Original Research Article

Software Development for the Prediction of Hydrate Formation Conditions

ABSTRACT (ARIAL, BOLD, 11 FONT, LEFT ALIGNED, CAPS)

Web-based software named "SEN ABLE" to run on both desktop workstations and mobile devices was developed to simulate hydrate formation temperature and pressure conditions using available data to develop the software. Twenty four (24) correlations were studied and streamlined down to 15. These fifteen correlations were coded into the 'senengine', the backend code that powers the software. The software and its engine were created in scarlet macaw and adobe dreamweaver, using programming languages such as html5, php, css3, and javascript. The software was tested and debugged on mozilla firefox web browser and a blackberry™ mobile phone browser. It was further tested on other major web and mobile browsers for compatibility. After various development and debugging phases, the software was tested and evaluated using experimental data. Also, the results from the software were compared with those gotten from using the experimental data in a commercial software, hydrate plus. The comparison showed pleasing results.

The software is a cheaper alternative to the other expensive commercial softwares that perform the same functions. Also, with the additional advantage of being web based, it is now possible to do hydrate simulations on the go, which erstwhile could not have been done. It is also intended to further develop the software into a fully robust solids deposition simulation suite.

*Keywords: [*Web; software; hydrate plus; deposition; hydrate temperature; hydrate pressure*} (Arial, inclined, 10 font, justified)*

ARTICLE ROADMAP

1. INTRODUCTION (ARIAL, BOLD, 11 FONT, LEFT ALIGNED, CAPS)

Gas hydrates (or clathrate hydrates) are ice like crystalline structures that are composed of gas components such as methane and carbon dioxide as guest molecules entrapped into cavities formed by water molecules. Most low molecular weight gases, including O_2, H_2, N_2, CO_2, CH_4, H_2S, Ar., Kr and Xe, as well as some higher hydrocarbons and freons, will form hydrates at suitable temperatures and pressures. Gas hydrate formation is always expected whenever a system of natural gas and water exists at especially high pressure and low temperature conditions.

They usually occur in gas and gas/condensate wells, as well as in oil wells. The intensity and location of hydrate accumulations in a well vary and depend on:

a) operating regime

b) design
c) fluid composition
d) geothermal gradient in the well
e) other factors.

Different methods are currently in use for reducing hydrate problems in hydrocarbon transfer lines and process facilities. The most practical methods are;

a) at fixed pressure, operating at temperatures above the hydrate formation temperature. this can be achieved by insulation or heating of the equipment.
b) at fixed temperatures, operating at pressures below hydrate formation pressure.
c) dehydration i.e. reducing water concentration to an extent of avoiding hydrate formation.
d) inhibition of the hydrate formation conditions by using chemicals such as methanol and salts. the most common inhibitors are thermodynamic inhibitors such as methanol and glycols; however, produced water that contains electrolytes also has inhibiting effects.
e) changing the feed composition by reducing the hydrate forming compounds or adding non hydrate forming compounds.
f) preventing, or delaying hydrate formation by adding kinetic inhibitors.
g) preventing hydrate clustering by using hydrate growth modifiers or coating of working surfaces with hydrophobic substances.
h) preventing, or delaying hydrate formation by adding kinetic inhibitors.

The first step in controlling hydrate formation is to understand which pressure and temperature conditions/locations in the specific system are conducive to gas hydrate formation. A number of computer simulators are available for this purpose, usually as adjuncts to more general phase pvt simulators. The models vary in how well they compute the chemical activity of the water phase, the effect of higher-molecular-weight hydrocarbons, and the effect of hydrate inhibitors. Besides the dissociation pressure/temperature (P/T) points for the hydrate, the information required and derivable from such models is the amount of hydrate formed, the composition of all phases, and the distribution of inhibitors throughout all phases.

The second control step is the comparison of this information with the measured or expected (P/T) profile within the production system. A method of coping with hydrate formation is then selected (e.g., producing the hydrocarbons under conditions that avoid the hydrate P/T formation zone or using a suitable inhibition method. An estimated amount of $1.5 billion is spent on removing hydrates yearly. Instead of spending this amount of money on cleaning up pipelines, a better option is to make use of the preventive method. The prevention-by-well-design scenario, albeit initially potentially expensive, may be more cost effective throughout the life of the well as against cleaning or dissolving. The cost of treatments involves not only the chemical itself but the deferred and/or lost oil production attendant to the well's downtime for the treatment. In the light of this, it is usually simpler and more economical to use a computer program that

will predict the precipitation and deposition conditions of gas hydrates. A software that predicts the gas hydrate formation pressure and temperature conditions, in three-phase equilibrium (liquid water, hydrocarbon gas, and solid hydrate) using various mathematical correlations and models is to be developed. Also, to ensure mobility and cross platform compatibility, which is the ICT trend of the day, the software is to be developed using web technologies. These web technologies include **HTML**, **CSS**, and **JAVASCRIPT** for the front end and **PHP** and **MYSQL** for the backend. The software will be hosted on a remote server. This way, predicting the solid precipitation and deposition conditions will not be restricted to using desktop systems again, but will also be doable on mobile phones, tablet computers and laptop computers.

