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Optimization of Reagents Consumption in the Transportation of Gases

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Abstract

This article investigates the gas hydrate formation during the natural gas production, gathering, and pipeline transportation under changing thermodynamic conditions and evaluates its impact on the gas treatment efficiency and the reliable operation of gas transportation systems. Based on studies conducted at gas-condensate fields and an analysis of existing research, the integrated gas treatment process prior to transportation is examined. Thermodynamic modeling principles were applied to evaluate hydrate formation conditions by considering gas composition, pressure, and temperature. A calculation methodology is proposed to determine the optimum methanol consumption rate for the hydrate inhibition under the different operating conditions. The obtained results made it possible to establish optimal methanol dosage values for various technological regimes and to assess their effect on hydrate prevention. The study demonstrates that optimization of inhibitor consumption significantly reduces the risk of hydrate formation, improves gas treatment quality, enhances the operational safety of technological equipment, and decreases reagent consumption and operating costs. The proposed methodology can be effectively applied to improve the reliability, efficiency, and economic performance of natural gas transportation systems.

Keywords: gas condensate, methanol, hydrate, consumption rate, thermodynamics.

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Qazların nəqli zamanı reagentlərin sərfinin optimallaşdırılması

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Xülasə

Məqalədə qazın hasilatı, toplanması və magistral qaz kəmərləri ilə nəqli zamanı termodinamik şəraitin dəyişməsi nəticəsində yaranan hidratəmələgəlmə problemləri, onların qazın nəqlə hazırlanması prosesinə və qaz nəqli sistemlərinin etibarlı istismarına təsiri araşdırılmışdır. Tədqiqat çərçivəsində qaz-kondensat yataqlarında aparılan müşahidələr və mövcud elmi yanaşmalar əsasında qazın kompleks hazırlanması prosesləri təhlil edilmiş, hidratəmələgəlmə şəraitinin qiymətləndirilməsi üçün termodinamik modelləşdirmə prinsiplərindən istifadə olunmuşdur. Qazın tərkibi, təzyiq və temperatur parametrləri nəzərə alınmaqla hidrat inhibitoru kimi istifadə olunan metanolun optimal sərf normasının müəyyən edilməsi üçün hesablama metodikası təklif edilmişdir. Aparılmış hesablamalar nəticəsində müxtəlif texnoloji şəraitlər üçün optimal metanol sərfiyyatı müəyyən edilmiş, reagent sərfinin normallaşdırılmasının hidrat əmələgəlməsi riskini azaltdığı, qazın hazırlanma keyfiyyətini yüksəltdiyi və texnoloji avadanlığın təhlükəsiz istismarını təmin etdiyi göstərilmişdir. Müəyyən edilmişdir ki, təklif olunan metodikadan istifadə reagent sərfinin optimallaşdırılmasına, istismar xərclərinin azaldılmasına və qaz nəqli sistemlərinin etibarlılığının artırılmasına imkan verir.

Açar sözlər: qaz kondensatı, metanol, hidrat, sərfiyyat dərəcəsi, termodinamika.

Оптимизация расхода реагентов при транспортировке газов

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Аннотация

В статье исследованы процессы образования газовых гидратов, возникающие при добыче, сборе и транспортировке природного газа вследствие изменения термодинамических условий, а также их влияние на эффективность подготовки газа и надежность эксплуатации газотранспортных систем. На основе исследований, проведенных на газоконденсатных месторождениях, и анализа современных научных подходов рассмотрены процессы комплексной подготовки газа к транспортировке. Для оценки условий гидратообразования использованы принципы термодинамического моделирования, учитывающие состав газа, давление и температуру. Предложена расчетная методика определения оптимального расхода метанола, применяемого в качестве ингибитора гидратообразования. По результатам расчетов установлены оптимальные нормы расхода реагента для различных технологических режимов эксплуатации. Показано, что нормирование расхода метанола позволяет снизить вероятность образования гидратных пробок, повысить качество подготовки газа, обеспечить безопасную работу технологического оборудования и уменьшить эксплуатационные затраты. Предлагаемый подход способствует повышению надежности и эффективности функционирования систем транспортировки природного газа.

Ключевые слова: газоконденсат, метанол, гидрат, норма расхода, термодинамика.

