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APPLYING A DIMENSIONAL ANALYSIS METHOD FOR IMPLEMENTING THE GAS HYDRATE TECHNOLOGY WHEN TRANSPORTING AND STORING NATURAL GAS

Larysa Pedchenko
PhD, Associate Professor*
Nazar Pedchenko
Doctor of Philosophy (PhD)*
Mykhailo Pedchenko
Corresponding author
PhD, Associate Professor*
E-mail: pedchenkomm@ukr.net
*Department of Oil and Gas Engineering and Technology
National University "Yuri Kondratyuk Poltava Polytechnic"
Vitaliya Hrytsayenka ave., 24, Poltava, Ukraine, 36011

This study's object is the process parameters for the forced conservation of gas hydrate by a layer of ice to ensure its stability under non-equilibrium conditions. These conditions are atmospheric pressure and a slight negative temperature. However, the application of water in the liquid phase and its subsequent crystallization, with the release of a corresponding amount of process energy, is accompanied by the dissociation of the surface layer of gas hydrate, and therefore the loss of gas. A decrease in the melting level of gas hydrate could be achieved by increasing the cooling level of gas hydrate structures. However, this increases the operating costs of the technology. This paper proposes a variant for substantiating the process parameters for the forced conservation of natural gas hydrate structures.

To this end, by using the dimensionality analysis method, dimensionless complexes were established: Fourier criterion (Fo); criterion characterizing the relative thermal conductivity of bodies (K_ε), Kosovich criterion (Ko), dimensionless temperature criterion Θ_1/Θ_0, and the criterion equation. The coefficient and degrees of dimensionless complexes in the criterion equation were experimentally determined. They make it possible to describe the process of forced conservation of gas hydrate structures regardless of size and shape.

The application of dimensionless dependences and criterion equations could make it possible to determine in a simplified form parameters for the technological process of industrial production of gas hydrate structures with the required properties (stability during transportation and storage under non-equilibrium thermobaric conditions without gas loss).

Based on experimental studies, the limits of the applied parameters of the stability of gas hydrate structures under non-equilibrium conditions have been established. For them, the criterion of dimensionless temperature Θ_1/Θ_0 is within 0.02–0.04

Keywords: gas hydrate technology of natural gas storage, mathematical modeling, dimensional analysis method

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1. Introduction

Natural gas is currently the basis of Ukraine's energy sector. Gas and gas condensate fields are being actively developed to provide the country's population and industry with local hydrocarbons. Further preparation of extracted hydrocarbons and transportation to the consumer requires complex equipment and an extensive pipeline system.

Over 100 gas and gas condensate fields are operated in Ukraine. Among them, a significant part is small. Most of the new fields that are being discovered are small. Often, the construction of a pipeline and the necessary infrastructure for such fields is economically inexpedient.

In addition, since the beginning of the full-scale invasion of the aggressor country into Ukraine, the vulnerability of the entire energy infrastructure of the state has become obvious.

Therefore, to accelerate the utilization of resources from small undeveloped fields and increase the stability of the natural gas supply system, consumers need alternative techniques for transporting it at lower initial costs.

Given the identified tasks, the issue of finding and researching alternative techniques for transporting and storing natural gas for consumers at small settlements is particularly relevant today.

Therefore, research into the problems of storing structures formed under non-equilibrium conditions of hydrate formation with the dimensions necessary for industrial production, with the aim of applying innovative technology for transporting and storing natural gas, is a relevant task.

2. Literature review and problem statement

Currently available commercial natural gas storage technologies are compressed natural gas (CNG) and liquefied natural gas (LNG). However, none of them provides simultaneous large-scale and long-term storage of natural gas. In the case of CNG, because of the need for high pressure (20–25 MPa) and safety issues (explosion hazard) [1], or extremely low storage temperature (–162°C) and boiling problems for LNG [2].

In addition, these technologies are advisable to use only for significant volumes of gas and large deposits.

