrate inhibitors (KHIs) and anti-agglomerants (AAs).

THIs shift the hydrate phase boundary to a relatively low temperature and high pressure, allowing the operation conditions outside the hydrate stability zone (HSZ). Methanol (MeOH) and mono ethylene glycol (MEG) are two of the most widely used THIs. In contrast, KHIs do not prevent hydrate formation but delay hydrate nucleation and hinder hydrate crystal growth within certain degree of subcooling, providing sufficient time for the hydrocarbon fluids to pass through the length of a transport pipeline where the thermodynamic conditions are in the HSZ [3-

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7]. AAs are different from both THIs and KHIs. AAs allow hydrate formation but prevent individual hydrate crystals from agglomerating together, therefore, maintain the hydrocarbon system transportable [7, 8]. KHIs and AAs together are further called “low dosage hydrate inhibitors (LDHIs)” as the concentrations of KHIs or AAs are low (typically, less than 3%) in comparison with those of THIs [8, 9], where in some cases high concentrations of up to 60 mass% of MeOH or MEG may be needed to have sufficient inhibition [10, 11].

In general, the amount of a hydrate inhibitor is determined based on the predicted or measured hydrate phase boundary and the operation conditions such as temperature and pressure, water-cut, and possible loss of the inhibitor to non-aqueous phases. However, as a common flow assurance strategy, excessive dosages of an inhibitor have to be applied to minimize the risk of pipeline blockage due to hydrate formation. This often results in even more cost and severe impact on the environment.

In recent years a great deal of effort has been made to optimize hydrate inhibitor injection to minimize the cost and environmental impact. Willmon and Edwards [12] suggested experience-based rules of thumb to help rationalizing the dosage of hydrate inhibitors. A research programme has been carried out in the Centre for Gas Hydrate Research at the Institute of Petroleum Engineering, Heriot-Watt University, since 2004, aiming to develop techniques for hydrate monitoring and early warning [13, 14]. In this communication, we report the development of a novel technique for monitoring the hydrate safety margin. By measuring acoustic velocity and electrical conductivity in downstream aqueous samples it determines not only the inhibitor and salt concentrations, but also the hydrate phase boundary hence the hydrate safety margin given that the hydrocarbon composition and the temperature and pressure in the pipeline are known.

## METHODOLOGY

### Hydrate monitoring approach

This work is aimed at developing a technique that can monitor the degree of hydrate inhibition, i.e., how far away the operating conditions are from the HSZ. Hydrate safety margin was introduced to describe the degree of hydrate inhibition. It is

defined as the temperature difference between the actual fluid temperature and the hydrate dissociation temperature at a given pressure. The developed technique measures concentrations of the salt and inhibitor in the aqueous phase and determines the hydrate safety margin. The determined hydrate safety margin can be used as traffic lights to help the operators have an appropriate control of hydrate inhibition. In Figure 1, a red light can be shown if the determined hydrate safety margin is negative, which alarms the operator that the pipeline conditions are inside the HSZ, i.e., the pipeline has already been exposed to hydrate formation risk, therefore, more hydrate inhibitor must be injected into the pipeline immediately; a green light indicates that the system is safe from hydrate formation and the current injection rate of the inhibitor is appropriate; an amber light reminds the operator that the system is close to the HSZ and the inhibitor injection rate needs to be increased a bit; finally, a blue light means over inhibition, therefore, the inhibitor injection rate should be reduced. Furthermore, the increment or decrement of hydrate inhibitors can be quantitatively estimated using the determined hydrate safety margin.



Figure 1 Hydrate safety margin determined by the developed hydrate monitoring technique could be used as traffic lights to help the operators have a better control of hydrate inhibition.

### Velocity-conductivity method

For salt aqueous solutions in the absence of any other chemical additives, the hydrate suppression temperature (i.e., dissociation temperature shift) can be determined by measuring the electrical conductivity [15]. To characterize liquid mixtures for industrial processes, an acoustic multi-sensor system was developed to measure the concentrations of the chemicals such as

MeOH and MEG in the solutions without salts [16]. However, these methods may not be applicable to most hydrocarbon transport pipelines where salts and at least one inhibitor often coexist in the aqueous phase. Sandengen and Kaasa [17] developed an empirical correlation that determined the MEG and NaCl concentrations by measuring the density and electrical conductivity of water samples under examination. However, the critical weakness of this method is that it requires high accuracy of the density measurement, which prevents it from application to real produced water samples that usually contain solid particles (sands and clays) and oil droplets.