Introduction

One of the operating costs encountered in the process of extracting, collecting and transporting oil and natural gas in offshore fields is related to the consumption of chemical reagents used to prevent hydrate formation and other technological difficulties. If this consumption is large, it increases the cost of the produced gas. Therefore, regulating the quality and quantity indicators of the reagents used to ensure the unhindered transportation of gases is one of the urgent and important issues in oil and gas production.

Taking into account the above, scientific research and mining work was carried out to improve the working principle of the devices and technological system and to develop new scientific and technical proposals to regulate the consumption rate of reagents used in the process of extracting, drying and transporting gas from gas and condensate fields.

Research work was carried out in several directions. When studying the operation of individual devices of the gas preparation unit operating at the OGDI of the PU, it was found that methodological reports were not prepared taking into account actual indicators to regulate the consumption of reagents used in the system [1-3]. The results of research conducted in offshore fields showed that since the extraction, accumulation and preparation of gas condensate for transportation are carried out under complex conditions, it is advisable to improve the composition and technologies of reagents used to prevent hydrate formation in the technological system.

Problem Statement

A general approach to the thermodynamic modeling of phase equilibrium in water–gas hydrate systems is proposed. Special attention is given to the investigation of vapor–liquid equilibrium in the natural gas–hydrocarbon condensate system, as it provides the basis for predicting hydrate formation and improving the efficiency of gas preparation and transportation processes. However, the application of a methodological approach to modeling the equilibrium of the condensed water phase - water, cooled water, aqueous solution of inhibitors, transformation of the ice phase into hydrates, and the thermodynamic state of each phase in contact were studied separately. This method has a number of advantages for gas preparation under thermodynamic conditions in the well, which ensure the accuracy of reporting phase equilibrium parameters and normal ization of hydrate formation inhibitors. In order to accurately report the phase imbalance in mining conditions, it is necessary to accurately report the volatile or non-volatile components of the gas phase, including CO_2, H_2S, N_2, H_2O and methanol. For this purpose, a modified thermodynamic model based on the Redlich-Kwong equation for a hydrocarbon or non-hydrocarbon gas phase has been developed. The model used according to the modification includes additional parameters characterized by second-order virial coefficients and the compressibility coefficient of gas mixtures at high pressure. The parameterization of the modified equation shows that the calculation of the volatile components of natural gas results in an error of 1-2%, which is more accurately calculated by the Peng-Robinson equation.

Literature Review

The advantage of the modification of the equation of state is the ease of adding additional changes due to second-order coefficients when conducting new experiments, and the disadvantage is the high number of parameters. This

is characterized by an increase in the accuracy of the description of the Hibbs energy in the gas phase. The effective range of use of the equation is -40°C ÷ +100-150°C, and the pressure is 0,1MPa÷12-14 MPa.

Recently, it was considered necessary to update the parameterization of the hydrate phase model based on the specification of more accurate data, such as the difference in chemical potentials - $\Delta\mu^0$ and the enthalpy of the empty hydrate network of water-ice hydrate - Δh^0, and therefore the Langmuir coefficient of individual components of natural gas mixtures has been recalibrated. Currently, the new values are as follows:

For structure I:

$\Delta\mu^0$=1203 J/mol;

Δh^0=1170 J/mol;

T=273,15° K at temperature;

For structure II:

$\Delta\mu^0$=1077 J/mol;

Δh^0=1294 J/mol;

T=273,15° K at temperature.

The ice-water enthalpy Δh^f is equal to 6008 J/mol.

Using the new data, the Langmuir coefficient was reconstructed, i.e., the parameters of the gas-hydrate phase were updated, which improved the description of the three-phase equilibrium "gas-water-hydrate" for the complex gas phase, but did not affect the description of the three-phase equilibrium with pure gases. Based on the above models, a complex reporting program was developed for the equilibrium of natural gas with water, individual hydrate compounds in gas in the gas industry or gas mixtures at a given pressure and temperature.

With the help of the complex reporting program, simple and relevant analytical dependencies were obtained for the purpose of determining the norm of natural gas mixtures from various fields, hydrate formation, and the consumption of hydrate inhibitors in the gas collection and preparation system. To confirm the results of the calculation of the gas-hydrate equilibrium of gases with complex composition, it is advisable to use the thermodynamic method of comparative reporting, including the method of empirical correction of the thermodynamic model of the gas-hydrate phase using the hydrate formation curve of a complex gas. The thermodynamic extrapolation method of three-phase equilibrium test data at negative or positive temperatures in a wide temperature range, with a pressure of 100 MPa and higher, is used.

