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GAS PROCESS SYSTEM EMPIRICAL TOOL FOR PREDICTING HYDRATE FORMATION

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Abstract

The rapid formation of gas hydrates, promoted by typical high pressure/ low temperature operating conditions in deep water installations, is considered one of the most difficult problems with flow assurance. Understanding the conditions for the formation of hydrates is necessary to overcome the problems associated with hydrates. Ideally, the conditions for the formation of gas hydrates are determined experimentally in the laboratory; but this data is not always available. Therefore, correlation is used to determine the conditions for gas hydrate formation. Several models have been proposed that require more complex and longer computations to predict the conditions for the formation of gas hydrate over the years. In this study, it is crucial to develop a reliable and easy-to-use method for oil and gas practitioners'. The proposed correlation extends over a wide range of pressure (2000 to 25000kPa) and molecular weights (16 to 27). Consistent and accurate results of the proposed pressure range, temperature, and molecular weight are presented. Statistical error analysis is used to appraise the efficiency and accuracy of the correlation coefficient for estimating the formation of gas hydrate. This will guide designer and operator to select the optimal correlation for a particular application.

Keywords: Gas hydrate; Gas process system; hydrate prediction; correlations; Instability in Pipeline.

1. Introduction

Natural gas is becoming more important all over the world, as an important source of energy and as raw material for the processing industry. The increase in demand for natural gas in the energy matrix throughout the world has caused a great demand for exploration and production of the offshore proven reserves. The offshore industry is developing from shallow water fixed platforms to the development of deepwater field with floating facilities. This has created new problems for the prevention of hydrate during transportation and processing of natural gas in the subsea environment [1]. Hydration requires a condition that consists of having water in the pipeline; the pressure must be high, with a temperature that is low and the existence of methane, carbon dioxide [2].

The exploration deeper waters have rendered transporting of hydrocarbons and water mixture to onshore facilities through a single multiphase pipeline the only practical way. Currently, the responsibilities of processing the fluids on-site are considered very strict due to the cost of building and maintaining the complex offshore platforms with processing facilities [3]

In 1934, Hammerschmidt [4] discovered that the obstacle to the gas pipeline was not due to the formation of ice, but due to the formation of natural gas clathrate hydrate. History had it that, this discovery was the determining factor in raising a practical interest for oil and gas companies [1]. Gas hydrates are one of the serious economic and safety challenges in oil and gas industry in the exploration, extraction, production, transportation, and processing of natural gas and liquid. Hydrate formation can block pipelines and processing equipment. Formation of gas hydrates can occur during process operation and shut in periods. It can be very complex, difficult and expensive to remove them.

Hydrate plugging of hydrocarbon production conduits causes serious operational problems that can lead to significant and considerable economic losses. The ability to model and predict the emergence of hydrate plugging would help to improve design and operation of the facility in order to reduce the extent of such events. Methods for preventing and eliminating hydrates have been studied and developed using various approaches [5].

The purpose of this study is to develop a tool that can predict the conditions for the production of hydrates. The predicted conditions are compared with published used empirical methods.

2. Literature review

There are different approaches which can be applied to mitigation a plug of gas hydrates such as dehydration, heat management, and chemical inhibition [6]. Three methods have been used in the field; and among these methods, chemical inhibition is the most common to prevent and reduce gas hydrate formation [7].