It is well known that electrical conductivity of liquid solutions depends on the concentration of ions and their activity. The aqueous fluids in pipelines usually are electrolyte solutions and the conductivity is proportional to the salt concentration. The activity of the ions is related to temperature, and impurity like nonconductive chemical additives. Measurements of electrical conductivity could directly reflect the concentrations of chemicals such as salts, THIs (alcohol) and KHIs (polymers). Clay and Medwin [18] presented a simple correlation in which the sound velocity in sea water was described as a function of salinity and temperature. Acoustic velocity has been successfully applied to investigate a variety of solutions and binary gas mixtures [19-22]. As a result, electrical conductivity and acoustic velocity were chosen as two parameters to simultaneously determine both salt and inhibitor concentrations.

Artificial neural network (ANN) provides a numerical tool for such applications in which multi-parameter correlations are needed but the interaction and the relations between the parameters are not well known [23, 24]. Therefore, ANN correlations were developed to determine salt and inhibitor concentrations using the measured electrical conductivity, acoustic velocity, and temperature.

#### Determination of hydrate safety margin

A thermodynamic model HWHYD is employed to determine the hydrate safety margin [25, 26]. Firstly, the hydrate phase boundary is determined by the model using the measured salt and inhibitor concentrations and the composition of the hydrocarbon fluids (oil or gas) in the pipeline.

Then the hydrate safety margin is calculated by the difference between the determined hydrate dissociation temperature and the pipeline temperature at the pipeline pressure.

A methodological chart of the hydrate inhibition monitoring system is shown in Figure 2. Both the acoustic velocity  $V$  and electrical conductivity  $\sigma$  are a function of the salt concentration  $C_{Salt}$ , the inhibitor concentration  $C_{HI}$ , and temperature  $T$ . The velocity, conductivity, and temperature of an aqueous sample under examination are measured and fed to an ANN correlation that has previously been trained using a set of electrical conductivity and acoustic velocity data. The ANN correlation determines the salt and inhibitor concentrations. After loading the determined concentrations, the measured temperature, and the given hydrocarbon composition, the model HWHYD predicts the hydrate phase boundary. Finally, the hydrate safety margin is simply calculated given that the pipeline temperature and pressure are known.

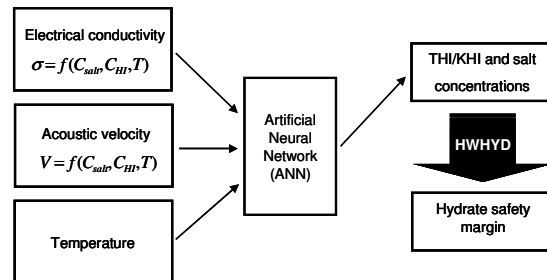


Figure 2 Methodological chart of the hydrate inhibition monitoring system

## DEVELOPMENT OF PROTOTYPE DEVICE

### Instrumentation

A prototype device of the conductivity and velocity (C-V) technique has been developed. The device mainly consists of three units, including a sample cell, a measurement unit, and a personal computer. The sample cell is made of stainless steel for aqueous samples under examination. The measurement unit is equipped with a conductivity meter with a 4-pole conductivity probe, an ultrasonic pulser/receiver with two ultrasonic transducers that are mounted on the two opposite sides of the sample cell. The measured electrical conductivity and temperature

(by the conductivity meter) and the acoustic velocity (by the pulser/receiver) are acquired and sent to the computer by a high-speed data acquisition card. Specific software is developed and installed in the computer. The software is capable of determining salt and inhibitor concentrations using the measured conductivity, velocity, and temperature. By integration with a thermodynamic model HWHYD, the software can also determine the hydrate phase boundary, hence the hydrate safety margin given the hydrocarbon composition and the pipeline conditions.

Figure 3 shows a picture of the prototype C-V device. A graphic user interface (GUI) was built to make the C-V device software user friendly. In Figure 4 the GUI shows the measured temperature, electrical conductivity, acoustic velocity, and the determined salt and inhibitor concentrations. By clicking on “HSZ” on the menu bar, a HSZ window will appear and the determined salt and inhibitor concentrations will be automatically fed to the integrated thermodynamic model (Figure 5). The hydrate phase boundary is determined by the model after the hydrocarbon composition is loaded. The determined hydrate phase boundary can be shown in a plot (the blue curve in the insertion in Figure 5) or saved in a Microsoft Excel data file. The hydrate safety margin can be calculated if the operating temperature and pressure are known. In the insertion in Figure 5, as an example, the cross represents the operating conditions, the calculated hydrate safety margin is represented by  $\Delta T$ .