Analysis of the thermodynamic model shows that the test data obtained, provided that the degree of filling of the gas-hydrate space is not higher than 1, is calculated as follows [4-6].

$$\ln f - H(p,T) - \frac{\Delta V\ p}{(\nu_1+\nu_2)RT} \quad \div \quad \frac{1}{T}$$

f – volatility of the component; ΔV – difference in molar volumes of water and hydrate, cm^3;

p – pressure, MPa; $H\ (p,\ T)$ – Henry's constant;

ν_1, ν_2 – crystal-chemical constants (characterizing the type of hydrate structure).

Approximation dependence of data in the coordinate system:

$$Y = A + \frac{B}{T} + C \cdot \ln(T)$$

where: A, B, C are coefficients that describe the data at the test points in more detail, allowing for a broad extrapolation of the dependence obtained at negative or positive temperatures. The equilibrium pressure at a given temperature is determined by the following formula:

$$A + \frac{B}{T} + C \cdot \ln(T) = \ln(f) - \frac{p \cdot \Delta V}{(\nu_1+\nu_2)\cdot RT} + \frac{\ln x_{H_2O}}{\nu_1+\nu_2}$$

Determination of gas volatility, water density and water solubility in gas at high pressure is mandatory. The extrapolation method was carried out for two gases (methane, nitrogen) and the results obtained are sufficient for the temperature equilibrium range of hydrate formation for methane. For methane it is up to ~307° K (110 MPa) and for nitrogen it is up to ~301° K (230 MPa), the error percentage in the report is ~ 5-6% (depending on pressure).

The thermodynamic properties of the metastable phase are shown in relation to hexagonal ice, cooled water and ice cube. Cooled water and ice cube are formed during the separation of gas hydrate as an intermediate phase. The state temperature of hexagonal ice is standardly T_o=273,15° K.

The thermodynamic report of the difference in chemical potentials between hexagonal ice and cooled water was carried out based on the thermodynamic properties of the condensed water phase based on the experimental data provided in the literature. The phase equilibrium used to calculate the difference in chemical potentials between cooled water and ice cubes was calculated with an error of a few percent in the temperature range of 240–273° K. A model of slow hydrate separation is presented and the main experimental data are summarized. The theoretical model is typified and the mechanism of hydrate separation and the surface separation property of gas hydrate are explained in detail.

In order to expand the possibilities of gas hydrate conservation technology, Istomin V.A. proposed that in order to slow down the separation of gas hydrate, its upper surface should be covered with another solid-phase thermodynamically stable coating, unlike the hexagonal ice layer [7-10]. The realization of the kinetic mechanism of separation depends on the gas content in the hydrate, the morphology and structure of the hydrate, and the thermobaric conditions of its formation and separation.

The driving force of the separation process is determined by several methods: for example, ΔT is the temperature of the separation process and the temperature difference between the separation of the hydrate into the gas and water phases and is calculated as follows:

$$\Delta G = RT \ln f / f_0 \approx RT \ln p / p_0$$

where f, p - gas volatility and pressure in the system, f_0, p_0 - equilibrium value of volatility and pressure.

During the test, the driving force may change at the initial stage, the results of the test data show the properties of the mechanism of the gas hydrate separation process and its kinetics at negative temperatures:

- staged (conservation effect resulting in an initial fast stage and a subsequent slow separation stage);
- a clear demonstration of the self-conservation effect of hydrate methane and natural gas in a certain temperature range;
- the formation of various metastable water phases at the initial separation stage.
- the formation of a metastable surface phase at different times depending on the thermobaric conditions.

Problem Solution

Depending on the test conditions, the mechanism of gas hydrate separation at negative temperatures can be divided into options.

- For a precise separation of the conservation effect, the initial stage is the separation of hydrate into gas and cooled water, followed by crystallization of the cooled water layer under optimal conditions.
- Long-term storage of cooled water in an uncrystallized state, and in exceptional cases,

the complete separation of the hydrate particle into cooled water and gas. In this case, it is impossible to stop the process. During the conservation effect, one part of the hydrate particles is completely separated, and the other part is self-conserved.

If the first surface separation of the hydrate occurs with ice and gas, an ice layer forms on the top layer of the hydrate and, although the process is slow, a sharp course does not occur.