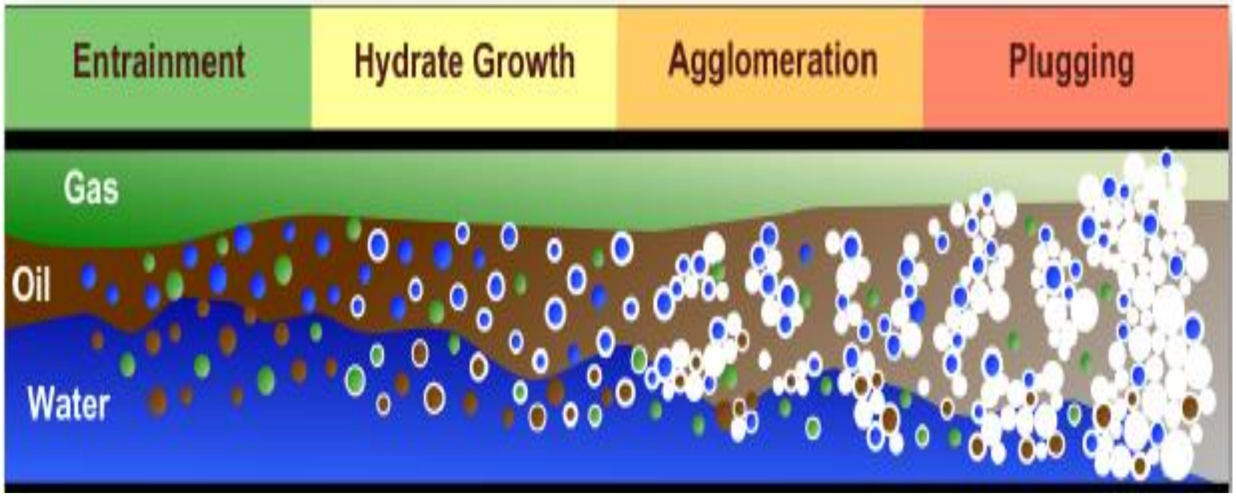


Figure 1. A conceptual model for hydrate formation (*Source: Turner* [8])

When speaking about the formation of hydrate, the vital elements relates to economics [9]. Since these problems can interfere with the process operations; thus, its higher value is directly related to economics. According to Obanijesu *et al.* [10], intensifying and improving research to explore more opportunities to avoid, identify, prevent or eliminate hydrate occurrence in the pipeline is a worthwhile investment as a consequence is catastrophic.

According to Huijie [11], the thermodynamics of hydrate formation has been extensively studied by researchers for many years, and data on some thermodynamic inhibitors have been acquired. These data can be useful for testing industrial design applications and predictive models. Several methods have been published for predicting hydrate formation in the presence of inhibitors.

The methods are mostly based on using cubic equations of state for the fluid phase. The statistical associated fluid theory has been investigated extensively since proposed and is very advantageous over traditional cubic equation of state. Salam *et al.* [12] in their study on prediction of hydrate formation conditions, outline some of the improvement that has been done on methods of hydrate formation prediction.

Bhangole *et al.* [13] extended van der Waals and Platteuw's to predictions of hydrate formation in the reservoir. They developed a model for pore-freezing that can be used to envisage gas hydrate equilibrium for pure CH_4, CO_2, and mixed CH_4 –CO_2 system for any pure size distribution. They used Equation of State developed for bulk gas hydrate equilibrium conditions based on van der Waals–Platteuw's model. The model was implemented with FORTRAN code, and it can calculate gas hydrate saturation equilibrium at any given pressure, temperature; and also calculate hydrate equilibrium condition for salt in a mixed CH_4-CO_2 gas hydrate system. Another model applied to an offshore and deep water environment is the work of Mohammadi and Tohidi [14]. They developed a model to predict the hydrate free zone in mixed salt and chemical inhibitors for deep water and offshore applications.

Prevention of formation of hydrate is of paramount important because this definitely saves 5-8% of total plant cost and several methods have been adopted in the prediction of hydrate free zone when designing transportation of gas. Gas has a different composition which makes their behavior and hydrate formation condition differs [12].

To optimize the cost of cleaning or remediation, the accuracy of forecasting conditions for the production of natural gas hydrates is very vital. The best way to determine the conditions for the formation of hydrate is to experimentally measure the formation of the composition of interest at high pressure and low temperature.

From a practical view, it will be almost impossible to establish the gas hydrate formation conditions for an infinite number of specific gas composition needed. Also, the experimental activities are both expensive and time consuming relative to industrial needs. It is necessary to form the prediction methods that will help for interpolation between the measured ones. Therefore, an accurate and simplified model is desirable for predicting natural gas hydrate conditions [19].