Researchers have already made significant progress in understanding the structure of hydrates and conditions that make them form. They have even gone ahead to perform experiments and create mathematical expressions that can be used to estimate these formation conditions. Von- stackelberg and Muller (1951)[1] studied the hydrate structure using x-ray diffraction methods. Their work along with works by Claussen (1951)[2] identified two hydrate structures; structure I (SI) and structure II (SII). The SI hydrates were discovered to have 46 water molecules per eight cavities, two small spherical cavities with 12 pentagonal faces (5^{12}) and six large oblate cavities with two hexagonal faces and 12 pentagonal faces ($5^{12}6^{2}$). The SII hydrates consist of 136 water molecules per 32 cavities, 16 small cavities with 12 pentagonal faces (5^{12}) and eight large cavities with 12 pentagonal and four hexagonal faces ($5^{12}6^{4}$), all in a spherical shape. Ripmeester et al. (1987)[3] discovered a third type of hydrate structure (structure H). The formation of hydrate SH requires both small and large molecules to be stabilized. The hydrates with SH contain 34 water molecules per six cavities, three cavities formed by 12 pentagonal (5^{12}), two cavities formed by three square, six pentagonal, and three hexagonal faces ($4^{3}5^{6}6^{3}$), and one large cavity formed by 12 pentagonal and eight hexagonal faces ($5^{12}6^{8}$). Jeffrey et al. (1965)[4] studied crystallography on hydrates SI and SII. A summary of their experience showed that hydrates are “clathrates”. Hammerschmidt (1934)[5] determined that natural gas hydrates could block the gas transmission pipelines sometimes at temperatures above the freezing point of water. This discovery opened the eyes of the industry to the importance of hydrates and how crucial it is to combat them.

After Hammerschmidt (1934)[5] discovered the phenomenon of hydrates blocking, Katz et al. (1959)[6] started an experimental study on hydrates. It is practically impossible to measure the hydrate formation pressure and temperature for every gas composition, so Katz et al’s method was created to estimate the hydrate formation conditions for natural gases using two approaches; for SI and SII. Hence, they can only predict for SI and SII hydrate structures. The k-value method was generated before the determination of the hydrate-crystal structure and was improved upon by Katz and co-workers[6]. The k-value charts were generated for methane, propane, butane, carbon dioxide, hydrogen sulphide and nitrogen. using the k-value of every component in the mixture at three-phase (l_w-h-v) equilibrium, the hydrate formation pressure at any given temperature could be estimated or vice versa. In a rather different approach, Katz

and his students, in 1959, developed the gas gravity chart. It is a plot that relates the hydrate pressure and temperature with specific gravity of natural gases, not including non-hydrocarbons. They generated this plot from limited experimental data from Deaton and Frost (1946)[7], Wilcox et al (1949)[8], Kobayashi and Katz (1941)[9] and significant calculations based on the k-value method. The method is simple and may be used for initial estimation of hydrate formation conditions. based on the gas-gravity plot. Kobayashi et al (1987)[10] developed the following equation to estimate the hydrate formation temperature at a given pressure.

$$t = 1/[c_1 + c_2(\ln p) + c_3(\ln\gamma) + c_4(\ln p)^2 + c_5(\ln p)(\ln\gamma) + c_6(\ln\gamma)^2 + c_7(\ln p)^3 + c_8(\ln\gamma)(\ln p)^2 + c_9(\ln\gamma)^2(\ln p) + c_{10}(\ln\gamma)^3 + c_{11}(\ln p)^4 + c_{12}(\ln\gamma)(\ln p)^3 + c_{13}(\ln\gamma)^2(\ln p)^2 + c_{14}(\ln\gamma)^3(\ln p) + c_{15}(\ln\gamma)^4] \quad (1)$$

By having the pressure in bar and temperature in ^{o}C, the Kobayashi et al (1987)[10] correlation would have the best results with experimental data. the coefficients for this correlation are shown in Table 1 below.