At the same time, a promising technology is based on the ability of gas and water molecules to form gas hydrates (NGH technology). Natural gas is proposed to be transported and stored as gas hydrates. Up to 164 m^3 of gas can be stored in 1 m^3 of gas hydrate under standard conditions [3]. However, given the properties of gas hydrates, the challenge is to achieve this level of natural gas content during its intensive industrial production and then maintain it during gas hydrate transportation and storage.

Natural gas storage based on gas hydrates has a number of advantages, including lower energy requirements (in most cases), reduced environmental hazards, almost 100% recovery, and higher safety [4]. However, a problem that needs to be further addressed is ensuring maximum stability of the produced gas hydrate to prevent gas losses as a factor of hazard, environmental impact, and technology efficiency.

The temperature of natural gas hydrate (NGH) during transportation remains higher than that of liquefied natural gas (LNG). In addition, NGH transportation is expected to be more flexible in terms of its facilities and equipment. NGH technology may be suitable for transporting natural gas from small gas fields for which LNG technology is not economically viable [5].

Thus, the technology of storing natural gas in hydrate form demonstrates great potential for commercially profitable non-explosive storage on an industrial scale under moderate pressure and temperature conditions [6]. However, the maximum possible competitiveness of this technology, based on the properties of natural gas hydrate, will be provided by the production of gas hydrate structures suitable for long-term storage without gas loss at atmospheric pressure.

In work [7], a detailed economic assessment of NGH technology is provided and a methodology for calculating its economic efficiency is proposed. However, this technology is currently not commercially implemented since it is at the stage of development and improvement of the main elements. Currently, NGH technology requires the design and construction of specialized equipment for intensive production of gas hydrate, as well as the development of parameters for the technological process of this production.

In work [8], the design of a plant for continuous generation of natural gas hydrates and optimal conditions for their generation are considered. However, the proposed technological solutions and process parameters do not sufficiently take into account the need to produce gas hydrate structures for their storage outside the thermobaric conditions of hydrate stability (for example, at atmospheric pressure).

At the same time, natural gas storage using NGH technology would have the lowest cost if the produced gas hydrate were stored under non-equilibrium conditions. Therefore, the parameters for the production of gas hydrate structures that will be stable under such conditions require further research.

A number of papers, for example [9], have investigated the effect of surfactants on the intensification of hydrate formation. However, surfactants may negatively affect the stability of the produced gas hydrate, exhibiting the properties of hydrate formation inhibitors.

In [10], a method of applying a liquid (tetrahydrofuran (THF), cyclopentane (CP), cyclohexane and n-tetradecane) to methane hydrate to enhance its self-preservation effect was investigated. It was found that the applied liquid could form a new layer of hydrate or another hardened layer on the surface of the hydrate, which prevents its dissociation below the freezing point of water. However, it cannot prevent the melting of methane hydrate at positive temperatures. Therefore, this technique may only enhance the stability of the hydrate below the crystallization point of water but cannot stimulate the manifestation of the self-preservation effect.

In [11], the storage duration of CH_4, C_2H_6, C_3H_8 hydrates was investigated from the point of view of industrial application of the technology. The experiment lasted for 3 months at close to equilibrium thermobaric conditions (temperature 253 K and atmospheric pressure). The formation of thermal cracks on the surface of the granules was recorded, which over time were "healed" by a layer of ice up to 100 microns thick. This ensured a high degree of stability of the hydrates. However, the surface of the granules was not completely covered with an ice crust, which only partially prevented gas loss.

In work [12], it is proposed to form gas hydrate into granules and store it at a temperature of 258 K and atmospheric pressure. However, granular gas hydrate has a number of disadvantages. The granules will freeze during storage. This will create problems when unloading it. Also, when using granular gas hydrate, the filling of the volume of vehicles and hydrate storage facilities will be far from maximum.

In [13], the stability of pure methane hydrate and hydrate of a mixture of methane and tetrahydrofuran (THF) formed into cylindrical samples was studied. Hydrate structures were stored at atmospheric pressure and temperatures of 253 K and 268 K and demonstrated long-term stability. However, during long-term storage at 268 K, a gas loss of 7% from methane hydrate – THF, and 40–50% from methane hydrate was recorded.

