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## THEORETICAL AND EXPERIMENTAL JUSTIFICATION OF THE METHOD FOR DETERMINING THE PARAMETERS OF THE MOMENT OF GAS HYDRATES MASS CRYSTALLIZATION

The processes of oil and gas production – extraction, preparation, storage and transportation of oil, gas and condensate – are accompanied by risks of man-made hydrate formation. Such man-made gas hydrates cause serious problems for the oil and gas production industry. Oil and gas companies bear significant material costs in connection with the prevention of these processes. For prevent or eliminate it in each specific case, it is necessary to understand the physics of processes and parameters of hydrate formation. Therefore, establishing the peculiarities of the kinetics and thermobaric parameters of the hydrate formation process is an urgent problem. Thus, the object for research is the parameters of the beginning of mass gas hydrates crystallization in reservoir systems. At the same time, the most reliable results can be obtained in the process of laboratory monitoring of processes in reservoir systems and technological equipment directly at industrial facilities.

The process of hydrate formation at the phase boundary is manifested by the formation of a thin hydrate layer in the form of a film. In the course of experimental studies, it was established that this process is visually fixed by the transformation of the mirror surface of the phase boundary into a matte one. The distortion effect of the interphase boundary is explained by the formation, growth, massive and chaotic accumulation of gas hydrate microcrystals at this boundary. In the work, based on the results of theoretical and experimental studies, the methodology for operational laboratory determination of parameters of mass gas hydrate crystallization is substantiated. The essence of the technique is to establish the parameters for the moment of mass gas hydrates crystallization based on the fixation of the optical distortion effect of the reflection of the light source on the mirror of the liquid-gas interphase surface. The results of empirical studies are based on optical phenomena observed at the interfacial surface of the gas hydrate layer and gas. They were studied using microscopy, fixation and image processing methods. The main experiments result was the information recorded by the optical system and obtained after fixing the pressure and temperature.

The technique can be used to establish and operationally control the moment of mass gas hydrates crystallization directly at the objects of the oil and gas industry (during the implementation of technological processes). This will make it possible to effectively prevent clogging of technological equipment with the solid gas hydrate phase, as well as to prevent overuse of hydrate formation inhibitors. At the same time, the only limitation of the application for this technique may be the low light permeability of the aqueous solution as part of the formation system.

**Keywords:** gas hydrates, phase transitions, mass crystallization, interphase contact, induction period of hydrate formation, gas bubble.

Received date: 08.01.2023

Accepted date: 16.02.2023

Published date: 22.02.2023

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### How to cite

Pedchenko, L., Pedchenko, M., Yelchenko-Lobovska, A. (2023). Theoretical and experimental justification of the method for determining the parameters of the moment of gas hydrates mass crystallization. *Technology Audit and Production Reserves*, 1 (1 (69)), 26–31. doi: <https://doi.org/10.15587/2706-5448.2023.274183>

### 1. Introduction

The processes of extraction, preparation, storage and transportation of oil, gas and condensate are accompanied by the problem of the formation of man-made gas hydrate plugs. Gas hydrates are formed from gases and water under certain thermobaric conditions [1]. The composition of the system and the gas mixture in it affects the parameters of the thermodynamic hydrate equilibrium.

The analysis of the purpose of technological operations and equipment for extraction and preparation of natural

and oil gas shows that a significant part of them is related to the prevention of hydrate formation. To prevent hydrate formation or remove formed hydrates in each specific case, an understanding of the physics for hydrate formation processes and parameters is necessary. Possible zones formation of hydrate plugs are determined by gas composition, thermobaric conditions, and its moisture content [2].

Process prevention methods involve the elimination of at least one of the necessary factors: temperature, pressure, or gas moisture. At the same time, in many cases of operation of objects, it is impossible to achieve a non-hydrated

regime by maintaining appropriate thermobaric conditions. Then substances-inhibitors for this process are used [1].

Therefore, management of hydrate formation processes has become necessary for safe and reliable oil and gas extraction [3]. Development of methods for detecting early signs of hydrate formation may be one such option. This will allow operators sufficient time to take appropriate measures to prevent hydrate formation.

There are various analytical methods that are used to identify the structures of gas hydrates that occur at the interface of the water and vapor phases [4]. For example, a method based on a change in the composition of the gas phase due to the formation of hydrates [5]. In the process of hydrates formation, the components of natural gas are selectively captured in hydrate cells and the composition of the gas phase changes [6]. Next, the speed of sound, thermal conductivity are determined, and the equivalent concentration of the components for the gas mixture is established [5].