Table 1: Coefficients for Kobayashi et al (1987) correlation

coefficients for calculating the hydrate formation temperature from equation 2.3		
$c_1 = 2.7707715 \times 10^{-3}$	$c_2 = 2.782238 \times 10^{-3}$	$c_3 = -5.649288 \times 10^{-4}$
$c_4 = -1.298593 \times 10^{-3}$	$c_5 = 1.407119 \times 10^{-3}$	$c_6 = 1.785744 \times 10^{-4}$
$c_7 = 1.130284 \times 10^{-3}$	$c_8 = 5.9728235 \times 10^{-4}$	$c_9 = -2.3279181 \times 10^{-4}$
$c_{10} = -2.6840758 \times 10^{-3}$	$c_{11} = 4.6610555 \times 10^{-3}$	$c_{12} = 5.5542412 \times 10^{-4}$
$c_{13} = -1.4727765 \times 10^{-3}$	$c_{14} = 1.3938082 \times 10^{-5}$	$c_{15} = 1.4885010 \times 10^{-6}$

Based on a statistical method by Van der waals and Platteeuw (1952)[11], Parrish and Prausnitz (1972)[12] developed the first thermodynamic model for calculating the hydrate formation conditions. This work was further developed by Du and Guo (1990)[13] to predict the hydrate formation conditions for systems including alcohol solutions. Another model developed by Javanmardi and Moshfeghian (2000)[14] can predict the hydrate formation conditions for systems including electrolyte solutions. If there is a combination of electrolytes and alcohol in the solution, the model by Nasrifar et al (1998)[15], and the model by Nasrifar and Moshfeghian (2000)[16] can be used to predict the hydrate formation conditions.

This article has been organized into four sections. The first section is the introduction which contain a general overview of the formation, accumulation, and methods of control of hydrates in gas and gas/condensate wells. The focus of the second section is on a new approach of applying web technologies to develop a software in such a way that work done on a desktop workstation could be

continued on a mobile device, such as a tablet computer or even a smartphone. The third section is on the findings and analysis of the models used in hydrate prediction and the final section is the concluding part of the study.

2. MATERIAL AND METHODS / EXPERIMENTAL DETAILS / METHODOLOGY (ARIAL, BOLD, 11 FONT, LEFT ALIGNED, CAPS)

To predict the formation pressures and temperatures of gas hydrates, a study was carried out to determine various applicable models and correlations that have been developed over time. 24 different models were obtained and tested. The testing phase involved comparing the predictions by the models with experimental data. These 24 models were further streamlined to 15, which were then hard coded into the software.

To develop the software, a new approach of applying web technologies was adopted. The software was developed in such a way that work done on a desktop workstation could be continued on a mobile device, such as a tablet computer or even a smartphone. Both jobs are constantly in sync and can be updated from any of the two ends. The software also has an extra memory feature, such that, even when it is closed, or when the user logs out, the data used for the simulation are stored in a database and will reappear the next time the user logs in.

The web technologies employed for the development of the software include;

a) **HTML5, CSS3, JQUERY** for the front end development and user interface.
b) **JAVASCRIPT, PHP5 and MYSQL** for the engine.

The software requests data in a most user friendly manner, and outputs results using charts and tables. Detailed review of the software and steps carried out in its development are discussed in subsequent sub-sections.

The models used in the estimation of the formation conditions of hydrates were grouped into 2 on an input/output basis. The groupings are shown in Table 2 below.

Table 2: Groupings of models on the basis of input and output parameters

Input: Temperature (K)	**Input: Pressure (psia)**
Output: Pressure (psia)	**Output:Temperature (K)**
Ostergaard et al 2000	Berge 1986
Wilcox et al 1941	Kobayashi et al 1987
Hammerschmidt 1934	Hammerschmidt
Makogan 1981, 1997	Sun et al 2003
Safamirzael 2011	Safamirzael
Molar mass method	Motiee 1991
Density method	Wilcox et al
	Towler et al 2005

These models have varying degrees of accuracy in the estimation of the hydrate formation pressure and temperatures.

Developing the software; Input data required

Results of analysing the different correlations, models, and methods to be employed in the development of the software showed the following required input parameters.

Table 5: Input parameters required for the different correlations

Input parameters	**Models, Correlations or Methods that make use of it.**
Pressure (psia)	All
Temperature (K)	All
Gas composition: C_1 (%) **	All
Gas composition: C_2 (%)**	All
Gas composition: C_3 (%) **	All
Gas composition: nC_4 (%) **	All
Gas composition: iC_4 (%) **	All
Gas composition: CO_2 (%) **	All
Gas composition: N_2 (%) **	All
Gas composition: H_2S (%) **	All
Mole weight (lb/mol)	All
Gas gravity	All (optional)
Density	Density method

NB: When the gas gravity value is available, there is no need to enter the values of the gas composition.