Important for NGH technology of natural gas storage is the property of anomalous stability of gas hydrate structures – self-preservation by an ice crust [14]. However, the thickness of the formed crust (of the order of hundredths of a millimeter) is not enough for reliable and long-term preservation of gas hydrate during storage.

In work [15], it is shown that the anomalous stability of gas hydrates outside their stability zone below the melting point of ice depends on the type of guest molecule, the morphology of hexagonal ice growing during hydrate dissociation, and the method of dissociation. These results indicate a significant range of parameters on which the stability of gas hydrate depends and therefore emphasize the inadmissibility of counting on the effect of its self-preservation in industrial technology.

In [16], the dissociation of methane hydrates at negative temperatures, which differ in macro- and microstructural parameters, was studied. It was found that the density of the pore distribution in the crust of the formed ice decreases by several orders of magnitude, which significantly changes the dissociation rate. These results emphasize the need for the operation of forced conservation of gas hydrate, since even its constant presence under conditions of significant cooling cannot prevent its dissociation.

In paper [17], a modern understanding of the effect of spontaneous conservation of gas hydrates and a variant of its engineering application in gas storage and transportation technology are outlined. It is shown that self-preservation depends on the porosity of the hydrate, the size of its crystals, and the microstructure of the ice crust formed as a result of hydrate dissociation. However, the spontaneous process of self-preservation is not enough to prevent losses of natural gas during transportation and storage of gas hydrate structures.

In work [18], in order to increase the efficiency of the NGH technology for transportation and storage of natural gas, it was proposed to form the produced gas hydrate into large cylindrical blocks. To increase the stability of the gas hydrate, it was proposed to cement the surface layer of the blocks with ice. For this purpose, water was applied to the surface of these cooled structures. After its crystallization, a gas-impermeable shell was formed in the pores and on the surface of the blocks.

In [19], the positive effect of reducing the porosity of the gas hydrate as a result of compaction on the manifestation of the effect of self-preservation and freezing of the ice layer on its surface was recorded. It was established that for high-quality forced preservation of gas hydrate structures by a layer of ice, the porosity of the gas hydrate should be within 0.07–0.15, the initial temperature should be 248–259 K. However, it is important to establish the relationship of these parameters with the size and shape of the gas hydrate structures, as well as the thermobaric parameters of the implementation of this process.

Thus, regardless of the size and shape of gas hydrate structures, to prevent natural gas losses due to gas hydrate dissociation during transportation and storage under non-equilibrium conditions, the main problem is their reliable conservation. At the same time, regardless of the size and shape of these structures, the set of parameters that determine the possibility of the crystallization process of deposited water in the surface layer of gas hydrate is important. Such parameters, first of all, are the initial and final (after water crystallization) surface temperatures of the gas hydrate structure [19].

Therefore, an important scientific and technical issue related to the NGH technology that needs to be studied is process parameters for the production, transportation, and storage of natural gas hydrate at minimal excess pressure up to atmospheric.

At the same time, all physical parameters of the processes occurring in such systems can be described through dimensionless complexes or similarity criteria [20]. However, in the case of a large number of such parameters (factors), the mutual influence and dependence between the similarity criteria is described by the criterion heat transfer equation. The use of similarity theory or dimensional theory, in the case when the mathematical description of the process by differential equations is complicated, allows us to obtain criterion equations based on theoretical solutions. Based on the results of experiments and data processing, we can obtain the coefficients and powers of these equations. The parameters included in the equations do not depend on geometric dimensions, properties, scales, and are universal. It is such equations that have found wide application for the study of heat transfer processes [21].

To use the results of experimental studies when implementing NGH technology for transportation and storage of natural gas, it is necessary to establish the dependence between the parameters of the studied system. That is, to determine the parameters of gas hydrate structures for their industrial production, such relationships between parameters should be represented in the form of dimensionless complexes and a criterion equation.

3. The aim and objectives of the study

The purpose of our study is to determine the criteria and parameters for the technological process of producing gas hydrate structures regardless of their shape and size based on the dimensional analysis method. This will make it possible to calculate the process parameters for the forced conservation of gas hydrate structures by a layer of ice, which will minimize the dissociation of the surface layer of gas hydrate during the crystallization of applied water and prevent excessive energy (cold) consumption.