The concentration of salts and thermodynamic and kinetic inhibitors of hydrates is determined on the basis of the measurement of electrical conductivity and acoustic speed data of the chemical composition for the aqueous phase, and the stability hydrates zone is determined [7]. In [8], a method for determining the reserve of stability for the hydrate zone based on the measurement of the water content in the gas phase is proposed.

In [9], the authors propose monitoring systems to prevent hydrate formation based on dielectric properties and determination of the freezing point of the aqueous phase.

However, a significant complex of gas hydrates properties, a diverse composition of reservoir systems, a wide range of thermobaric parameters and their fluctuations do not allow obtaining an acceptable unambiguous result for most of the known methods. At the same time, an important task remains the improvement of the method of operational hydrate formation forecasting in systems of collection and preparation of industrial products.

Therefore, *the aim of research* is theoretical and experimental substantiation of the method for finding parameters at the moment of mass gas hydrates crystallization. This will allow controlling the process of hydrate formation in technological lines.

## 2. Materials and Methods

*The object of research* is the parameters of the beginning of mass gas hydrates crystallization in reservoir systems.

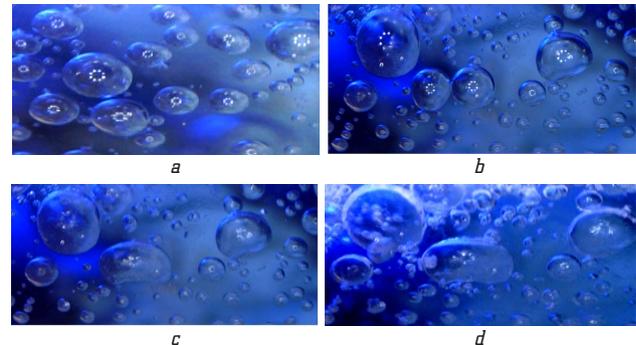
*The formation of defects on the mirror images of the liquid-gas interphase contact*

It is known that the formation of hydrates is characterized by an induction period, which includes the appearance of crystal nuclei and their growth [10]. Next, agglomeration of crystals takes place – the phase transition stage, when hydrate plugs are formed [8, 11].

As is known, the most intense process of hydrate formation occurs at the boundary of contacting phases (gas and water). The process at the phase boundary (liquid mirrors, drops and bubbles in a laboratory reactor or technological equipment) is manifested by the formation of a thin hydrate layer in the form of a film. Visually, this process is fixed by the transformation of the initially mirror surface of the phase boundary into a matte one, with defects in the form of reflections for light sources. The dynamics for this process is illustrated in Fig. 1, 2.



**Fig. 1.** Mirror interfacial surface of bubbles for hydrate-forming gas in water (gas hydrate film is absent)



**Fig. 2.** Dynamics of distortion of reflections for point light sources on images of gas bubbles in water as a result of the formation of a gas hydrate film on the interfacial surface: *a* – reflection of point light sources on the surface of gas bubbles; *b* – the beginning of the process of distortion reflection for the light sources; *c* – turbidity of the interfacial surface; *d* – formation of a gas hydrate film

This effect of successive distortion of the interphase surface mirror can be explained by the formation and chaotic accumulation of gas hydrates at this boundary. At the same time, a significant deviation of thermobaric parameters at the initial stage 1 (matte surface of the bubble) is practically not recorded by control systems (Fig. 3).

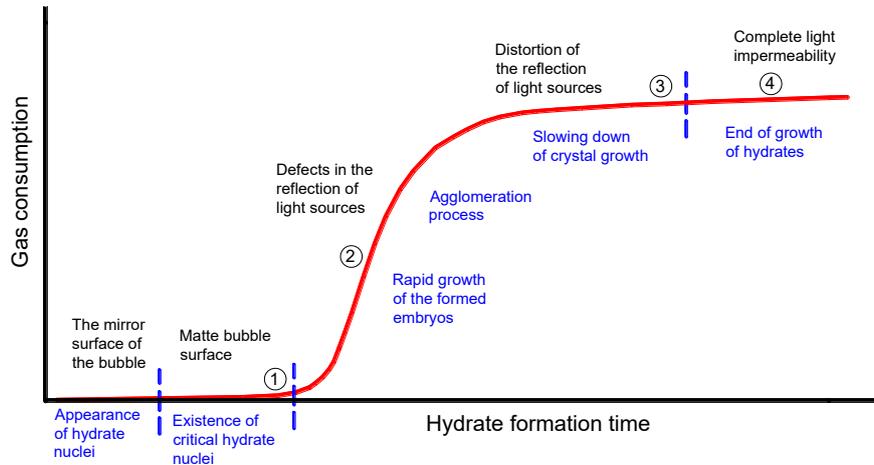
At the initial stage 1 (Fig. 3), as a result of the increase in Gibbs energy  $\Delta G$ , nucleation of hydrate nuclei (nuclei) occurs. At the same time, from the point of view of the dispersed systems classification, the solution of gas molecules and nuclei in water can be considered ideal. One with the important properties characteristic of non-colored systems of a dispersion molecular degree is the complete passage of light, transparency, and the absence of a scattering process. Therefore, a gas bubble surface in a liquid visually looks like a mirror when it is magnified and illuminated.