In addition to the input parameters to be given by the user, the following standard parameters were used behind-the-scenes.

Table 6: Standard parameters

Gas Component	**Molar mass (g/mol)**
C_1	16.04246
C_2	30.06904
C_3	44.09562
nC_4	58.1222
iC_4	58.1222
CO_2	44.0095
N_2	28.0134
H_2S	34.08088

The Integrated Development Environment (IDE) used for the development of the software include;

- **Scarlet Macaw (1.5.14)**
- **Adobe Dreamweaver (2015 Release)**

Scarlet Macaw is a windows software used for website prototyping. Because the software to be developed is based on web technologies, it was used to design the user interface of the software. The programming languages used in Scarlet Macaw include:

HTML5 – Short for HyperText Markup Language 5, HTML5 was used for designing the basic layout and structure of the software (a prototype), which was to be transferred to Adobe Dreamweaver for further work.

CSS3 – Short for Cascading Style Sheet 3, CSS3 was used in the beautification of the software, for positioning and basic animations.

JAVASCRIPT – JAVASCRIPT was used for advanced animations in Scarlet Macaw.

After work was done in Scarlet Macaw, the code for the software was transferred into Adobe Dreamweaver 2015 Release. Adobe Dreamweaver, developed by Adobe Systems is a software used for advanced web design, client-side and server-side programming. It was used to code the **SenEngine**, which is the system that controls the calculation, coordination and final presentation of the software.

The programming languages used in Adobe Dreamweaver include the following:

HTML5 – HTML5 was used for further refinement of the prototype that had been created in Scarlet Macaw.

JAVASCRIPT – JAVASCRIPT was used in Adobe Dreamweaver for advanced animations, calculations and manipulations. It was also used for communicating with an online server via PHP.

PHP – Short for PHP Hypertext Preprocessor, PHP was used to manipulate values in an online database on a remote server. It was also used to create the license validation system.

MYSQL – Short form for Microsoft Structured Query Language, MYSQL was used in conjunction with PHP to connect with the database and insert, retrieve and update values.

The software was created to use a combination of charts, tables and numerical values to display calculated results.

The **SenEngine** that powers the software has, built into it, a unit conversion system that handles all the unit conversion needs for the software. This system was developed based on the unit conversion chart developed.

To ensure responsiveness and fluidity of operation, while the software was being developed, it was constantly tested on various desktop computers and mobile devices. Various bugs and errors were encountered while testing. These errors ranged from malfunctioning user interface, to damage due to wrong code and so on. The errors were fixed as soon as they were corrected, in order to avoid pile up of errors. Also, to fine tune the software, an initial simulation was carried out using experimental data gotten from literature. The results gotten were used to generate the accuracy scale, which is then used in the output section of the software. The accuracy scale so generated is given by;

Decreasing Accuracy

Temperature models, Correlations and Methods	Pressure models, Correlations and Methods
Motiee	Ostergaard
Wilcox et al	Molar mass method
Sun et al	Wilcox et al
Safamirzael	Modified Makogon
Towler et al	Safamirzael
Berge	Hammerschmidt et al
Hammerschmidt	Density method
Kobayashi et al	

As the software was created using web technologies, it had to be hosted on a remote network. After this was done, the domain for the software (i.e. the web address through which the software can be accessed) was created. The domain is given as http://bit.ly/sensim. For testing purposes, any user can login with user id as 'betatester' and security key as '12345678'.