To achieve this aim, the following objectives were accomplished:

– based on the analysis of the mathematical model of the process of conservation of gas hydrate structures by a layer of ice, to establish the physical parameters that affect the course of the studied heat transfer process;

– to use the method of dimensional analysis (dimensional theory), namely Buckingham's theorem, to establish dimensionless complexes that are included in the process of conservation of gas hydrate structures, and the criterion equation that "controls" the process of conservation of gas hydrate structures regardless of their size;

– to assess correctness of the established dimensionless complexes and coefficients and powers in the criterion equation for describing the studied process based on experimental data.

4. The study materials and methods

The object of our research is process parameters for the forced preservation of gas hydrate by a layer of ice to ensure its stability under non-equilibrium conditions.

The main hypothesis of the research assumes that the process of preserving gas hydrate structures by a layer of ice depends on a number of physical parameters that are mutually related in certain ratios. Therefore, it was assumed that the physical parameters of the process could form certain dimensionless complexes connected in a criterion equation. This is what had to be confirmed by experimental studies.

Under industrial conditions, it is unrealistic to arrange a production line for the forced preservation of gas hydrate structures by a layer of ice in a high-pressure chamber (for example, above 3.5 MPa). Therefore, it is assumed that this operation could be implemented in a sealed and thermally insulated chamber at atmospheric pressure. Based on this, the research was carried out in a thermally insulated chamber with forced ventilation at atmospheric pressure.

Experimental studies were performed to determine the influence of the initial temperature of gas hydrate structures on the crystallization time of applied water and the change in the thickness of the frozen ice layer on the time of its crystallization process. For this purpose, water was applied to the surface of cylindrical samples (diameter 0.08 m, height 0.11 m) and porosity 0.12 with different initial temperatures by spraying in amounts of 15, 30, and 45 ml according to the plan of a single-factor experiment (Table 1).

In [19], a study was carried out on the process of preserving a gas hydrate structure (cylindrical block) with a layer of ice to prevent losses of natural gas during the implementation of the NGH technology of its transportation and storage. It was established that the desired conservation effect is achieved with an ice layer thickness of 0.002 m.

The main factor influencing the process of crystallization of the ice layer on the surface of a gas hydrate sample formed into blocks is a decrease in entropy due to the cold accumulated in it.

Table 1

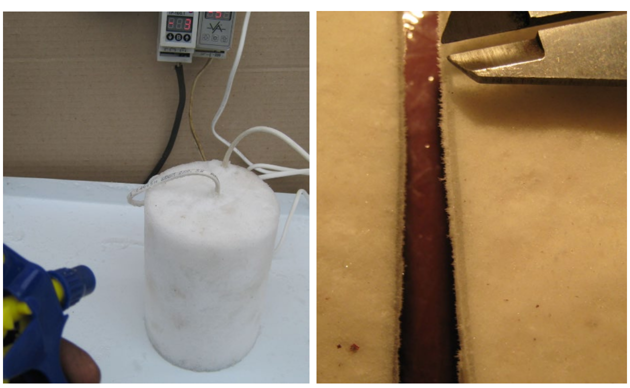
Investigated factors of the crystallization process of deposited water on the surface of gas hydrate samples

Factor		
Resulting factor	Factor under investigation	
Crystallization time, s	Initial sample temperature, K	253.0
		256.0
		259.0
Crystallization time, s	Thickness of the wetted layer (ice layer) on the sample surface, m	0.002
		0.004
		0.006

The process of freezing the ice layer on the surface of a cylindrical gas hydrate sample with a porosity of 0.1–0.15 was carried out under adiabatic conditions. The sample was pre-cooled to negative temperatures. Water with a temperature close to 273 K and a volume equal to the volume of pores of the conditionally selected surface part of the sample was evenly applied to its surface. The thickness of this layer is much less than the radius of the sample, but not less than 0.002 m. The applied water completely fills the pore space of this layer and after some time crystallizes, making it impermeable to gas.