Several empirical correlations to predict hydrate formation conditions have been published by different researchers' [15-18]. It is obvious that a model that can predict hydrate plugging in a given production system will become a valuable tool. The results would be useful in both design and operations.

3. Methodology

In this study, correlations to determine the conditions for gas hydrate formation based on gas gravity method was programmed. The tool was assessed to match existing experimental data published in the literature under different system conditions. In the industry, precise forecasting tools are now required for hydrate plugging. Knowledge of the risk of hydrate plugging with greater accuracy will help to develop a system and also provide operational support.

In this study, over 30 data point was collected in three-phase equilibria of various gas systems. Data also include those published in some literature, and a review of the literature showed that pressure, temperature and specific gravity are common correlation variables.

3.1. Development of the new Tool

An accurate and simplified model is desirable for predicting natural gas hydrate. Among the thermodynamic models in the literature, only a few examples can predict reliable hydrate-formation conditions for the complex systems with a minimal error.

The proposed model was developed using the statistical analysis software and experimental data points (from literature) to find the top correlations midst the variables. Multiple regressions provided a powerful method used to correlate the formation temperature of hydrate with pressure, specific gravity and water-vapour pressure.

3.2.1. The new and existing correlations

In this study, the following method was used to develop this new correlation. Firstly, the hydrate formation temperatures were correlated as a function of pressure for various molecular weights. The calculated coefficients of these polynomials are correlated as a function of molecular weight. Already developed polynomials were applied to calculate the new coefficients so as to predict the condition for hydrate formation for natural gases.

Bahadori et al. correlation [20]

$$\ln T = a + b\left(\frac{1}{P}\right) + c\left(\frac{1}{P}\right)^2 + d\left(\frac{1}{P}\right)^3 \qquad (1)$$

where, $a = A_1 + B_1M + C_1M^2 + D_1M^3$; $b = A_2 + B_2M + C_2M^2 + D_2M^3$; $c = A_3 + B_3M + C_3M^2 + D_3M^3$; $d = A_4 + B_4M + C_4M^2 + D_4M^3$.

The coefficients are given in the literature [20] .

Kobayashi correlation* [21] *,

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

where: $A_1 = 2.7707715 \text{ X } 10^{-3}$; $A_2 = -2.782238 \text{ X } 10^{-3}$; $A_3 = -5.649288 \text{ X } 10^{-4}$;
$A_4 = -1.298593 \text{ X } 10^{-3}$; $A_5 = 1.407119 \text{ X } 10^{-3}$; $A_6 = 1.785944 \text{ X } 10^{-4}$
$A_7 = 1.130284 \text{ X } 10^{-3}$; $A_8 = 5.9728235 \text{ X } 10^{-4}$; $A_9 = -2.3279181 \text{ X } 10^{-4}$
$A_{10} = -2.6840758 \text{ X } 10^{-5}$; $A_{11} = 4.6610555 \text{ X } 10^{-3}$; $A_{12} = 5.5542412 \text{ X } 10^{-6}$
$A_{13} = -1.4727765 \text{ X } 10^{-5}$; $A_{14} = 1.3938082 \text{ X } 10^{-5}$; $A_{15} = 1.4885010 \text{ X } 10^{-6}$
Also, $r_g = M_g/29$

***Berg correlation* [22]**

$For\ 0.555 \le r_g < 0.58$

$$T = -96.03 + 25.37 \ln P - 0.64(\ln P)^2 + \frac{r_g - 0.555}{0.025}\left[80.61P + \frac{1.16 \text{ X } 10^4}{P + 596.16} - \left(-96.03 + 25.37 \ln P - 0.64(\ln P)^2\right)\right] \tag{3}$$