Figure 3 Picture of the prototype C-V device

### Development of the ANNs

Several ANNs have been developed for THI-salt systems and KHI-salt systems, including MEG-salt systems, MeOH-salt systems, and KHI systems. In Table 1 KHI-a is Luvicap EG<sup>®</sup> (a poly(N-vinylcaprolactam (PVCap)-based kinetic inhibitor, supplied by BASF), KHI-b is also polymer-based and its formula was not disclosed by the supplier. The ANNs were developed for a temperature range from 0 to 25 °C and typical concentrations of hydrate inhibitors: 0 to 50 mass% of MEG, 0 to 20 mass% of MeOH, 0 to 2 mass% of PVCap (active polymer) of KHI-a, 0 to 3 mass% of KHI-b. The salt concentrations were different for individual inhibition systems: 0 to 10 mass% for the MEG and MeOH systems, and the KHI-b system, 0 to 7 mass% for the KHI-a system. Table 1 summarizes the results of training, validation, and testing. In general, the ANN outputs are in good agreement with the experimental data of the salt (NaCl) and inhibitor concentrations.

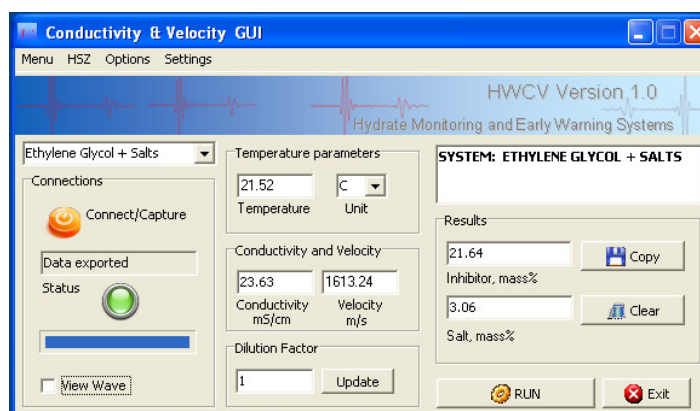


Figure 4 GUI shows the measured temperature, electrical conductivity, acoustic velocity, and the determined salt and inhibitor concentrations.

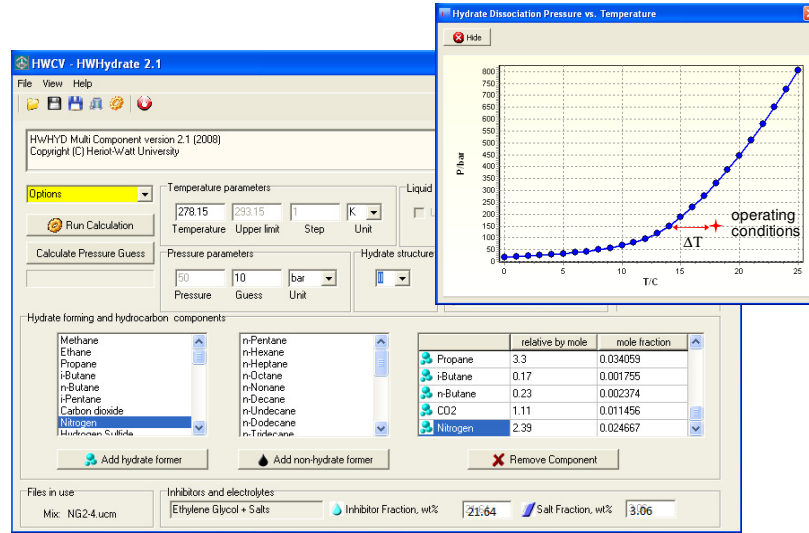


Figure 5 HSZ window shows the loaded hydrocarbon composition and the determined salt and inhibitor concentrations (In the insertion, the cross point denotes the operating conditions,  $\Delta T$  denotes the determined hydrate safety margin).

Table 1 Results of the ANN evaluation, in mass%

Hydrate inhibitors	Deviation in training		Deviation in validation		Deviation in testing	
	Average	Maximum	Average	Maximum	Average	Maximum
MEG	0.08	0.4	0.09	0.4	0.18	0.4
NaCl	0.08	0.5	0.10	0.5	0.12	0.27
MeOH	0.06	0.3	0.06	0.4	0.10	0.25
NaCl	0.04	0.23	0.04	0.17	0.15	0.23
KHI-a	0.04	0.18	0.03	0.20	0.02	0.04
NaCl	0.04	0.21	0.04	0.24	0.08	0.15
KHI-b	0.06	0.4	0.06	0.24	0.07	0.3
NaCl	0.04	0.3	0.04	0.3	0.04	0.14