If the volume of applied water on the surface of the cylinder is insignificant, and a sufficient amount of cold is accumulated in the sample (conditionally an internal source of energy), then its crystallization occurs in a certain time τ. Under adiabatic conditions, the heat of the exothermic process is removed to the center of the sample.

A sign of the completion of the crystallization process of the applied water was the beginning of a decrease in the temperature of the surface layer of the gas hydrate sample. Fig. 1, *a* shows the cementation process of the surface layer of the sample, Fig. 1, *b* – the cemented surface layer in the section of the sample.



a *b*

Fig. 1. Forced preservation of a gas hydrate sample: *a* — the process of applying water to the surface of a pre-cooled sample; *b* — view of the cemented surface layer in the cross-section of the sample

Table 2 gives the results of an experiment on the freezing of an ice layer on the surface of a gas hydrate sample; Table 3 gives the dependence of the thickness of the frozen ice layer on the crystallization time of applied water.

Theoretical, empirical, and statistical methods were used in the research process.

Table 2

Dynamics of changes in the surface temperature of a gas hydrate sample during the crystallization of deposited water when a 2 mm thick ice layer is frozen and the initial sample temperature is 259 K (maximum for the implementation of this process)

Time, s	Sample surface temperature, K				
	Repetitions				
	1	2	3	4	5
0	259.0	258.9	258.7	259.1	259.1
40	263.1	263.0	264.5	263.2	263.9
80	264.8	265.0	266.4	265.4	266.2
120	266.4	266.0	267.3	266.3	267.2
160	267.5	266.6	267.8	267.0	268.1
200	268.0	267.3	268.1	267.6	268.6
240	268.2	267.6	268.2	267.7	268.8
280	268.5	267.8	268.2	267.9	268.9
320	268.6	268.0	268.2	268.0	268.8
360	268.5	267.9	268.2	268.0	268.8
400	268.4	267.9	268.1	268.1	268.7
440	268.3	267.9	268.0	267.9	268.6
480	268.1	267.8	267.9	267.8	268.3
520	267.9	267.7	267.8	267.8	268.2
560	267.7	267.5	267.7	267.6	268.1
600	267.5	267.4	267.6	267.5	268.0
640	267.4	267.3	267.5	267.4	267.9
680	267.3	267.3	267.3	267.3	267.8
720	267.1	267.2	267.2	267.2	267.6
760	267.0	267.2	267.1	267.0	267.5
800	266.8	267.1	267.0	266.9	267.3
840	266.7	267.0	266.8	266.7	267.2
880	266.6	266.9	266.7	266.7	267.1
920	266.5	266.8	266.6	266.5	266.9
960	266.4	266.7	266.4	266.3	266.7
1000	266.2	266.6	266.3	266.3	266.6
1040	266.1	266.5	266.2	266.2	266.5
1080	266.0	266.4	266.0	266.2	266.4
1120	265.8	266.4	265.9	266.1	266.3
1160	265.7	266.3	265.8	266.1	266.2
1200	265.5	266.2	265.6	265.9	266.1

Table 3

Dependence of the thickness of the frozen ice layer on the surface of a gas hydrate sample with the corresponding amount of applied water on the time of its crystallization (initial sample temperature 259 K)

Crust thickness, m	Crystallization time, s		
	repetitions		
	1	2	3
0.002	274	296	277
0.004	695	712	756
0.006	1267	1295	1302

5. Results of determining the criteria and parameters for the production of gas hydrate structures based on the dimensional analysis method

5. 1. Establishment of physical parameters that affect the course of the studied heat transfer process

To describe the process of cementation of the surface of a gas hydrate sample with ice, a boundary value one-dimensional problem of non-stationary thermal conductivity was stated. The propagation of thermal energy in the surface wetted layer occurs due to thermal conductivity and is described by the following equation:

$$c_{cr}\rho_{ck}\frac{\partial T_{cr}}{\partial \tau}=\lambda_{cr}\left(\frac{\partial^2 T_{cr}}{\partial x^2}+\frac{1}{x}\frac{\partial T_{cr}}{\partial x}\right)+\rho_{ice}L\frac{dV_w}{v_{cr}d\tau},$$