$For\ 0.58 < r_g \le 1.0$

$$T = \left. 80.61P - 2.1 \text{ X } 10^4 - \frac{1.23 \text{ X } 10^3}{r_g - 0.535} - \left(1.23 \text{ X } 10^4 + \frac{1.71 \text{ X } 10^3}{r_g - 0.509}\right) \middle/ P - \left(-260.12 - \frac{15.18}{r_g - 0.535}\right) \right. \tag{4}$$

3.3. This study correlation

The hydrate formation temperature ***T*** is a functional relationship between the operating pressure ***P*** and gas gravity γ_g of the mixture. In this study non-linear regression analysis was used to develop such relationship. The model utilized is

$$\boldsymbol{T = a_1 + {}^{p}/\gamma_g} \ \ldots\ldots\ldots\ldots \tag{5}$$

The least-square minimization to solve equation 5 was formulated as follow. An objective function ***f(c)*** is defined by the error function given as:

$$\boldsymbol{f(c) = \sum_{i=1}^{N} r_i^{\,2}} \tag{6}$$

where the vector c = [a_1, a_2, a_3 …., a_{16}] represents 16 independent variables called regression constants. The elements or ***r*** are dependent variables called residues.

The residue expresses the relative difference between the experimental hydrate formation temperature ***(p, γ_g, T)*** and the study's simulated hydrate temperature ***(p, γ_g, T)***. The residue for ***N*** number of data points is defined by

$$\boldsymbol{r_i = \frac{\left[\check{T}(p,\gamma_g,c)_i - (p,\gamma_g,T)_i\right]}{(p,\gamma_g,T)_i}, i = 1, 2, 3, \ldots, N} \tag{7}$$

The requirement non-linear optimization is to find the update

$$\boldsymbol{c_{i+1} = arg\, Min\{F(c)\}} \tag{8}$$

To ensure convergence to a realistic constant, the non-linear optimization is controlled to enforce the descent direction such that the objective function at an iteration stage is greater than the objective function at the next iteration stage. That is,

$$\boldsymbol{f(c_{k+1}) < f(c_k)} \tag{9}$$

Once, the descent direction is located, a step size that would give a good decrease in the objective function is chosen. Thus, the derivation of the hydrate formation model for this study;

$$\boldsymbol{\ln T = A_1 + A_2\gamma_g + A_3{\gamma_g}^2 + A_4{\gamma_g}^3 + A_5P^{-1} + A_6\gamma_gP^{-1} + A_7{\gamma_g}^2P^{-1} + A_8{\gamma_g}^3P^{-1} + A_9P^{-2} + A_{10}\gamma_gP^{-2} + A_{11}{\gamma_g}^2P^{-2} + A_{12}{\gamma_g}^3P^{-2} + A_{13}P^{-3} + A_{14}\gamma_gP^{-3} + A_{15}{\gamma_g}^2P^{-3} + A_{16}{\gamma_g}^3P^{-3}} \tag{10}$$

The coefficients of A_1 to A_{16} are given in table 1.

Table 1 Tuned coefficients used to predict hydrate formation in this study

Coefficients	120kpa<P<40000 kPa $\gamma_g > 0.79$	120kpa<P<5000 kPa $\gamma_g < 0.79$	5000kpa<P<40000 kPa $\gamma_g < 0.79$
A1	6.4185	-4.181	7.096
A2	-2.552493	42.688	-6.322
A3	2.991437	-61.17434	9.5033
A4	-1.158472622	28.998521	-4.6997603
A5	-8642.6	45280	-125800
A6	29705.57	-198998	550710
A7	-34452.406	287958.4	-801136.6
A8	13279.8105	-137602.738	385346.2
A9	11590000	-83170000	921900000
A10	-40165000	365400000	-4068700000
A11	46549350	-529914100	5955962000
A12	-17899087.1	253645600	-2882535910
A13	-4020000000	5858000000	-2.105E+12
A14	13893900000	-28022700000	9.3177E+12
A15	-16004230000	43176940000	-1.36864E+13
A16	6124077900	-21652554200	6.64571E+12

3.4. Error analysis

The statistical error analyses were applied to test the performance and accuracy of the proposed hydrate formation correlations and table 4 data was used for this process. The criteria adopted in this study was average percent relative error.