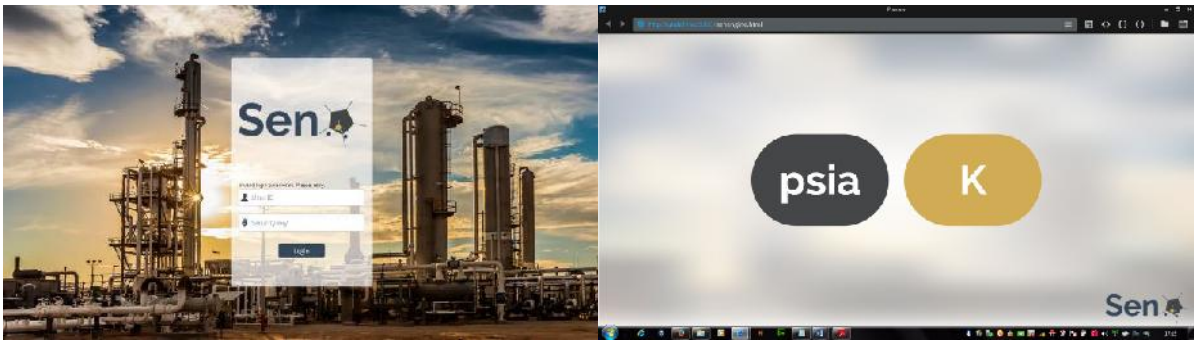
The Sen Algorithm

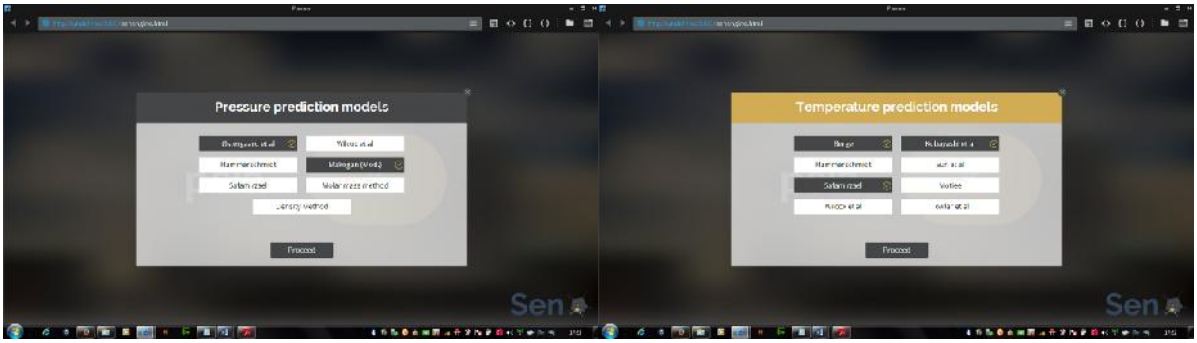
Step	Details
Step 1	Ask the user to input User ID and Security key.
Step 2	Check inputted User ID and Security key against database. If a match is found, allow user to use the software. If not, ask the user to input licensing details again.
Step 3	Ask the user to select a task from the task pane.
Step 4	Based on the selected task, display a list of available models, correlations and methods to use in the simulation.
Step 5	When the user has selected all the models he wants to use, and has clicked the 'proceed' button, display the data input page. On this page, display the input boxes for the required data based on the selected task.
Step 5	When user clicks the button to begin solve, check if the required data has been filled by the user. If it has, begin to calculate temperature or pressure values based on the selected models, correlations or methods. If not, ask user to input the missing data.
Step 6	Check the selected models, correlations, and methods on the accuracy scale. Display the results calculated using the model, correlation or

	method with the highest rank on the accuracy scale, on the main results display page.
Step 7	Smartly pick a range of values based on the inputted pressure or temperature value, while setting the inputted pressure or temperature to be the median value. With these values, rerun the simulation using the selected models.
Step 8	With the results gotten from the re-run simulation, plot a Hydrate formation Pressure vs Temperature chart. Also, insert these values into the hydrate P/T table.

3. RESULTS AND DISCUSSION

After development, the following screenshots of the software were taken on both desktop workstations and a mobile device.





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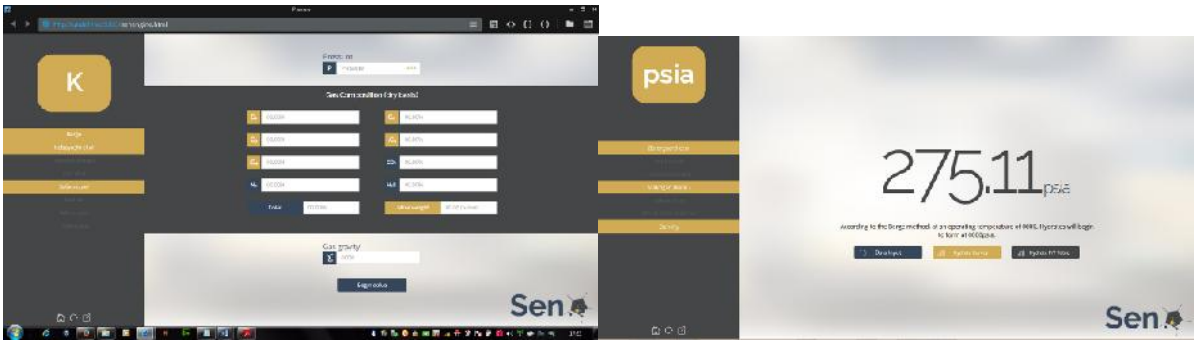