Table 2 Evaluation results of the prototype C-V device, in mass%

Inhibition system	Water	NaCl concentration			Inhibitor concentration*		
		Exp.	C-V	AD	Exp.	C-V	AD
MEG-NaCl	DW	3.50	3.51	0.01	25.00	24.93	-0.07
MEG-NaCl	DW	6.00	5.85	-0.15	35.00	34.80	-0.20
MEG-salts	PW	4.97	4.90	-0.10	25.00	25.16	0.16
MeOH-NaCl	DW	2.50	2.58	0.08	18.00	17.57	-0.4
MeOH-NaCl	DW	8.00	7.77	-0.23	15.00	14.93	-0.07
MeOH-NaCl	DW	4.00	3.16	-0.8	24.00	23.32	-0.7
KHI-a-NaCl	DW	4.50	4.47	-0.03	0.80	0.85	0.05
KHI-a-NaCl	DW	4.00	3.96	-0.04	1.50	1.51	0.01
KHI-b-NaCl	DW	1.00	1.03	0.03	1.50	1.64	0.14
KHI-b-NaCl	DW	1.50	1.54	0.04	2.20	2.38	0.18
KHI-a-salts	PW	4.97	4.83	-0.14	1.20	1.19	-0.01
KHI-a-salts	PW+CI+SI	4.97	4.86	-0.11	1.10	1.10	0.00

\* For KHI-a systems the inhibitor concentration means the concentration of PVCap.

### Evaluation of the prototype C-V device

The prototype C-V device was evaluated using synthetic solutions containing various concentrations of salt (NaCl) and inhibitors. Table 2 shows the evaluation results (In Table 2, DW and PW stand for distilled water and produced water, respectively; AD stands for absolute deviation, defined as a difference between the measured and experimental concentrations). The measured salt and inhibitor concentrations (“C-V” in Table 2) agree well with the experimental data (“Exp.” in Table 2). In addition, there are slightly larger deviations observed for the solution containing 24 mass% MeOH and 4 mass% NaCl. This infers that larger deviations may occur if salt or inhibitor concentrations are beyond the range of the ANN training data.

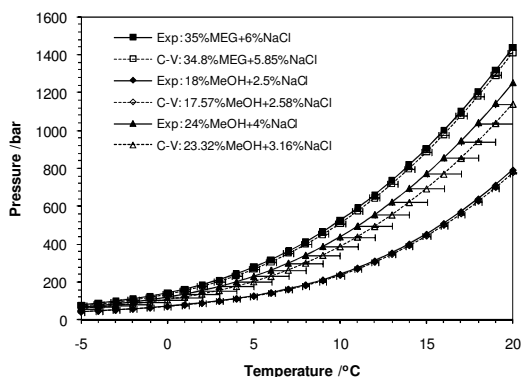


Figure 6 Comparison of the methane hydrate phase boundaries determined using the measured and experimental concentrations of the salt and inhibitors. (Error bands =  $\pm 0.3$  °C for the curve with 34.80 mass% MEG and 5.85 mass% NaCl,  $\pm 0.2$  °C for the curve with 17.57 mass% MeOH and 2.58 mass% NaCl,  $\pm 1$  °C for the curve with 23.32 mass% MeOH and 3.16 mass% NaCl).

It is essential to know how the measurement errors of the salt and inhibitor concentrations shift the hydrate phase boundary. Figure 6 shows the shifts in the hydrate phase boundary for the three evaluation measurements (in Table 2) with the largest deviations between the measured and the experimental concentrations of MEG and MeOH. Methane was used for simplicity. It can be seen that the measurement errors of the prototype C-V device are within 0.3 °C for the MEG-salt solution, 0.2 °C for the MeOH-salt solution (MeOH  $\leq$  20 mass%). However, for the

solution with 24 mass% of MeOH, the measurement errors shift the hydrate phase boundary about 1 °C, which is mainly due to the measured MeOH concentration which is beyond the ANN training range.