$$A\ge V_w\ge 0,\ 0<x<l,\ \tau>0, \quad (1)$$

where T_{cr} is the temperature of the wetted layer of gas hydrate, K; τ is the crystallization time, s; l is the thickness of the wetted layer of gas hydrate, m; V_w, v_{cr} are the volumes of applied water and the surface layer of gas hydrate wetted with water, respectively, m^3; c_{cr} is the specific heat capacity of water-wetted gas hydrate, J/kg·K; ρ_{ck}, ρ_{ice} are the densities of the wetted layer and ice, respectively, kg/m^3; λ_{cr} is the thermal conductivity coefficient of the wetted layer, W/(m·K); L is the specific heat of crystallization of water, J/kg; A is the volume of applied water per sample, m^3.

The spread of thermal energy in the unwetted (dry) part of the gas hydrate sample over a certain time also occurs due to thermal conductivity and is described by the following equation

$$\frac{\partial T_{g.h}}{\partial \tau}=a_{g.h}\left(\frac{\partial^2 T_{g.h}}{\partial x^2}+\frac{1}{x}\frac{\partial T_{g.h}}{\partial x}\right),\ \tau>0,\ l<x<R, \quad (2)$$

where $T_{g.h}$ is the temperature of the dry part of the gas hydrate sample, K; a is the thermal conductivity coefficient of the dry gas hydrate, m^2/s; R is the radius of the sample, m.

Initial conditions

$$T_{cr}(x,0)=T_0,\ T_{g.h}(x,0)=T_1. \quad (3)$$

For a correct problem statement, we set the condition of thermal symmetry in the center of the sample. It is assumed that the gas hydrate sample is under adiabatic conditions. The boundary conditions of the fourth kind correspond to the problem and take the following form:

$$T_{g.h}(R-l;\tau)=f(x);\ T_{cr}(l;\tau)=\phi(x,\tau), \quad (4)$$

$$\lambda_{cr}\frac{\partial T_{cr}(x,\tau)}{\partial x}=\lambda_{g.h}\frac{\partial T_{g.h}(x,\tau)}{\partial x}, \quad (5)$$

where $\lambda_{g.h}$ is the thermal conductivity coefficient of dry gas hydrate, W/(m·K).

The rate of heat transfer by thermal conductivity is described for both selected parts of equations (1) and (2) by terms $\left(\frac{\partial^2 T_{cr}}{\partial x^2}+\frac{1}{x}\frac{\partial T_{cr}}{\partial x}\right)$ and $\left(\frac{\partial^2 T_{g.h}}{\partial x^2}+\frac{1}{x}\frac{\partial T_{g.h}}{\partial x}\right)$. They describe the rate of change of internal energy per unit volume. The heat of crystallization of water – an internal source of energy – is described by term $\rho_{ice}L\frac{dV_w}{V_{cr}d\tau}$ from equation (1) [19].

Analysis of the model of the process of preservation of gas hydrate structures by a layer of ice revealed that the equations include a number of physical parameters that directly affect the course of the crystallization process. Such parameters are initial t_0 and final t_1 temperatures, coefficients of thermal conductivity λ_1 and thermal diffusivity a_1 of the surface layer of the sample, and coefficients of thermal conductivity λ_2 and thermal diffusivity a_2 of its central part.

They should also include the amount of heat of crystallization of saturated water Q_{cr}; the amount of energy Q_{heat}, to change the temperature of the wetted layer and adjacent dry gas hydrate to the final temperature.

5. 2. Establishing dimensionless complexes that "control" the process of gas hydrate structure conservation

Thus, based on our analysis of the mathematical model of the thermal process (1) to (5), physical parameters have been established that directly affect the process of crystallization of deposited water and can be included in dimensionless complexes or similarity criteria.

To confirm or refute the hypothesis about the presence of dependences between the studied parameters in the form of interconnected dimensionless complexes, the theory of similarity (dimension methods) should be applied. In addition, based on the results of the experiments, the coefficients and powers of the criterion equation should be determined.

When confirming the hypothesis, the results can be used to simplify the calculation of geometric and technological parameters of gas hydrate structures for their high-quality conservation by a layer of ice in the NGH technology of natural gas transportation and storage.

