

Original Research

# Investigation of Hydrate Formation Phenomenon and Hydrate Inhibitors

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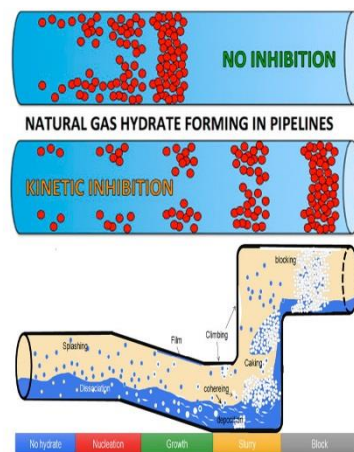
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## ABSTRACT

Natural gas hydrate is a solid crystalline compound formed from the combination of water and gas and is part of the clathrate family. Guest gas molecules are trapped within the cavities of the water network, which is formed by hydrogen bonding between water molecules. They fell. A good example of these gas molecules are compounds smaller than pentane in natural gas such as methane, ethane, propane and carbon dioxide. Gaseous hydrates can be formed by pure gas or a gas mixture consisting of two or more components. They are considered a solid solution in which guest gas molecules and hydrate gases are in contact with the host network (water). Chemical inhibitors are used to prevent gaseous hydrates in gas transmission pipelines. Thermodynamic and kinetic inhibitors are divided into two groups. These inhibitors also differ in the type and extent of use in pipelines.

## GRAPHICAL ABSTRACT



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## Introduction

Hydrate is a water network with a set of empty spaces at short distances. If some of this space is filled with gaseous molecules, hydrate can be considered as a stable solid. In other words, gaseous hydrates are complex crystalline molecules made up of a mixture of water and suitable gaseous molecules. Water molecules form unstable networks due to hydrogen bonds, and gaseous molecules occupy the space between the network gaps. When a small amount of lattice holes is filled, the gaseous hydrate molecules crystallize at temperatures even above freezing. The structure of the hydrated gas will be as follows: Structure I, Structure II and, more recently, Structure H have also been identified. Historically, studies on the phenomenon of gaseous hydrates can be classified into three important periods:

-The first period, from its discovery in 1810 until now, has been to study hydrate as an unknown scientific phenomenon in which water and gas are transported into a solid network.

-The second period, from 1934 until now, has studied the phenomenon of hydrates in the gas transmission industry.

-The third period, from 1960 to the present, began with the discovery of underground hydrate sources in the deep oceans and deep-frozen areas [1-8].

In 1778, Joseph Priestley left the window open as he left the laboratory in the winter evening. Priestley, therefore, is believed to have discovered it thirty years before the hydrate

demon was discovered. The discovery of hydrate by an independent Davy is considered to be the first observation of hydrate [9-13].

Sir Davy Humphrey was the first to notice the formation of chlorine hydrate in 1811 while testing an aqueous solution of chlorine saturated at -40 degrees Fahrenheit.

From 1810 to 1934, studies on hydrates were conducted on the following two main axes:

(1) Identify all the compounds that can form hydrates.

(2) Quantitative description of compounds in terms of percentage composition and physical properties.

During the century since the discovery of hydrate, the number of articles published during this period has been reported to be approximately 40, while in 2000 this number alone reached 400, and this indicates the peak of importance and attention to this issue in the present world. From 1934 onwards, research on hydrates led to ways to prevent this phenomenon from occurring in gas transmission lines, because at that time Hammerschmidt realized that clogged pipelines were due to hydrate formation, not frost. Hydrate has naturally doubled since its discovery in parts of Alaska, Siberia and Canada. These hydrate mineral reserves are recognized as a pristine source of energy and have been noted for the large amount of gas stored in them [14-18].

*Investigation of operating conditions of hydrate formation*

To investigate the operating conditions of hydrate formation (required temperature and pressure), there are different methods, one of the common methods for this purpose is to draw the logarithm of the pressure change curve in terms of gas temperature. In this case, the curve is a straight line the structure of hydrates in the water-gas range is maintained by molecules of solution in the aqueous phase region [19-21]. As a result, H<sub>2</sub>S and CO<sub>2</sub> accelerate the formation of hydrates at high temperatures because they are more soluble in water than most hydrocarbons. Pressure causes the lattice to deform. In the case of pentane and larger molecules, the pressure breaks the lattice and prevents the formation of stable hydrates. Hydrates increase like crystals. They form sediments. In fact, high pressure and low temperature in places such as the cap of a pipe in perforated plates and valves cause sediment, which the maximum force from the variable flow can also prevent the formation of hydrates in these places. So high turbulence can easily alter their actual structure. Hydrocarbon liquids aid this process with a thorough wash. These structures are related to the size of the spaces in the network. This space can easily hold methane, H<sub>2</sub>S and CO<sub>2</sub> and similar molecules. That is, the substitute molecule must be of a suitable size to be able to enter the blue groove [22-28].