Table 2. Experimental data used for comparison (*Source: Bahadori and Vuthaluru* [23])

Pressure (kPa)	Gas molecular meight	Experimental temperature	Pressure (kPa)	Gas molecular meight	Experimental temperature
3157.8	16	274.82	4757.38	20.3	288.71
4136.85	16	277.59	23442	20.3	297.04
5515.81	16	280.37	496.42	23.2	272.04
1723.69	17,4	277.59	1930.53	23.2	283.15
3309.48	17.4	283.15	11721.09	23.2	294.26
6756.86	17.4	288.71	758.42	26.1	277.59
18098.74	17.4	294.26	1585.79	26.1	283.15
2688.96	18,85	283.15	17926.37	26.1	297.04
14134.25	18,85	294.26	413.69	29	274.82
827.37	20.3	274.82	1344.48	29	283.15
2344.2	20.3	283.15	3033.69	29	288.71

4. Results and discussion

The type of hydrate data applied is the formation pressures and temperatures. This type of data is most significant for natural gas applications. We summarized the most accessible experimental data of this type, which were compiled as a basis for comparing reputable correlations such as Berg, Bahadori, and Kobayashi correlations. The experimental data on hydrate formation conditions were compared with those calculated by the proposed correlation and included in the program.

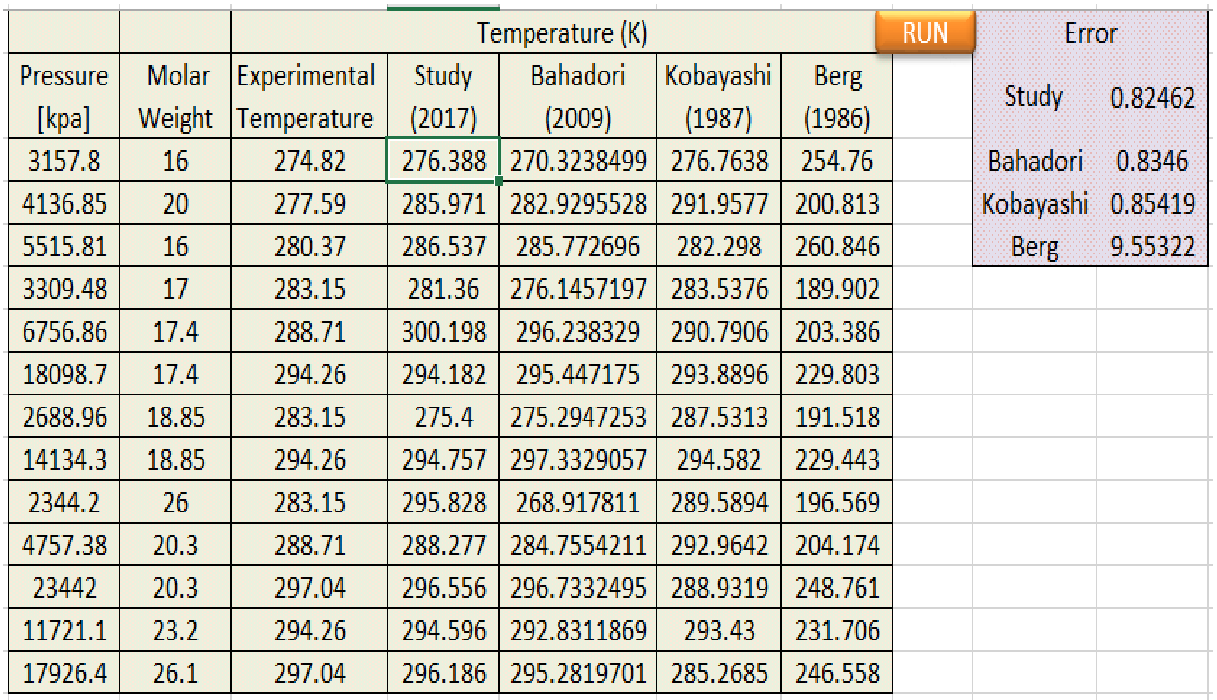
Using software for statistical analysis, we applied a regression model to the experimental data points to find the best correlations among the variables. The data points include several samples ranging from gas mixtures to non-hydrocarbon and pure hydrocarbon components. To confirm the correlations accuracy and compare the predicted results with the experimental data, a statistical error analysis was used for these correlations.