Further growth and overcoming the energy barrier  $\Delta G_{cr}$  leads to the nucleus reaching the critical size  $r_{cr}$ , the value of which can be calculated using the formula:

$$r_{cr} = -\frac{2\sigma'_{ef}T_0}{\Delta H\Delta T}, \quad (1)$$

where  $\Delta H$  – heat of hydrate former crystallization, kJ/mol;  $T_0$  – equilibrium temperature of hydrate formation for the corresponding pressure, K;  $\Delta T = T - T_0$  – relative cooling of the system;  $\sigma'_{ef}$  – effective specific surface energy of the grain, mJ/m<sup>2</sup>.

The value of the critical radius  $r_{cr}$  for the system «gas (in a bubble) – water» depends inversely on the supercooling temperature  $\Delta T$  and will decrease with its growth (decrease in system temperature). As a result, the induction period of hydrate formation is reduced.



**Fig. 3.** Kinetics of the gas hydrates formation on the bubble surface: 1 – nucleation of hydrate nuclei; 2 – rapid growth of hydrate crystals and their agglomeration; 3 – slowing down the growth of hydrate particles; 4 – completion of hydrate formation

At the end of the induction period, water with formed critical hydrates nuclei can conditionally be considered a colloidal system (the size of «true colloidal» particles is within 1–100 nm) [12]. At the same time, the dispersed phase of the system is nuclei of a critical size. For example, for natural gas hydrate (density about 900 kg/m<sup>3</sup>) at  $T_0=278$  K and  $\Delta T=3$  K, the radius of the critical nucleus will be  $r_{cr} \approx 15$  nm, and at  $T_0=280$  K and  $\Delta T=1$  K – about 45 nm.

Since the resolution of an optical microscope does not exceed  $10^{-7}$  m, colloidal particles, which are the nuclei of gas hydrate, cannot be fixed with its help. However, when the light wavelength is longer than the linear dimensions of the dispersed phase particles, scattering (opalescence) is a characteristic optical property for colloidal systems [13]. (The sizes of the critical  $r_{cr}$  nuclei at the end of the induction period are much smaller than the wavelength of visible light ( $400 < \lambda < 750$  nm)). Therefore, the presence of gas hydrate nuclei can be recorded when the sample is illuminated from the side by the fact of its scattering (the surface of the bubble becomes slightly matte (Fig. 2)).

For describe a colloidal system with a particle size of the dispersed phase up to 0.1 wavelength of light (40–50 nm), the Rayleigh equation is used:

$$I_{scat} = \frac{24\pi^3}{\lambda^4} \left( \frac{n_1^2 - n_0^2}{n_1^2 + n_0^2} \right)^2 \gamma V^2 I_0, \quad (2)$$

where  $\gamma$  – concentration of dispersed particles;  $V$  – the particle volume;  $\lambda$  – wavelength of light;  $n_1, n_0$  – refractive indices of the dispersed phase and dispersed medium;  $I_0, I_{scat}$  – intensity of incident and scattered light.

Therefore, the greater the difference between  $n_1$  and  $n_0$ , the greater the turbidity and the stronger the light scattering.

The next stage 2 of the process is the rapid growth of hydrate crystals and their agglomeration (stage 2, Fig. 3). It depends on the heat removal method of the hydrate formation process. For a reactor with a stirrer, the growth rate of crystals is determined by the empirical relationship:

$$v = \alpha \Delta T^\beta, \quad (3)$$

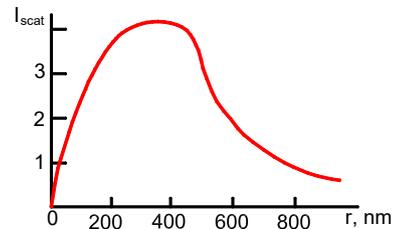
where  $v$  – rate of gas absorption during hydrate formation;  $\alpha$  and  $\beta$  – constants, the values of which depend on the energy for mixing the components of the system and are determined experimentally [14].