Ensuring the reliability of the results of theoretical research for gas hydrate structures of different sizes requires necessary and sufficient conditions for the existence of similarity. So, to confirm the hypothesis, we shall use the first Newton-Bertrand theorem and the second Buckingham π-similarity theorem (hereinafter referred to as the π-theorem). According to the Newton-Bertrand theorem, for similar phenomena there must be the same similarity criteria, regardless of the type of model constructed, but numerically the same for similar phenomena. The first theorem forms the necessary conditions for the existence of similarity (the same similarity criteria).

To construct a criterion equality consisting of dimensionless complexes, one should use the π-similarity theorem. According to it, any physical dependence between n variables can be expressed in terms of n-k dimensionless complexes, where k is the number of basic dimensions. Therefore, according to the π-theorem, a physical process written by a mathematical equality can be represented by a functional dependence between the established similarity criteria obtained from the parameters included in the process. The second theorem establishes the possibility of representing the differential equation of a physical process as a function of appropriately constructed dimensionless similarity criteria or dimensionless complexes.

Based on our analysis of the dimensions of physical parameters of a water crystallization process in the surface layer of a gas hydrate sample, it is possible to establish the criteria for their similarity and derive the criterion equation, even in the absence of differential equations of the studied physical process.

The π-theorem, where all dimensionless parameters (similarity criteria) are denoted as π, is solved by the method of sequential elimination of dimensions.

According to Buckingham's π-theorem, it is possible to establish the number of independent dimensionless criteria [22]

$$k = n - m - 1, \quad (6)$$

where k is the maximum number of dimensionless criteria; n_1 is the number of dimensional parameters that define and characterize the process; m_1 is the number of basic dimensionalities of these parameters (quantities).

Therefore, it is necessary to establish dimensionless parameters that relate the initial temperature t_1 and the final temperature t_2 in the surface layer with a thickness r, which changes over time τ, with:

– coefficients of thermal conductivity λ_1 and thermal diffusivity a_1 in the surface layer and coefficients of thermal conductivity λ_2 and thermal diffusivity a_2 of some adjacent dry part of the gas hydrate;

– the amount of heat Q_{cr}, crystallization of water per unit volume of the surface layer;

– the amount of energy Q_{heat} spent to change the temperature of the wetted surface layer and some adjacent dry part of the gas hydrate to its final temperature.

According to the theorem

$$\pi = a_1^a \cdot \tau^b \cdot r^c \cdot \lambda_1^d \cdot \lambda_2^e \cdot a_2^f \cdot t_1^g \cdot t_2^h \cdot Q_{cr}^i \cdot Q_{heat}^j. \quad (7)$$

Given the dimensionality of the selected parameters:

$$[a] = L^2T^{-1}; [\tau] = T; [r] = L; [t] = \Theta;$$

$$[\lambda] = LMT^3\Theta^{-1}; [m] = M; [Q] = ML^2T^{-2}. \quad (8)$$

After substituting (8) into equation (7), a π-equation is obtained in the form

$$\pi = \left(\frac{L^2}{T}\right)^a \cdot T^b \cdot L^c \cdot \left(\frac{LM}{\tau^2\Theta}\right)^d \cdot \left(\frac{LM}{\tau^2\Theta}\right)^e \times$$
$$\times \left(\frac{L^2}{T}\right)^f \cdot \Theta^g \cdot \Theta^h \cdot \left(\frac{ML^2}{T^2}\right)^i \cdot \left(\frac{ML^2}{T^2}\right)^j. \quad (9)$$