There is no industrially larger molecule than isobutane that can be replaced in these gaps. However, in recent experiments, normal butane has been identified as the largest hydrated molecule that can be complexed in a

network of large numbers of small molecules. It should be noted that normal butane is not able to form a stable hydrate on its own, but needs the help of another gas to enter the hydrate network and form a stable building. Hydrate is a solid but different from the structure of ice. Because it is a crystalline solid. Hydrate is a type of chemical compound called clathrate (solid molecular compound, one component of which is encapsulated in cage-shaped cavities). Which is the nomenclature used to identify compounds that may be present in fixed species [29-31].

#### *New methods of molding hydrated gases*

A two-stage mechanism for the production of hydrated gases has been proposed:

- (1) Perform a semi-chemical reaction to produce alkaline hydrates
- (2) Adsorption of small gas molecules from basic alkali hydrate chains.

This new concept is based on previous studies and two types of equilibrium: the semi-chemical equilibrium reaction in the first stage and the equilibrium physical absorption in the second stage.

The results obtained during successive experiments show that the new models have the ability to predict the formation of hydrates for pure gases and a combination of gases [32-38].

Many thermodynamic models are available to predict hydrate formation from the various modifications and corrections proposed by the Vdwp model proposed by Andrews and Plato. The basis and purpose of the vdwp model is

based on the relative similarity between hydrate formation and Langmuir uptake. Although the adsorption mechanism has the ability to interpret the properties and non-stoichiometric states of hydrates, there are many differences between the two processes. In this section, we will try to describe the real mechanism of hydrates to some extent that is similar to the hydrate model [39-42].

*Proposed mechanism of hydrate formation*

A two-step mechanism for hydrate formation is proposed as follows:

Step 1: Stoichiometric formation of alkaline hydrates during a semi-chemical reaction.

Step 2: Absorption of gas molecules among a number of alkaline hydrate chains during a non-stoichiometric process.

In the first step, based on our long and cylindrical design, we assume that the gas molecules are dissolved in water, which is done by surrounding the guest molecules with a few water molecules that surround it. This continuous encapsulation process, called alkaline hydration, involves the alkaline side of one of these molecules being captured and occupied by the gas molecules and the other side of the chain remaining intact [43-47].

We describe this process by the following complex process:



In this regard, G indicates the specific gravity of gas,  $\lambda_2$  refers to the number of gas molecules and the number of water molecules in alkaline hydrates. During this stage, the intact part (in the chain) is covered by other alkaline

hydrates. In the second stage, molecules that are small in size, such as (Ar, N<sub>2</sub>, O<sub>2</sub>, CH<sub>4</sub>, etc.) that are soluble in water, move towards chain rings (adsorption process). This action is the result of non-stoichiometric properties of hydrates. However, this will not happen at this stage for larger molecules, such as (ethane, propane, -n-butane, and -i-butane), as well as for those molecules that are able to separate from the chain. Therefore, the final stoichiometric form of alkaline hydrates will be formed only in the first stage. So, it can be said that alkaline hydrates are not hypothetical but these molecules are physically present. These were the logical reasons used in Langmuir's theory of adsorption to describe the saturation of chain links by gas molecules [48-52].

*Thermodynamic roots of the model and evaluation of pure hydrate gas parameters*

Based on the two-step hydrate formation mechanism described, there are two types of equilibria in the system. A semi-chemical equilibrium reaction that occurred in the first phase, and the physical equilibrium adsorption that saturated the gas molecules around the chain links during the second phase [53-58]. For the reaction described in formula (1), if we use chemical equilibrium constants we will have:

$$\mu_B = \mu_w + \lambda_2 \mu_g \quad (2)$$

Where  $\mu_B$  are the chemical potentials of alkaline hydrates,  $\mu_w$  and  $\mu_g$  are the standard chemical potentials of water and gas, respectively. After the adsorption of gas molecules in the chain links, the chemical

potential of alkaline hydrates will decrease.  $\mu_B$  can be displayed as follows.