The process of surface separation of gas hydrates is divided into three main stages at negative temperatures: the first stage is the stage of separation into the metastable phase, the stage of crystallization or recrystallization of the metastable water phase, and then the stage of slow separation by the diffusion mechanism. Sometimes the stages may not occur or the separation stage may end at the first stage. In order to more fully study the initial stage of the process of separation of the methane hydrate part, a three-phase calculation of the water phase and the methane hydrate part at low (272.95°K) temperature was carried out.

When transferring the system from area 1 to area 3 (Fig. 1.) and in the presence of ice in the hydrate, as a result of the separation of methane hydrate in the first stage, it passes into the cooled water phase, and water directly crystallizes and turns into ice. Water creates a dynamic layer between the hydrate and ice, then the separation rate decreases, the water layer softens and disappears, and thus the hydrate conservation stage occurs.

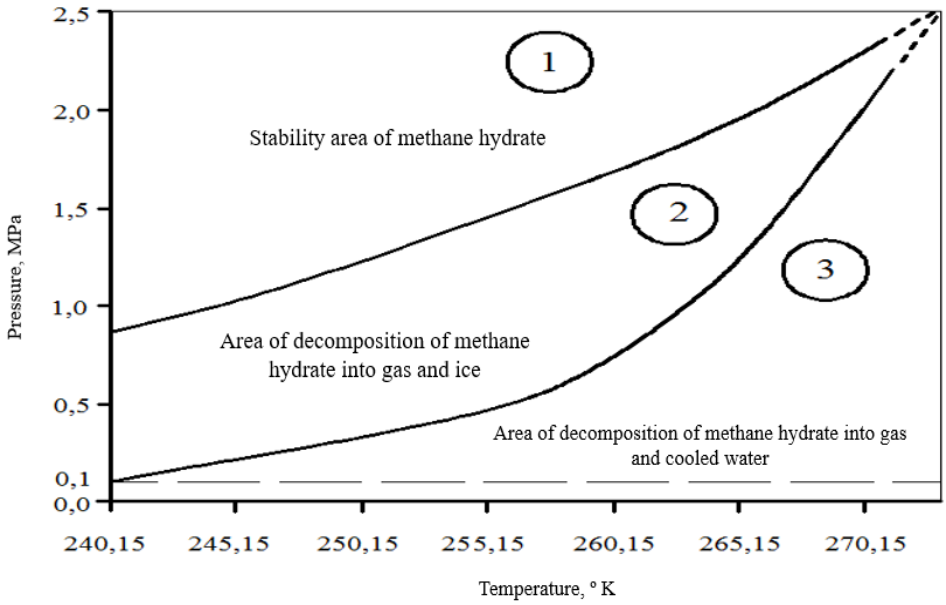

Figure 1 – Separation of methane hydrate into “gas-ice” and “gas-cooled water” phases in different thermobaric zones as a result of increasing pressure.

It should be noted that the formation of cooled water is visually visible (the "darkening" of hydrate particles is observed in the light passing through the window of the hydrate pressure chamber). This visual representation of the decomposition process allows us to study the experimental equilibrium line "gas (methane, propane) - hydrate - cooled water".

In order to determine the possible temperature range of the manifestation of the self-preservation effect, similar studies have been conducted for hydrates of other gases. Reports

have also been made for propane, ethane, and isopropane by taking a different temperature range, and as can be seen from Table 1, the lower limit of the cooled water for the obtained nitrogen hydrate is ~240 K below the theoretical point, in which case the cooled water practically does not exist.

Therefore, the determination of the temperature range when the pressure is reduced to atmospheric pressure is an important element in the optimization and search for a method of preserving hydrates of various gases.

Thermodynamic analysis of the surface separation of hydrates shows that it is possible to switch to the preservation method and control the process. Here, the main parameter is pressure.

A decrease in pressure to a certain value leads to the formation of cooled water in the upper layer of the hydrate.

One of the leading parameters in the conservation effect is temperature, the increase in which in studies leads to the surface separation of hydrate in the network of three-phase equilibrium lines "gas-hexagonal ice-hydrate" and "gas-cooled water-hydrate" in the corresponding sequence.

In the "gas-cooled water-hydrate" network, hydrate is converted into cooled water. Under optimal conditions, the crystallization of this water is important.