The sizes of hydrate particles have a significant effect on the intensity of scattering. The volume of the particle is determined by the formula:

$$V = (4/3)\pi r^3,$$

where  $r$  – the radius of the hydrate particle.

Then the scattering intensity  $I_{scat}$  according to formula (2) is proportional to the volume of the formed hydrate particles  $V$  and their radius  $r^6$ . Therefore, even with a slight increase in the size of gas hydrate particles, the scattering intensity or turbidity of the colloidal solution increases sharply (Fig. 4).



**Fig. 4.** Dependence of the scattering intensity on the linear dimensions of the particles [13]

The growth of hydrate crystals causes an increase in the effect of light scattering – clouding of the surface of the bubble. After the crystals reach a size that exceeds the length of light waves, their reflection occurs. However, the unevenness of their growth leads to the appearance of light reflection defects on the bubbles.

The next stage of the process is the slowing down of the growth for hydrate particles (stage 3, Fig. 3). Volume-diffusion growth of crystals will be observed on the formed gas-hydrate crust. The speed of the process is determined by the water diffusion rate  $M_w$  through a flat hydrate film of thickness  $h$  and surface  $F$  [15]:

$$M_w = D_w \cdot \frac{F \cdot \Delta f \cdot \rho_w}{h}, \quad h = \sqrt{\frac{2D_w \cdot \Delta f \cdot \tau}{n}}, \quad (4)$$

where  $D_w$  – the diffusion coefficient of water through the hydrate film (for natural gas of relative density 0.6  $D_w = 10^{-6}$  sm<sup>2</sup>/s);  $\Delta f$  – difference in volatility of water vapor over liquid and hydrate;  $\rho_w$  – density of water in the hydrated

state ( $0.757-0.792 \text{ g/sm}^3$ );  $h$  – hydrate film thickness during hydrate formation;  $\tau$  – time;  $n$  – mass ratio of water and gas in the hydrate.

Since at this stage the linear dimensions of the hydrate particles exceed the light wavelength  $\lambda$ , their reflection will occur.

The last stage of the process (stage 4, Fig. 3) is the completion of hydrate formation. There is a thick, dense and opaque crust that covers a gas bubble that is not part of the gas hydrate.

Therefore, based on the above analysis of the kinetics for hydrate formation, in order to prevent complications in the technological lines of oil and gas production facilities, the most valuable parameters are the moment for the beginning of its mass crystallization.

For the practical implementation of the method of laboratory finding of the parameters for the beginning of mass gas hydrate crystallization, a laboratory installation construction in which a capillary for low-dose gas supply is placed in the work [16] is proposed. At the same time, for the acceptable efficiency of bubbling mixing of the sample and the possibility of visual fixation of the process, a gas supply rate of 0.5 to 3.0 bubbles per minute is provided.

### 3. Results and Discussion

Consider the bubbling process of bubbles from a horizontal capillary, the end of which is cut at an angle of  $45^\circ$ . The time of a complete bubble cycle  $\tau$  consists of:

- time  $\tau_1$  – from the moment of bubble genesis to its separation from the capillary;
- time  $\tau_2$  – waiting time until the pressure in the capillary rises above the pressure in the reactor at the level of the capillary.

Then the bubble separation frequency  $f$  will be  $1/\tau$ .

Let the capillary with a diameter  $d_c$  be placed at a depth  $h$  from the surface of the liquid in the reactor. At the moment of separation, the bubble will have a volume  $V$  and a diameter  $d_w$ , and  $d_c < d_w$ . During the time  $\tau_1$ , the bubble will be affected by the forces of gravity  $F_{grav}$ , surface tension  $F_{sur.ten}$  and Archimedes  $F_{Arch}$  (Fig. 5).