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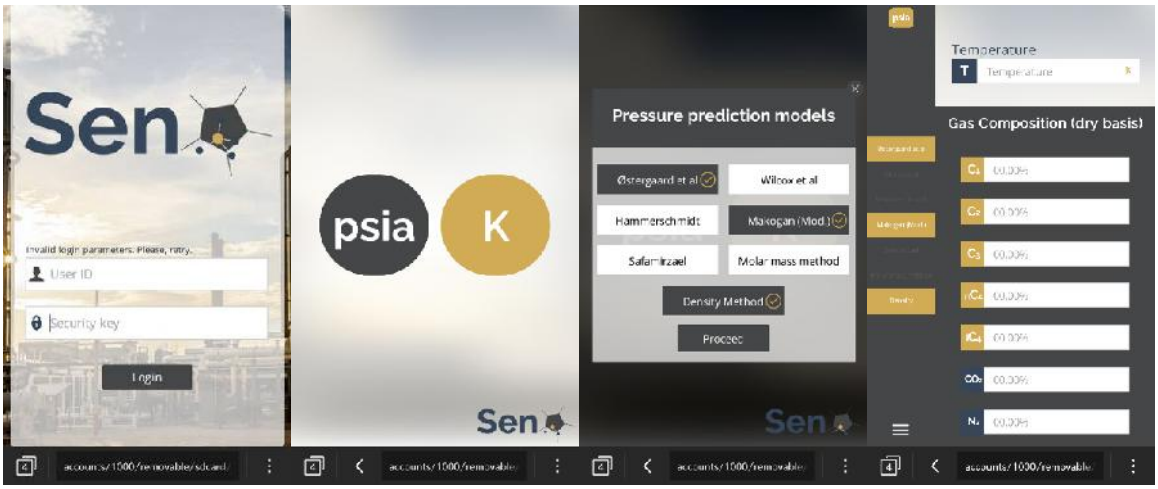

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Figure 1- Screenshots of the software taken on both desktop workstations and a mobile device.

3.1 Case Study: Using experimental data gotten from literature.

Experimental data gotten from literature was used to generate the accuracy scale. It was also used to verify the functionality of the fully deployed software.

The accuracy scale used by the software to display the default calculated value on the results display page. To generate this scale, the following experimental data gotten from literature (ADISASMITO and SLOAN) was used.

Table 7: Experimental Data for Accuracy Scale

Gas Component	Value(%)
Gas composition: C_1 (%) **	76.62
Gas composition: C_2 (%)**	11.92
Gas composition: C_3 (%) **	6.91
Gas composition: nC_4 (%) **	1.82
Gas composition: iC_4 (%) **	2.66
Gas composition: CO_2 (%) **	0.03
Gas composition: N_2 (%) **	0.02
Gas composition: H_2S (%) **	0.02

Experimental Pressure (MPa)	Experimental Temperature (K)
1.4	282.16

Based on these data, the software was able to calculate the predicted temperature and pressures. Also, the same experimental data was fed into a commercial software, Hydrate plus, used for the same purpose. The results of using both **Sen** and **Hydrate plus** are shown in Tables 8a and 8b for Temperature and Pressure respectively.

Table 8a: Predicted Hydrate Temperature conditions for Sen and Hydrate plus

Model, Correlation or Method	Calculated temperature values(K) from Sen	Hydrate Plus method	Calculated temperature values(K) from Hydrate plus
Motiee	280.18	Method I	283.22
Wilcox et al	280.02	Method II	281.77
Sun et al	284.39		

Safamirzael	279.83
Towler et al	279.66
Berge	278.99
Hammerschmidt	277.85
Kobayashi et al	255.52

Table 8b: Predicted Hydrate Pressure conditions for Sen and Hydrate plus

Model, Correlation or Method	Calculated pressure values(MPa) from Sen	Hydrate plus Method	Calculated pressure values (MPa) from Hydrate Plus
Ostergaard	1.60	Method I	1.26
Molar mass method	1.12	Method II	1.46
Wilcox et al	1.77		
Modified Makogon	1.80		

When compared with experimental values, the error percentage of the models, correlations and methods were calculated as follows.

$$\%Error = \frac{|Value_{model} - Value_{exp}|}{Value_{exp}} \times 100 \quad (2)$$

The models, correlations and methods were then arranged in ascending order of error percentage. The model, correlation or method with the least error percentage was given the highest rank while the model, correlation, or method with the greatest error percentage was given the lowest rank.