The resulting predictive tool will improve the design process of production facilities and estimate hydrate plugging risk in any specific scenario.

4.1. Comparison of new correlation with existing correlations

As a basis for comparison, several authoritative correlations were selected; Berg [22], Kobayashi *et al.* [21], and Bahadori *et al.* [23]. Using the same hydrate conditions, the results of the proposed new correlations were compared with the existing correlation. Figure 2 shows a comparison of the proposed method and existing correlation results. It was deduced that the proposed correlation shows a higher yield in comparison with other existing correlations and estimates the conditions for the formation of hydrates.

Figure 2. Proposed new correlation and existing correlations

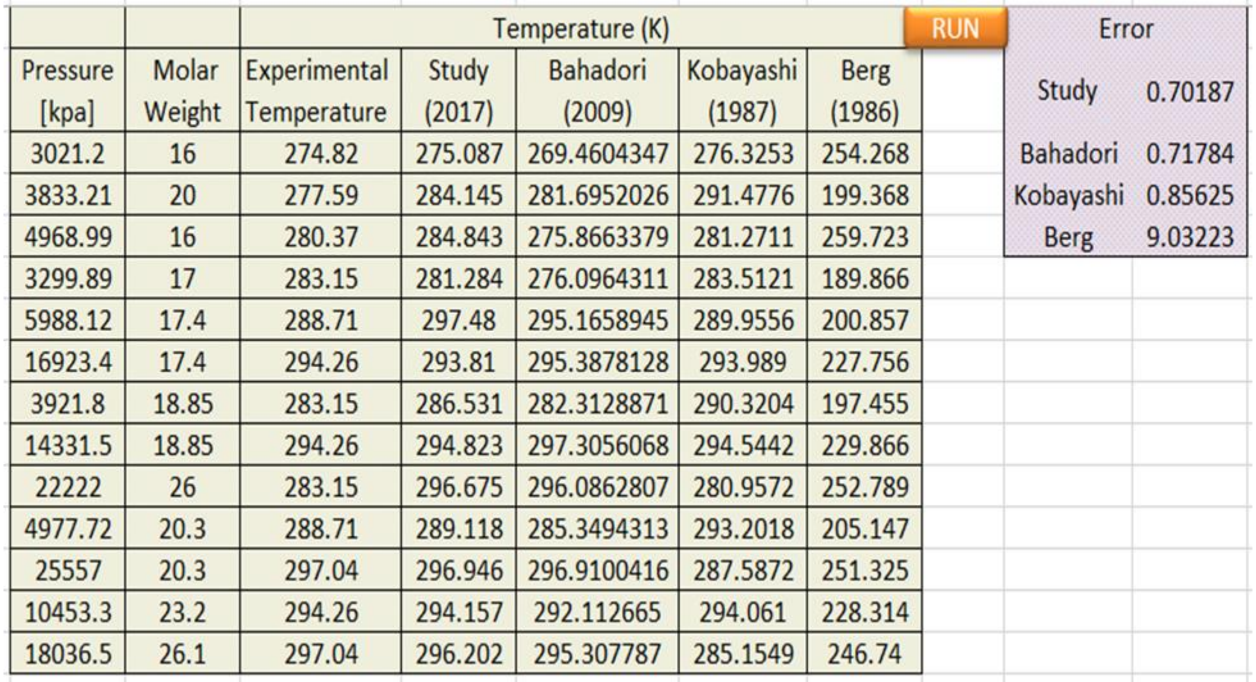


		Temperature (K)				
Pressure [kpa]	Molar Weight	Experimental Temperature	Study (2017)	Bahadori (2009)	Kobayashi (1987)	Berg (1986)
3157.8	16	274.82	276.388	270.3238499	276.7638	254.76
4136.85	20	277.59	285.971	282.9295528	291.9577	200.813
5515.81	16	280.37	286.537	285.772696	282.298	260.846
3309.48	17	283.15	281.36	276.1457197	283.5376	189.902
6756.86	17.4	288.71	300.198	296.238329	290.7906	203.386
18098.7	17.4	294.26	294.182	295.447175	293.8896	229.803
2688.96	18.85	283.15	275.4	275.2947253	287.5313	191.518
14134.3	18.85	294.26	294.757	297.3329057	294.582	229.443
2344.2	26	283.15	295.828	268.917811	289.5894	196.569
4757.38	20.3	288.71	288.277	284.7554211	292.9642	204.174
23442	20.3	297.04	296.556	296.7332495	288.9319	248.761
11721.1	23.2	294.26	294.596	292.8311869	293.43	231.706
17926.4	26.1	297.04	296.186	295.2819701	285.2685	246.558

Error	
Study	0.82462
Bahadori	0.8346
Kobayashi	0.85419
Berg	9.55322

From the study, we can agree that empirical expressions are a vital tool in predicting gas hydrate stability in different systems. Figure 2 illustrates that the proposed new correlation gives the accurate results in all pressure ranges and specific gas gravities when the temperature at which hydrate will form a certain pressure is predicted. This consistency was not matched by any of the correlations used in the survey range.

Figure 3. Proposed new correlation and existing correlations - another hydrate predictions