$$\mu_B = \mu_B^o + \lambda_1 RT \ln(1-\theta) \quad (3)$$

Where  $\theta$  represents the fraction of chain rings enclosed by the gas molecules,  $\mu_B^o$  represents the unsaturated chemical potential of alkaline hydrates ( $\theta=0$ ) and  $\lambda_1$  indicates the number of chain rings of small chains per molecule. Water has alkaline hydrates. According to Langmuir's theory,  $\theta$  is calculated as follows [59-62].

$$\theta = \frac{Cf}{(1+Cf)} \quad (4)$$

Where  $f$  denotes the special gas fugacity and  $C$  denotes the Langmuir constant. Using the general and basic thermodynamic relations, the specific chemical potential of the gas is calculated as follows.

$$\mu_g = \mu_g^o(T) + RT \ln f \quad (5)$$

Where  $\mu_g^o(T)$  represents the chemical potential in the ideal gas state, the product of the subtraction of Equations (2)-(5) is given in the following relation:

$$\mu_B^o + \lambda_1 RT \ln(1-\theta) = \mu_w + \lambda_2 [\mu_g^o(T) + RT \ln f] \quad (6)$$

And in the definition, we have:

$$f^o = \exp\left[\frac{\mu_B^o - \mu_w - \lambda_2 \mu_g^o(T)}{\lambda_2 RT}\right] \quad (7)$$

Equation (6) can be converted to the following equation after sorting:

$$f = f^o (1-\theta)^\alpha \quad (8)$$

Where  $\alpha = \lambda_1/\lambda_2$ ,  $\alpha = \frac{1}{3}$  for compound I and

$\alpha=2$  for compound II, in the equation shown in Equation (7),  $f^o$  is not only a function of  $T$  and  $P$  and  $\alpha w$  of water activity, Refer to Equation

(11), but also represents a property of the alkali hydrate formed. When  $\theta=0$ , equation (8) will look like this [63-68].

$$f = f^o \quad (9)$$

In this case,  $f^o$  exactly indicates the gas phase fugacity at equilibrium, when the alkaline hydrate is unsaturated. ( $\theta=0$ ).

The fraction  $(\mu_B^o - \mu_w)$  in Equation (7) can be calculated using multiple thermodynamic relations.

$$\mu_w = A_w + PV_w + RT \ln a_w \quad (10)$$

$$\mu_B^o - \mu_w = \Delta A + P\Delta V - RT \ln a_w \quad (11)$$

$A$  represents the number of moles of Helmutz free energy (which is basically related to the energy of the system) and the number of moles of volume change  $\Delta V$  whose constant value can be obtained. Equation (7) can be the result of three factors. The share of each of the components  $T$ ,  $P$  and  $a_w$  are displayed as follows:

$$f^o = f^o(T) f^o(P) f^o(a_w) \quad (12)$$

In the equation (13)

$$f^o(P) = \exp\left(\frac{\beta P}{T}\right) \quad (13)$$

Is that  $\beta = \frac{\Delta V}{\lambda_2 R}$  using it the constant value can

be obtained,  $\beta$  is equal to 0.4242 k/bar for compound I and 1.0224 k/bar for compound II, respectively.

$$f^o(a_w) = a_w^{-1/\lambda_2} \quad (14)$$

The number  $\lambda_2$  is equal to 3.23 for compound I and 1.17 for compound II, respectively.

$$f^o(T) = A' \exp\left(\frac{B'}{T - C'}\right) \quad (15)$$

In a special and pure composition which is composed of one substance or fat, the

constants of Antoine, A' .B' and C' can be obtained in a constant and suitable amount of hydrate gas formation, which will be the specific values of the same substance. These constant values for alternating compounds can be obtained by measuring a compound of two hydrates [69-71].

These constant values, which have a downward trend, can be seen for specific gases in the relevant table. If the hydrates are formed in the cooling state, the constants listed in this table will remain in force, but Equation (16) is corrected as follows:

$$f^\circ(T) = \exp\left(\frac{D(T - 273.15)}{T}\right) \times A' \exp\left(\frac{B'}{T - C'}\right) \quad (16)$$

The constant D in equation (17) is 22.5 for compound I and 49.5 for compound II, respectively.

To apply Equation (8) in the general case ( $\theta \neq 0$ ),  $\theta$  can be obtained by Equation (4). To investigate the Long Moir constants, one can determine the integration of two Lennard-Jones potash parameters, the values of which indicate that the long Muir constants are very close to the numbers of the sails [72-78].