Table 9a: Experimental vs predicted values for Hydrate Temperature

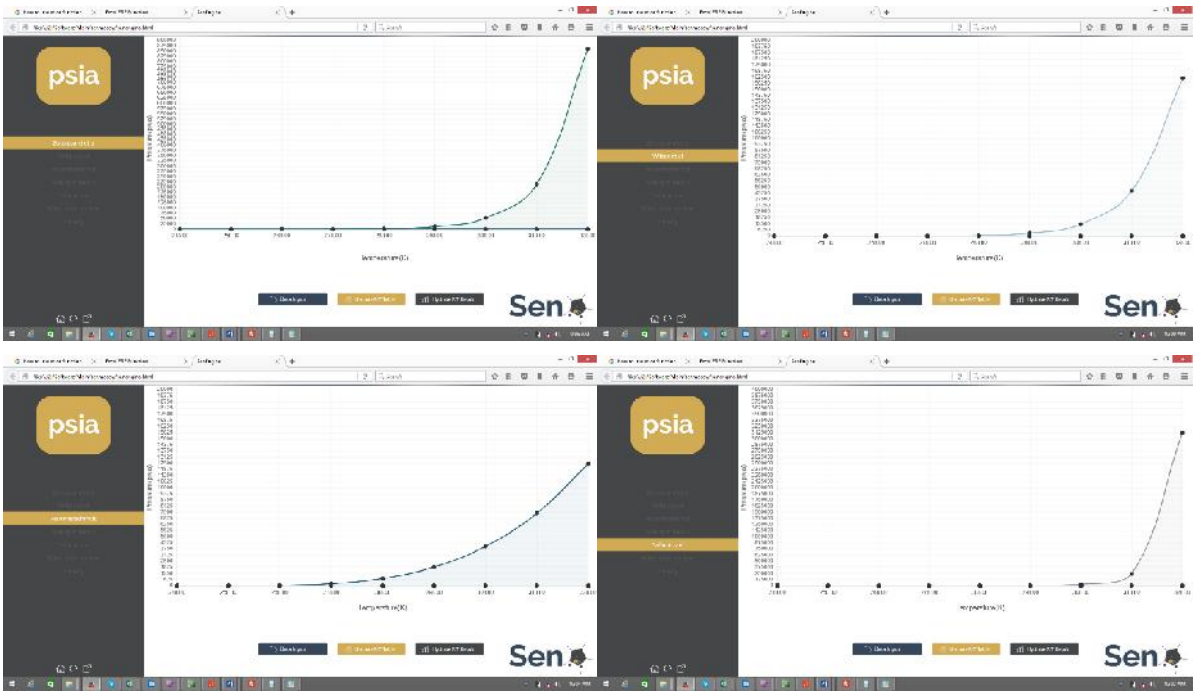
Temperature				
Model	**Predicted Value(K)**	**Experimental Value (K)**	**Error (%)**	**Rank**
Motiee	280.18	282.16	0.70	1
Wilcox et al	280.02	282.16	0.76	2
Sun et al	284.39	282.16	0.79	3
Safamirzael	279.83	282.16	0.83	4
Towler et al	279.66	282.16	0.89	5
Berge	278.99	282.16	1.12	6
Hammerschmidt	277.85	282.16	1.53	7

Kobayashi et al	255.52	282.16	9.44	8

Table 9b: Experimental vs predicted values for Hydrate Pressure

Model	Predicted Value(psia)	Experimental Value (psia)	Error (%)	Rank
Ostergaard	1.60	1.4	13.93	1
Molar mass method	1.12	1.4	19.76	2
Wilcox et al	1.77	1.4	26.27	3
Modified Makogon	1.80	1.4	28.84	4

Some of the hydrate curves generated by **Sen** are shown below.