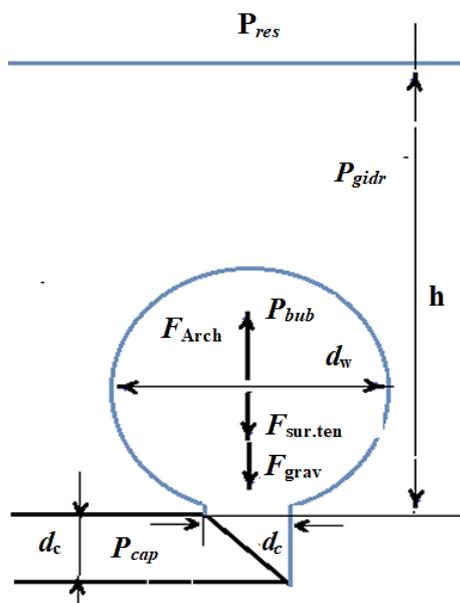


Fig. 5. Forces acting on the bubble

In the process of bubble growth, the first two forces will prevail over Archimedes' force, but will balance out over time:

$$\vec{F}_{grav} + \vec{F}_{sur.ten} - \vec{F}_{Arch} = \vec{0} \Rightarrow \vec{F}_{grav} + \vec{F}_{sur.ten} = \vec{F}_{Arch}, \quad (5)$$

where  $m$  – mass of gas in the bubble;  $g$  – free fall acceleration;  $\sigma$  – surface tension of water;  $\rho_w, \rho_g$  – density of water and natural gas;  $\pi=3.14$ ;  $V_{bub}$  – the volume of the bubble.

After mathematical transformations, the equation for determining the diameter of the bubble at the time of separation can be written in the form:

$$d_w = \sqrt[3]{\frac{6d_c\sigma}{g(\rho_w - \rho_g)}}. \quad (6)$$

The gas density in the bubble is unknown, therefore, to determine  $\rho_g$ , consider the change in pressure in the «capillary – bubble» system. Gas pressure in a stationary bubble according to Bernoulli's law:

$$P_{res} + P_{gidr} + P_{din} = P_{bub} = P_{cap}, \quad (7)$$

where  $P_{res}$  – pressure in the reservoir, MPa;  $P_{gidr} = \rho_w gh$  – hydrostatic pressure of the water column;  $P_{din} = \rho_g \omega^2 / 2$  – dynamic pressure of the moving gas (for a stationary bubble  $\omega = 0 \text{ m/s}$ );  $P_{cap}$  – gas pressure in the capillary;  $P_{bub} = P_{cap}$  – pressure in the bubble.

Based on the gas density  $\rho_g$ , let's calculate the diameter of the separation of the gas bubble from the capillary  $d_w$ .

The speed of movement of the bubble when rising to the surface, taking into account the condition  $\rho_w \gg \rho_g$  [17] can be calculated by the equation:

$$\omega = \sqrt{\frac{4d_w g}{3C}}, \quad (8)$$

where  $C$  – coefficient of resistance of water in the reactor.

The  $C$  coefficient in the event that the conditions are met, namely for the Weber criterion  $We > 1$ , the Reynolds number  $Re > 600$  and the Bond criterion  $VO < 13$ , can be calculated by the Harmonti formula [17]:

$$C = 0.568\sqrt{VO}, \quad VO = \frac{gd_w^2(\rho_w - \rho_g)}{\sigma}. \quad (9)$$

However, even under such conditions, it is quite difficult to record the moment of «cloudiness» of the interphase surface as clearly and unambiguously as possible [16]. Simultaneously with the process of clouding of the interphase surface, a gradual distortion of the reflections of light sources on it is observed (Fig. 2). Moreover, the beginning of their distortion was recorded much earlier than the visual signs of clouding of the mirror interphase surface (bubbles in the liquid or drops in the gas atmosphere). As can be seen in the photo (Fig. 2, b, c), no other visual signs of the beginning of the hydrate formation process, except for the distortion of the reflection of light sources, are practically not observed.

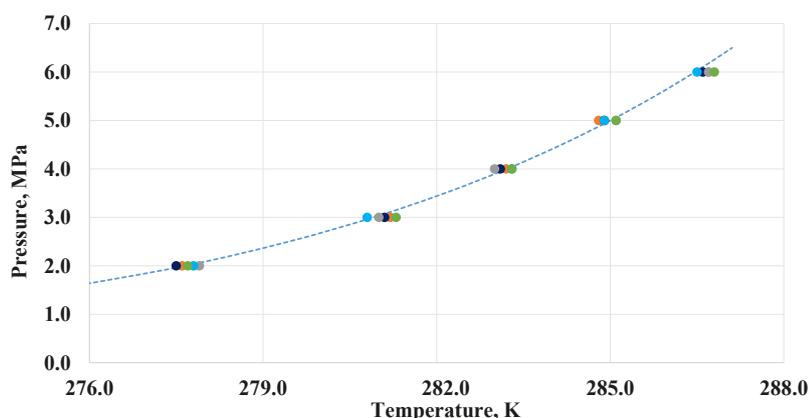
Thus, this optical effect makes it possible to record with high accuracy the changes that occur at the micro level with the mirror interphase «gas – liquid» interface. In our case, it allows to record the process of beginning mass crystallization of gas hydrate.