For the application engineer, it is very useful and convenient to connect the constant length of Moir C with temperature. In this operation, Antoine's equation is used to determine the value:

$$C = X \exp\left(\frac{\gamma}{T - Z}\right) \quad (17)$$

The constants X, y, Z, which are used to determine the Langmuir constant of C, are derived from the functions of Peta Nasil Leonard-Jones are calculated.

The measured constant values for each of these numbers indicate that, in order to suggest a mechanism, the formation of gas adsorption hydrates is done only by chain links.

#### Formation of hydrates in gas mixtures

The behavior and formation of hydrates consisting of gas mixtures is described in the previous pages as the concept of alkaline hydrate components. The mixture of alkaline hydrates has the same behavior as the composition of solid solutions and is called the components of alkaline hydrates. The general nature of the hydrate mixture depends only on the saturation of the gas molecules around the chain links.

In alkaline hydrates whose only difference in volume is summarized and has a composition They are the same, the extra volume and the additional entropy of the mixture of alkaline hydrates should be close to zero.

Therefore, it is reasonable to pay attention to the composition of the mixture of alkaline hydrates as a real solution [79].

Excluding the number of molecules in the solution that interact with each other (assuming the solution is ideal), the following procedure can be considered for the hydrate mixture:

$$\sum_j O_j = \frac{\sum_j f_j G_j}{1 + \sum_j F_j G_j} \quad (18)$$

$$\sum_j x_i = 1.0 \quad (19)$$

Where  $f_i$  denotes the fugacity of component i, calculated by the Patel-Tega equation of state.  $\theta_j$  on a part of the chain links occupied by the

gas component  $j$ .  $x_i$  Indicates the molar fraction of alkaline hydrate  $i$ . Determining the type and species of the hydrate model based on the Vdw-p contract, which is considered to be under the effect of calculating the chemical potential difference between water and the hydrate network ( $\Delta\mu_w$ ), was not possible. If we consider the effective factors to determine the hydrate model between molecules around the ring as an alkaline group, Equation (16) to calculate  $f_i^\circ(T)$  is corrected as follows:

$$f_i^\circ(T) = \exp\left(\frac{-\sum_j A_{ij} \theta_j}{T}\right) \times \left[A' \exp\left(\frac{B'}{T-C'}\right)\right] \quad (20)$$

Where  $A_{ij}$  is the effective two-component parameter between the components in our solution between the two components  $i$  and  $j$  ( $A_{ij} = A_{ji}$  ...,  $A_{ii} = A_{jj} = 0$ )

#### *Calculation of Antoine constants to compensate for a hydrate mixture*

The structure and composition of the hydrate mixture can be different from that of the constituent in pure form.

As mentioned before, in this case the structure of the hydrate mixture with the pure gas component can be different. The Antoine constants calculated for the two-component composition are similar to the structure and the hydrate mixture composition. The following is an acceptable solution for forming a gas component in a natural gas mixture.

The structure and composition of the hydrate mixture can be different from the constituent structure in its pure form Antoine's constants

$A'$   $B'$   $C'$  in Equations (16) and (22) can be calculated for both I and II.

As mentioned before, in this case the structure of the hydrate mixture with the pure gas component can be different. The Antoine constants calculated for the two-component composition are similar to the structure and the hydrate mixture composition. The following is an acceptable solution for forming a gas component in a natural gas mixture.

(1) To obtain the data for the formation of the two-component C<sub>2</sub>H<sub>6</sub>-C<sub>3</sub>H<sub>8</sub> system, the Antoine constants for both the C<sub>2</sub>H<sub>6</sub> compound (when mixed is composed of hydrate II) and the mixed C<sub>3</sub>H<sub>8</sub> (when mixed) can be obtained.

(2) To obtain the data for the formation of a two-component system CH<sub>4</sub>-Ar, we can use Antoine constants for the composition of CH<sub>4</sub> (when the mixture is formed from the combination of hydrate II) and Ar (when the mixture is formed from the combination of hydrate I).

(3) For H<sub>2</sub>S and CO<sub>2</sub>, when each of them is mixed with propane, the hydrate composition number II will be formed and the data formed according to the calculated data of the combination II will be Antoine constants for H<sub>2</sub>S and CO<sub>2</sub>.

(4) Calculation method: The calculation method to determine how hydrates are formed in the gas mixture is summarized and a separate diagram is shown in Figure 2. For simple hydrate gases (consisting of pure gases) the hydrate temperature can be calculated based on the same method.