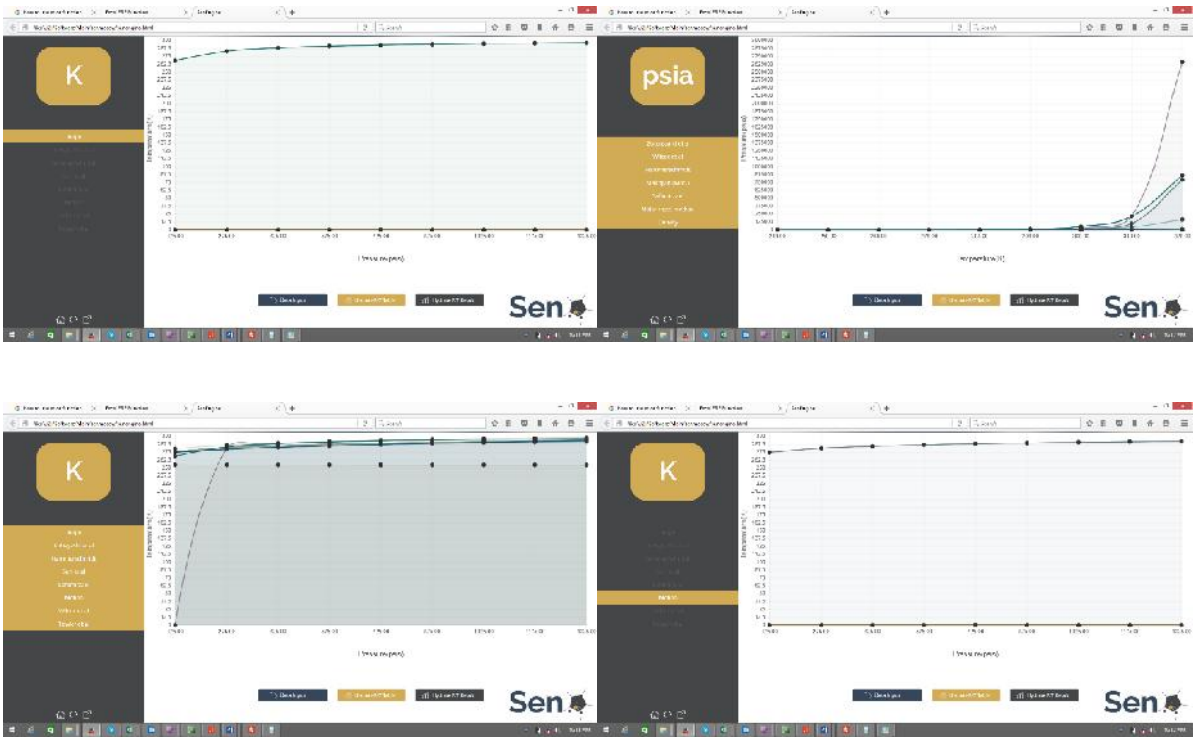


Figure 2- Hydrate plots generated from the software

From the plot generated, it can be seen that various models, correlations and methods have varying accuracies when simulating hydrate formation conditions. Also, it can also be inferred that operating in conditions to the right of the hydrate formation curve (Pressure versus Temperature) will prevent the formation of gas hydrates. The moment the line is crossed, and operations move to the left of the curve, gas hydrates begin to form. Also, on the Temperature versus Pressure hydrate formation curve, operating at conditions that fall to the right will cause hydrates to start to form.

Also, from comparison with the values gotten from commercial software, Hydrate plus, it can be seen that the software works very well and can be used instead of the more expensive counterparts.

4. CONCLUSION

Solids, in general, constitute a big nuisance in the oil and gas industry. Hydrates in particular, are a costly problem when they plug oil and gas pipelines. They reduce the flow rate, increase pressure drop drastically and cause large economic problems for the company.

The best way to tackle this hydrate problem is to operate below the formation conditions. To predict the formation pressure and temperature, the most effective way is to measure these conditions experimentally for every gas system. Since this is not cost efficient and rather time consuming, the only other alternative tool are correlations. Correlations have been developed to estimate the hydrate formation conditions to a very high accuracy when compared with sample experimental data.

A number of these correlations have been used to develop a software. The software, named Sen, was designed to work on both desktop workstations and mobile devices. It simulates hydrate formation conditions using a robust archive of correlations and data inputted by the user. The software can simulate from two angles;

- when only temperature data is available and
- when only pressure data is available

Although, the latter angle is usually preferred, both methods give very good simulation results. The software generates plots that can be used for further study. On the plots, operating to the right of the curve will reduce the risks of hydrate forming.

In all, the software provides a quick, user-friendly, mobile, accurate and cheap way to simulate hydrate formation conditions and can confidently stand where even other more expensive softwares, which perform the same job or less, cannot stand.

Nomenclature

A,B,C	=	Safamirzael correlation constants
ci	=	correlation constants
Fm	=	ratio of non-hydrate formers to hydrate formers
k	=	Makogon correlation constant
Kvs	=	vapor/solid equilibrium ratio
Mair	=	molecular weight of air
P	=	operating pressure of the system.
Pc	=	critical pressure
si	=	mol fraction of component i in solid phase
T	=	operating temperature of the system.
Tc	=	critical temperature
yi	=	mol fraction of component i in vapor phase
β	=	Makogon correlation constant
Δ	=	difference in properties
γ	=	gas gravity
ρ	=	Density of gas
ω	=	acentric factor

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