When the process power is low, the crystallization of water is accelerated, for example, it is possible to accelerate the re-cooling of the system or the combination of cooling by increasing the pressure. The presence of ice in the hydrate allows it to be controlled by the separation process and transferred to the conservation stage, since ice leads to the crystallization of cooled water.

Table 1 – The temperature range of the presence of cooled water during the separation of various gas hydrates when the pressure drops to atmospheric pressure.

Gas	Temperature, °K
Methane	~240 – 272,95*
Nitrogen	~223 – 271,85*
Ethan	~258 – 273,15*
CO_2	~251 – 273,0*
C_3H_8	~270,5 – 273,15*
Iso-butane	not available

It is possible to choose technological routes for each gas hydrate. The hydrate sample is pre-heated in a hydrate thermostatic pressure chamber. The change in the temperature of the chamber depends on the stabilization of the pressure, which means that the hydrate separation process is complete. Then the process continues at a higher temperature. At each temperature step, the separation process occurs, which results in an increase or a constant pressure of the hydrate sample. When the pressure increases, a re-hydration of the gas phase and water occurs on the upper layer of the first hydrate sample. (The composition of the new hydrate film is different). Thus, the initial hydrate separation process continues stably and the pressure in the system stabilizes. These experiments show that a hydrate of one composition is converted into a hydrate of another composition by conservation.

The process results in the creation of the effect of conservation of hydrate by hydrate.

Depending on the conditions, a sub-hydrate is separated, a hydrate is formed from the gas dissolved in water, which partially prevents the separation of the initial hydrate as a new stable hydrate. Another method for controlling the effect of slowing down the separation of hydrate has been discovered, which is the method of covering one hydrate with another hydrate

under thermobaric and stable thermodynamic conditions, at positive temperatures. It is shown from the reports that it is possible to conserve a hydrate of one composition with a hydrate of another composition, and its pressure is lower than the pressure of the other hydrate compound.

Thus, the composition of the gas mixture: 98% methane + 2% propane; 95% methane + 5% nitrogen; 70% isobutane + 30% nitrogen and 90% methane + 10% CO_2).

The conducted analysis and thermodynamic reports show that the following conditions form the possibilities of the effect of slow separation of gas hydrates:

- development of new methods of studying gas hydrate equilibrium;
- technology of storage and transportation of gas in the hydrate state;
- conditions of existence of gas hydrates in metastable conditions.
- a methodological approach to the standardization of hydrate formation inhibitors and their additional features with the use of volatile inhibitors are noted.

All data on the standardization of inhibitor consumption include information on its technological losses and other production needs.

Production losses of the inhibitor arising during its regeneration, storage, transportation is determined by the experimental-statistical method, other losses are determined by the reporting-analytical method.

In order to return the spent reagent to the technological cycle, it is necessary to install a regeneration unit for the spent inhibitor, and develop a recirculation technology for the use of hydrate formation inhibitors.

The methodological approach to the standardization of the inhibitor is as follows.

The consumption of chemical reagents in the production of all gas production is determined in group form (H) as follows:

$$H = \frac{\sum H_i \cdot Q_i}{\sum Q_i},$$

Here: H_i - individual consumption of chemical reagents per unit, kg/1000 m^3;

Q_i - gas production per unit at a certain time (month, quarter, year), mln. m^3.

Chemical reagent consumption H_i indicates the specific consumption rate per unit:

$$H_i = H_T - H_B + \sum g_i$$

Here: H_T – the rate of inhibitor consumption in the technological process;

H_B – the rate of specific consumption, the chemical reagent returned to the cycle;

$\sum g_i$ – the rate of final production losses.

The rate of inhibitor consumption in the technological process for each unit in the well is determined by the sum of the inhibitor consumption in each individual technological process in accordance with the well plume.

The rate of inhibitor consumption in each individual area is determined by the following formula:

$$H_{T,i} = G \cdot K$$

Here: G – minimum-necessary (theoretical) specific consumption of the inhibitor;

K – reserve coefficient in the range of 1.05-1.25 for factors not taken into account;

K – the distribution coefficient of the components by phase during non-isothermal and non-stationary flow of the gas-liquid mixture is determined experimentally.