		Temperature (K)				
Pressure [kpa]	Molar Weight	Experimental Temperature	Study (2017)	Bahadori (2009)	Kobayashi (1987)	Berg (1986)
3021.2	16	274.82	275.087	269.4604347	276.3253	254.268
3833.21	20	277.59	284.145	281.6952026	291.4776	199.368
4968.99	16	280.37	284.843	275.8663379	281.2711	259.723
3299.89	17	283.15	281.284	276.0964311	283.5121	189.866
5988.12	17.4	288.71	297.48	295.1658945	289.9556	200.857
16923.4	17.4	294.26	293.81	295.3878128	293.989	227.756
3921.8	18.85	283.15	286.531	282.3128871	290.3204	197.455
14331.5	18.85	294.26	294.823	297.3056068	294.5442	229.866
22222	26	283.15	296.675	296.0862807	280.9572	252.789
4977.72	20.3	288.71	289.118	285.3494313	293.2018	205.147
25557	20.3	297.04	296.946	296.9100416	287.5872	251.325
10453.3	23.2	294.26	294.157	292.112665	294.061	228.314
18036.5	26.1	297.04	296.202	295.307787	285.1549	246.74

RUN

Error	
Study	0.70187
Bahadori	0.71784
Kobayashi	0.85625
Berg	9.03223

5. Conclusion

The proposed correlation holds for a wide range of temperatures, pressure (2000 to 25000kPa) and molecular weights (16 to 27). Consistent and accurate results of the proposed pressure range, temperature, and molecular weight were presented. For all conditions, statistical error analysis was used to estimate the efficiency and accuracy of the correlation coefficient for estimating the formation of gas hydrate. The tool will guide the operator to select the best correlations for their particular applications. The results of the hydrate formation conditions estimated by the previous correlations were compared with the experimental data. From this comparative study, we can conclude that;

1. The current study and Kobayashi correlation results close to the experimental results at the investigated range of temperature, pressure and molecular weight.
2. The composition of the gas system plays a very important role in determining the temperature or pressure of hydrate formation. This means that two gas systems with the same specific gravity can form hydrate under different conditions.
3. The predicted results are in most cases are approximately the same with the experimental data; thus, the gas hydrate model developed in this study can be used to establish a flow assurance strategy.

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