### *Results of two-component and multi-component hydrate gas calculations*

Comparison of calculated results and values and values of experimental formation under the influence of pressure are present in two-component and multi-component systems separately in gas-hydrate systems.

For Ar-N<sub>2</sub>, Ar-CH<sub>4</sub>, CH<sub>4</sub>-H<sub>2</sub>S, C<sub>2</sub>H<sub>4</sub>-C<sub>2</sub>H<sub>6</sub>, C<sub>2</sub>H<sub>6</sub>-C<sub>3</sub>H<sub>8</sub> two-component systems, all A<sub>ij</sub>s options will be zero.

For other systems, the A<sub>ij</sub> crossover parameter is calculated in the system [80].

### **Natural hydrate gases**

If we consider the effect of the mixed molecules on each other, we set the value of zero for each pair of A<sub>ij</sub> for which quantity was not available. The mean absolute error of the predictions indicates that the new hydrate models on applied engineering are promising and have a bright future [81-83].

### *Discussions of hydrate building change*

I have known for years that if methane is mixed with a small amount of propane, the hydrate composition will change from I to II. Studies based on new models show that although the percentage of methane in the gas mixture is very high (95%), its alkaline hydrate volume is very low.

This implies that when a hydrate mixture is formed, most of it is encapsulated by propane, and methane only acts as a median in this process. From Equation 19 it can be seen that  $f_i$  is very sensitive and important in

determining the amount  $\sum_j \theta_j$ . For propane

the value  $\theta$  is equal to zero and  $F_i = f^{\circ}_i$ . However, for the composition and mixture (methane + propane), methane occupies most of the chain links. This is due to the presence of propane due to the slight decrease in hydrate formation pressure.

For example, when the molar fraction of methane around the chain link is 0.8, the partial pressure that propane will need to form the hydrate II compound is approximately 0.04 when the partial pressure of pure propane is required.

Although the concentration of propane in this operation is low. Based on the analysis of the above, naming the change in hydrate composition from methane would be somewhat misleading.

Interaction of molecules with each other The interaction effect of molecules on the hydrate phenomenon is examined in the following three categories.

- (1) The effect of the presence of molecules in solution on alkaline hydrate
- (2) The effect of exchange between molecules in solution and alkali hydrate and saturation around chain links by them
- (3) Interaction on molecules in solution and their adsorption by chain links.

The effect of  $f^{\circ}(aw)$  on equation 13:

From Equation 15 we have  $f^{\circ}(aw) = aw^{-7.67}$  for the hydrate compound number (I) and  $f^{\circ}(aw) = aw^{-17}$  for the hydrate compound number II.



This concept means that the pressure of product formation is very sensitive to water activity.

A slight decrease in water activity may cause a significant increase in the pressure formed. For systems consisting of water and gases with low solubility.

$X_w \rightarrow 1.0$  ,  $\gamma_w \rightarrow 1.0$  ,  $\alpha_w \approx x_w$  Therefore, Equation 15 can be shown as follows.

$$(21) \quad f^\circ(a_w) \approx [1-x_g]^{-1/\lambda_2} \approx 1+x_g/\lambda_2$$

$x_g$  represents the total molar fraction of gaseous species dissolved in the aqueous phase. When  $f^\circ(a_w)$ ,  $x_g = 10^{-13}$  equals 1.00768 for compound I and 1.017 for compound II, respectively.

On the other hand, for systems that contain gases with relatively high solubility in water, such as (H<sub>2</sub>S, CO<sub>2</sub>) and a salt dipole inhibitor, the contribution of  $f^\circ(a_w)$  to form in this case will not be minor [84-86].

For other compounds, the contribution of  $f^\circ(a_w)$  can be easily handled by a suitable combination of equilibrium gases based on available state equations or available activity coefficients.

## Conclusion

(1) Based on the two-step mechanism proposed for hydrate formation and the concepts reported in the previous pages, new hydrate models will be available.

(2) The complex algorithm of hydrate formation and simplification of the relevant calculations, and the direct calculation of  $\Delta\mu_w$  and the complex evaluations based on the Long

Muayer constant C, in addition to the optional selection of suitable parameters for the evaluation of  $\Delta\mu_w$  and C will not be possible.

(3) Methane hydrate changes when physically mixed with a small amount of propane can be physically addressed.

(4) The results of various experiments show that the proposal of new hydrate models is suitable for application engineers, especially when the combination of natural gases is desired.

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