During the initial stage of operation of the field, the maximum value of the coefficient is equal to 1.10÷1.25, the inhibitor consumption decreases to 1.05 during the implementation of

automation of production processes. The theoretical consumption of the inhibitor and the consumption of the water phase at the protected point for an individual section of the technological chain are determined by the following formula [11-13].

$$G_2 = \frac{X}{X_2} + \frac{G_1 \cdot X_1 + G_1^{recycle} \cdot X_1^{recycle} + 100 \cdot [(Q_1 - Q_2) + (q_1 - q_2)]}{X_2}$$

$$G = \frac{G_1 \cdot (X_2 - X_1) + G_1^{recycle} \cdot \left(X_2 - X_1^{recycle}\right) + (W_1 - W_2) \cdot X_2}{X - X_2} + \frac{100 - X_2}{X - X_2} \cdot [(Q_2 - Q_1) + (q_2 - q_1)]$$

G – specific consumption of the minimum-essential inhibitor, concentration X, kg/1000 m^3; G_1 - specific amount of the inhibitor entering the "liquid-water" phase from the previous technological area to point 1, kg/1000 m^3;

$G_1^{recycle}$ – specific amount of the recircuitlating aqueous solution of the inhibitor supplied to point 1, kg/1000 m^3;

G_2 – specific amount of the aqueous liquid phase of the inhibitor supplied to point 2, kg/1000 m3; X_1, X_2 - concentration of the aqueous phase of the inhibitor in front of point 1 and at point 2, respectively, % mass; *X* – concentration of the inhibitor supplied to point 1, % mass; $X_1^{recycle}$- concentration of the spent inhibitor supplied to point 1, % mass;

W_1, W_2 – moisture capacity of the gas at points 1 and 2 (equilibrated with the aqueous liquid phase), kg/1000 m^3; Q_1, Q_2 - the amount of inhibitor in the gas phase at points 1 and 2, kg/1000m^3; q_1, q_2 - the amount of inhibitor in the hydrocarbon condensate at points 1 and 2, kg/1000m^3;

G_1, $G_1^{recycle}$, X_1, X_2, $X_1^{recycle}$, *X* are assumed to be known quantities, and the quantities G and G_2 are found by calculation.

In practice, the theoretical consumption of the inhibitor is calculated several times, that is, calculations are made at several technological points, and the obtained values are summed up.

Here, the methodology is aimed at simplifying the thermodynamic equation of the gas-hydrate phase. The parameters are selected by the formula based on experimental data. It is possible to use the above-mentioned reporting methods to report the conditions of hydrate formation.

Calculation of the concentration of hydrate formation inhibitors at a given pressure and protection point ΔT (K) in gas and gas-condensate reservoirs and the dependence of the temperature difference of hydrate formation on the movement are determined as follows:

$$\Delta T = -A \cdot \ln \frac{(100 - X)/18}{(X / M + (100 - X)/18)}$$

A – empirical coefficient depending on the gas pressure, its composition, and the structure of the formed hydrates. This coefficient is selected individually, depending on the composition and pressure of the gas phase.

When analyzing the methodological approach to the practical application of the normalization of volatile and non-volatile inhibitors for hydrate formation inhibitors discussed

above, a number of advantages were revealed. The advantages of the volatile inhibitor - methanol - are associated with the non-linear nature of the distribution of hydrates in phases depending on temperature and pressure. In gas preparation devices used in low-temperature processes, the injection of methanol to protect against hydrates ensures the hydrate-free operation of the device. To calculate the norm of consumption of volatile hydrate formation inhibitors, it is necessary to calculate the dependence of the inhibitor consumption on the operation of the heat exchanger device under thermobaric conditions, and to calculate the inhibitor consumption, a maximum function should be selected.

Conclusion

A thermodynamic model and software have been developed, allowing for reporting of condensed water phase of hydrocarbon systems, including metastable phase, three- and two-phase equilibrium.

A correction method of the reporting model of test data, a thermodynamic extrapolation method for improving the description of the three-phase equilibrium "gas-condensed water-hydrate" are proposed.

A thermodynamic analysis of the surface separation of gas hydrates of various gases at negative temperatures has been developed, methods for controlling the separation process of gas hydrates and their conservation have been proposed.

A methodology for normalizing the consumption of hydrate formation inhibitors has been developed on the basis of experimental analysis and a general basis, and an analysis of the main features of its use for gas preparation technology has been given.

Conflict of Interest

The authors declare that there is no conflict of interest related to the publication of this article.

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