



Developing a simple and accurate correlation for initial estimation of hydrate formation temperature of sweet natural gases using an eclectic approach



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ABSTRACT

Formation of natural-gas hydrates which are crystalline solids composed of water and gas has been a problem to the natural-gas industry in production, processing facilities and transportation and caused substantial economic risks. Therefore, in order to overcome hydrate-related issues, it is important to predict the temperature and pressure under which gas hydrates form. Since the start of hydrates studies, several correlations have been proposed to predict their formation temperature (HFT) under circumstances of different known data such as indeterminate gas composition. However; most of these models have failed to give a good prediction of HFT over a wide range of pressure and gas gravity variation; furthermore, the presented models often require complicated and long computations. The purpose of this study is to present an accurate and simple correlation with only one set of coefficients, achieved using a novel fitting method which can apply for the large range of temperatures (273–299 K), pressures (350–30,000 kPa) and molecular weights (16–29 g/mol). The new proposed correlation shows to be the most accurate one among all the available correlations. The coefficient of determination (R^2) of this new formula is 0.9874 and the maximum absolute error is 1.3% for 545 experimental data. The results of the correlation are compared with seven widely used correlations along with a neural network model for predicting hydrate formation conditions using back propagation.

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1. Introduction

Gas hydrates (gas clathrates) are low energy, high pressure solid compounds of natural-gas molecules that are engaged within a crystal structure composed of water molecules (Sloan and Koh, 2007). Hydrate is similar in appearance to water ice and also dry ice and many but not all of its physical properties are similar to ice.

Gas hydrate formation is a concomitant process requiring the presence of both the host and guest molecular species. The most common guest molecules are methane, ethane, propane, isobutane, normal butane, nitrogen, carbon dioxide and hydrogen sulfide, of which methane occurs abundantly in natural hydrates (Bahadori and Vuthaluru, 2009).

Various models have been presented to predict hydrate formation temperature. These models can be classified into six major groups.

1.1. *K*-value method

The first method uses *K*-value which was initiated by Wilcox et al. This model utilizes the empirically estimated vapor–solid equilibrium constants to predict hydrate formation conditions (Wilcox et al., 1941). The method treats solid hydrate solutions similarly to an ideal liquid solution. He also defined the equilibrium ratio of a component in *Lw*–*H*–*V* equilibrium by the following equation (Katz et al., 1942):

$$K_i^{vs} = \frac{y_i}{s_i} \quad (1)$$

where y_i is the mole fraction of component i in the vapor phase and s_i is the mol fraction of component i in the solid phase. Therefore, the *K*-value charts are used to calculate the hydrate formation temperature or pressure of three-phase solution in a manner that satisfies the following equation:

$$\sum_{i=1}^n \frac{y_i}{K_i^{vs}} = 1 \quad (2)$$

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This method was improved by Katz and co-workers (Katz et al., 1942). Mann et al. presented new *K*-charts that could provide accurate results even for cases containing non hydrocarbon gasses such as CO₂, N₂ and H₂S which might have caused inaccurate results by the original Katz-chart (Elgibaly and Elkamel, 1998; Mann et al., 1989).

1.2. Gas gravity chart

The second method is the gas gravity plot developed by Katz (1945). The plot relates the hydrate formation pressure and temperature with gas gravity. This method is a simple graphical technique that may be useful for an estimate of hydrate formation condition (Ghiasi, 2012).

1.3. Charts of permissible expansion

The third method uses the charts of permissible expansion that a natural gas can undergo without risk of hydrate formation. These charts were redrawn with the aid of gas gravity charts using Joule–Thomson cooling curves (Elgibaly and Elkamel, 1998).

1.4. Statistical–Mechanical models based on model of Van der Waals and Platteeuw

The fourth method is based on the statistical–mechanical model of Van der Waals and Platteeuw (1952). Various thermodynamic models have been presented to calculate HFT according to Van der Waals and Platteeuw's model (Parrish and Prausnitz, 1972; Nasrifar and Moshfeghian, 2001). The newer models claim to account for solutions, including alcohol and electrolyte (Nasrifar and Moshfeghian, 2001).