Therefore, it is expedient to use the moment for fixation of the optical effect of the distortion of the light source reflection on the mirror of the «liquid – gas» interphase surface by gas hydrate microcrystals as the basis of the method of setting the parameters of the beginning of the stage of its mass crystallization. It is expedient to use the method for operational control of the parameters of this process in the conditions of industrial production.

Natural gas of the following composition was used to evaluate the proposed method, %: CH<sub>4</sub> – 87.17; C<sub>2</sub>H<sub>6</sub> – 5.16; C<sub>3</sub>H<sub>8</sub> – 2.48; *i*-C<sub>4</sub>H<sub>10</sub> – 0.67; *n*-C<sub>4</sub>H<sub>10</sub> – 0.90; *i*-C<sub>5</sub>H<sub>12</sub> – 0.12; *n*-C<sub>5</sub>H<sub>12</sub> – 0.17; C<sub>6</sub>H<sub>14</sub> – 0.17; C<sub>7</sub>H<sub>16</sub> – 0.28; CO<sub>2</sub> – 0.17; N<sub>2</sub> – 2.50; He – 0.21. The studied parameters are pressure and temperature at the moment for visual fixation of the beginning of the distortion of the light reflection on the bubbles surface. During the preparatory operations, slight pressure drop and temperature rise as a result of gas dissolution in water were taken into account. The results of the research and their comparison with the equilibrium parameters of hydrate formation, calculated according to the Barrer-Stewart equation, are presented in Fig. 6.

The hydrate formation process at the phase boundary is manifested by the formation of a thin hydrate layer in the form of a film. Such a process is visually fixed by the transformation of the mirror surface of the phase boundary into a matte one. The effect of distortion for the interphase boundary is explained by the formation, growth, massive and chaotic accumulation of hydrate microcrystals at this boundary. Simultaneously with the process of the interphase surface clouding, a significant distortion of the reflection points of the light source is visually recorded. The investigated optical effect allows to record changes occurring at the micro level at the gas-water interface with sufficiently high accuracy.

Thus, the proposed technique consists in establishing the parameters of the moment of mass gas hydrates crystallization based on the fixation of the optical effect of the distortion of the light source reflection on the mirror of the «liquid – gas» interphase surface. It can be used to establish and operationally control the moment of mass crystallization of gas hydrates directly at the objects of the oil and gas industry (during the implementation of technological processes). This will make it possible to effectively prevent clogging of technological equipment by the solid phase of gas hydrate, as well as overuse of hydrate formation inhibitors.



**Fig. 6.** Evaluation of the method of setting the parameters of the beginning of mass gas hydrate crystallization: ●, ●, ●, ●, ● – experimental data; - - - the equilibrium curve of hydrate formation, calculated according to the Barrer-Stewart equation

Thus, the proposed method is intended for use at industrial facilities for gas and oil production and preparation. Since the basis of the technique is the visual control of the interphase surface, so an important limiting factor of its application for control of oil and gas production technological processes is the light transmission (transparency) level of the studied samples for reservoir systems. When it decreases, the transparency (reliability) of the results decreases.

In further studies, it is expedient to evaluate the effectiveness of the proposed method for formation systems with average and high concentrations of hydrate formation inhibitor substances.

#### 4. Conclusions

1. The researched technique consists in establishing the parameters of the initiation moment of mass gas hydrates crystallization at the micro level based on the fixation of optical effects.
2. On the basis of the rank correlation analysis, a close relationship between the equilibrium parameters of hydrate formation and the parameters of the beginning of mass crystallization for gas hydrate in the studied samples is shown.
3. A uniform shift of the fixed parameters for the beginning of mass gas hydrate crystallization from the equilibrium parameters of hydrate formation was established. Such a trend is predictable, since the proposed technique involves fixing the moment of mass crystallization of gas hydrate in a system that has not yet reached a state of equilibrium.
4. The proposed technique can be used in the oil and gas industry for operational control of the technological process. The only limitation of the application of the technique is the low light transmission of the liquid phase for the formation system.

#### Conflict of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

#### Financing

The research was conducted without financial support.

#### Data availability

The manuscript has no associated data.

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