All the ANNs were initially trained using synthetic solutions in which NaCl was the only salt. Actually, real produced water usually contains multiple salts such as NaCl, KCl, CaCl<sub>2</sub>, MgCl<sub>2</sub>, etc. Moreover, corrosion inhibitors (CIs) and scale inhibitors (SIs) are often injected into a pipeline to prevent corrosion and scale formation. Therefore, further evaluation of the prototype C-V device was performed using a produced water to examine the effect of the presence of other salts on its performance. The produced water contains multiple salts: NaCl (3.11 mass%), CaCl<sub>2</sub> (0.16 mass%), KCl (0.34 mass%), MgCl<sub>2</sub> (0.33 mass%), others (1.03 mass%). In Table 2 the results demonstrate that the produced water with multiple salts did not noticeably affect the measurement accuracy for the MEG and KHI-a systems by comparison with the synthetic NaCl solutions. To investigate the possible effect of CIs and SIs, typical concentrations of a CI (500 ppm) and a SI (550 ppm) were added to the produced water. Considering the fact that KHI systems (typically low KHI concentrations) are more vulnerable to other chemicals, the produced water-CI-SI solution was used to test the prototype for KHI systems. The results in Table 2 also show that the presence of the CI and SI did not have measurable influence on the measurements of the KHI (KHI-a) and salt concentrations.

For MeOH-salt systems, two other produced water samples were used. The produced water samples were taken from the downstream of a pipeline that was inhibited with MeOH, and contained multiple salts and an unknown amount of a CI. The C-V device measured the first one (PWS-1) with 2.9 mass% of salts and 22.2 mass% of MeOH, and the second one (PWS-2) with 3.0 mass% of salts and 23.0 mass% of MeOH. For PWS-1, two hydrate phase boundaries were determined by the C-V device and the freezing point depression (FDP) method [14], for a typical natural gas that was composed of methane (88.3 mol%), ethane (5.4 mol%), propane (1.5 mol%), isobutene (0.2 mol%), normal butane (0.3 mol%), isopentane (0.1 mol%), normal pentane (0.09 mol%), nitrogen (2.39 mol%), carbon dioxide (1.72 mol%). For

PWS-2 with the same natural gas, the C-V device determined the hydrate phase boundary, and one hydrate dissociation point was experimentally measured at 7.4 °C and 139.4 bar. In Figure 7, comparison is made between the C-V device and the other methods. For PWS-1, the deviation in the two hydrate phase boundaries determined by the C-V device (solid curve) and by FPD method (dashed curve) is within 0.5 °C for pressure up to 200 bar. For PWS-2, the measured dissociation point (filled square point) is in line with the hydrate phase boundary (empty triangle points) that was determined by the prototype C-V device.

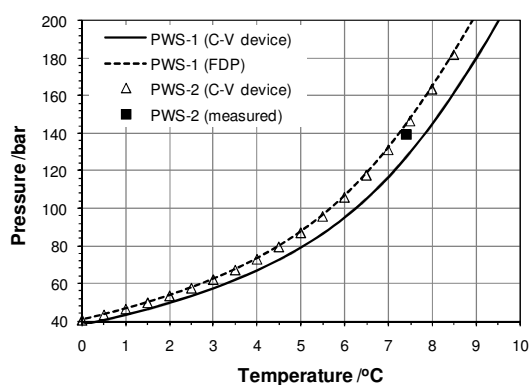


Figure 7 Evaluation results of the prototype C-V device using two real produced water samples

The prototype C-V device was also tested in field laboratories by some of the leading oil & gas and service companies, including Total in Pau (France), Statoil in Trondheim (Norway), Dolphin Energy in Qatar, Petronas in Nouakchott (Mauritania), NIGC in South Pars Gas Complex fields (Iran), Champion Technology in Aberdeen (UK). The field evaluations were conducted for a variety of hydrate inhibition systems such as MEG systems, MeOH systems, and KHI systems in the presence or absence of salts [27, 28]. The evaluation results suggested that the C-V technique provides a simple and reliable tool for monitoring of hydrate safety margin (THI systems), KHI injection process (KHI systems), and MEG regeneration process.

## CONCLUSIONS

A novel technique has been developed for determining the concentration of hydrate inhibitors in the aqueous phase and monitoring the hydrate safety margin. By measuring the

electrical conductivity and acoustic velocity, the developed C-V prototype device can determine the concentrations of thermodynamic hydrate inhibitors, kinetic hydrate inhibitors, and salts. Integrating with a thermodynamic model, it can also determine hydrate the stability zone hence the hydrate safety margin if the hydrocarbon composition and the operating conditions are known. Extensive evaluation was conducted under both lab and field conditions. The results demonstrate that the C-V technique provides a simple and reliable means for optimising hydrate inhibitor injection rates and minimising both the operating cost and the impact on the environment, as well as improving the reliability of hydrate prevention strategies.

## ACKNOWLEDGMENTS

This work was part of an industrial joint project “Hydrate Monitoring and Early Warning Systems” that was financially supported by BP, Chevron, NIGC, Petronas, Statoil, and TOTAL, which is gratefully acknowledged.

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