1.5. Statistical associating fluid theory (SAFT)

Most conventional equations of state are based on Van der Waals equation which assume molecules to be hard spheres. However, a hard-sphere reference is inappropriate for most fluids, which might contain molecules that are highly nonspherical and associating. In such cases, a more appropriate assumption is one that incorporates both chain length and molecular association. Such an approach was proposed by Chapman et al. and is termed the statistical associating fluid theory (SAFT) (Chapman et al., 1988). This model provides a more complex and also a more precise investigation of the hydrate formation and dissociation of non-spherical hydrates (Abolala et al., 2014; Li et al., 2007).

1.6. Empirical correlations

Most of these correlations use pressure and gas gravity as their input and predict the hydrate formation. Some of the widely used existing correlations are introduced below.

Hammerschmidt presented the following correlation as the results of his experiments which can predict HFT only as a function of pressure, in which pressure is stated in psi and temperature is calculated in °F, (Hammerschmidt, 1934).

$$T = 8.9 * p^{0.285} \quad (3)$$

Due to its simplicity, this correlation does not provide accurate predictions in average temperatures. However, in contrast with most of the other complicated models it doesn't show large deviations from experimental values in elevated temperatures and pressures.

Makogon presented a simple model that can predict hydrate formation pressure when the temperature and gas gravity are known (Makogon, 1981).

$$\text{Log}(P) = \beta + 0.0497(t + kt^2) - 1 \quad (4)$$

His equation can be rearranged to become temperature explicit as given below:

$$t = \sqrt{\frac{\text{Log}(P) + 1 - \beta}{0.0497k} + \frac{0.25}{k^2} - \frac{k}{1}} \quad (5)$$

In which γ is the gas gravity, pressure is in MPa and temperature is in °C (Örs, 2012). k and β can be found from graphical correlations introduced by him, however Elgibaly and Elkamel provided two simple correlations to calculate k and β (Elgibaly and Elkamel, 1998):

$$k = -0.006 + 0.011\gamma + 0.011\gamma^2 \quad (6)$$

$$\beta = 2.681 - 3.811\gamma + 1.679\gamma^2 \quad (7)$$

Berge was convinced that splitting correlations into two parts according to gas gravity could result in a more accurate correlation. He presented the following relationship (Berge, 1986).

For $0.555 \leq \gamma \leq 0.58$

$$t = -96.03 + 25.37 * \ln(P) - 0.64 * \ln(P)^2 + \frac{\gamma - .555}{0.025} * \left(80.61 * P + \frac{1.16 * 10^4}{P + 599.16} - 96.03 + 25.37 * \ln(P) - 0.64 * \ln(P)^2 \right) \quad (8)$$

In equation (8) pressure is used in psi and temperature is calculated in °F.

For $0.58 \leq \gamma \leq 1$

$$t = \frac{-1.23 * 10^4 + 80.61P - 2.1 * 10^4 - \frac{1.22 * 10^3}{\gamma - 0.535} - \frac{1.71 * 10^3}{\gamma - 0.509}}{P - 260.42 - \frac{15.18}{\gamma - 0.535}} \quad (9)$$

The original work (Berge, 1986) stated that pressure should be used in psi and temperature is calculated in °F, however these units provide unstable results and the correlation provides better results when pressure is in kPa and then the temperature is calculated in °F.

Kobayashi et al. developed an empirical equation using fifteen coefficients. The coefficients were determined by a regression method that predicts the hydrate temperatures at given pressures for systems. The correlation was fitted in the temperature range of 34–60 °F, the pressure range of 65–1500 psi, and the gas gravity range from 0.55 to 0.9. It should be noted that in this correlation pressure is used in psi and temperature is calculated in R (Kobayashi et al., 1987).

$$t = 1 / \left[a_1 + a_2 * \ln(\gamma) + a_3 * \ln(P) + a_4 * \ln(\gamma)^2 + a_5 * \ln(\gamma) * \ln(P) + a_6 * \ln(P)^2 + a_7 * \ln(\gamma)^3 + a_8 * \ln(\gamma)^2 * \ln(P) + a_9 * \ln(\gamma) * \ln(P)^2 + a_{10} * \ln(P)^3 + a_{11} * \ln(\gamma)^4 + a_{12} * \ln(\gamma)^3 * \ln(P) + a_{13} * \ln(\gamma)^2 * \ln(P)^2 + a_{14} * \ln(\gamma) * \ln(P)^3 + a_{15} * \ln(P)^4 \right] \quad (10)$$

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