According to equation (6), the number of physical parameters with certain dimensionalities is $n = 10$, the number of basic dimensionalities of these parameters is $m = 4$. Then the π-equation will contain 5 dimensionless criteria, namely:

$$\pi = \left(\frac{L^2}{T}\cdot\frac{T}{L^2}\right)^b \cdot \left(\frac{L^2}{T}\cdot\frac{T}{L^2}\right)^c \cdot \left(\frac{T^2\Theta}{ML}\cdot\frac{ML}{T^2\Theta}\right)^e \times$$
$$\times \left(\frac{\Theta}{\Theta}\right)^1 \cdot \left(\frac{ML^2}{T^2}\cdot\frac{T^2}{ML^2}\right)^j, \quad (10)$$

$$\pi = \left(\frac{a_1}{a_2}\right)^b \cdot (Fo)^c \cdot \left(\frac{\lambda_2}{\lambda_1}\right)^e \cdot \left(\frac{\Theta}{\Theta}\right)^1 \cdot (Ko)^j =$$
$$= K_a^b \cdot Fo^c \cdot K_\lambda^e \cdot \left(\frac{\Theta_1}{\Theta_2}\right)^1 \cdot Ko^j, \quad (11)$$

$$\frac{\Theta_2}{\Theta_1} = K_a^b \cdot Fo^c \cdot K_\lambda^e \cdot Ko^j, \quad (12)$$

where K_a – criterion characterizing the relative thermal inertia of bodies; Fo – Fourier criterion, K_λ – criterion characterizing the relative thermal conductivity of bodies; Ko – Kosovich criterion.

Due to the known dimensionless criteria, for example K_a and K_λ it is possible to write the dimensionless criterion, which describes the relative thermal activity of bodies

$$K_\varepsilon = K_\lambda / \sqrt{K_a}. \quad (13)$$

Then the criterion equation, including the selected dimensionless complexes, will take the following form

$$\frac{\Theta_2}{\Theta_1} = A' \cdot Fo^\alpha \cdot K_\varepsilon^\beta \cdot Ko^\gamma, \quad (14)$$

where A', α, β, γ are the empirical coefficients and powers of dimensionless complexes; $Fo = a\tau/r^2$ is the Fourier criterion.

The Kosovich criterion is defined as

$$Ko = \frac{Q_{cr}}{Q_{heat}} = \frac{Lw\rho_w r}{c_{ice.g.h}\rho_{ice.g.h}(T_1 - T_2)(1-w)(r+0.002)}, \quad (15)$$

where L is the energy of water crystallization, J/kg; w – humidity of gas hydrate mass, kg/kg; c_{ice} – specific heat capacity of the surface layer of gas hydrate with ice in the pores; T_1, T_2 – temperature of the surface layer at the beginning and at the end of the water crystallization process, respectively, K; ρ_w, $\rho_{ice.g.h}$ – density of water and density of the surface layer of gas hydrate with ice in the pores, respectively, kg/m^3.

5. 3. Assessing correctness of the established dimensionless complexes and the criterion equation

According to the results of statistical processing of the results of the experiment on the preservation of a gas hydrate sample by a layer of ice (Table 2), a regression equation (16) was derived, which describes the dependence of the temperature change of its surface layer on time. The plot of this dependence for the initial temperature of the gas hydrate sample at 259 K is shown in Fig. 2

$$T = -1.53 \cdot \left(\ln\left(\frac{\tau}{330} + 0.08\right)\right)^2 - 4.77 + 273, \quad (16)$$

where T is the sample surface temperature, K; τ is the crystallization time of the deposited water, s.

Based on the statistical treatment of results from our experiment to establish the dependence of the thickness of the formed ice layer on the surface of a gas hydrate sample on the crystallization time of applied water (Table 3), the following regression equation (17) was derived

$$r = 18.4 \cdot \tau^2 + 103.4\tau, \quad (17)$$

where r is the thickness of the formed ice layer on the surface of a sample, m; τ is the crystallization time of applied water, s.

The plot of this dependence for the initial temperature of a gas hydrate sample of 259 K is shown in Fig. 3.

Thus, the duration of crystallization of water deposited on the surface of a gas hydrate sample depends on the initial temperature of the sample and the amount of water deposited.

Thus, the dimensionless complexes Fo (Fourier criterion), K_ε (criterion characterizing the relative thermal conductivity of bodies), Ko (Kosovich criterion), and the type of the criterion equation were established. To check the possibility of their application to describe the process of forced conservation of gas hydrate by an ice layer, and to establish empirical coefficient A' and degrees for dimensionless complexes in the criterion equation, experimental data were used.

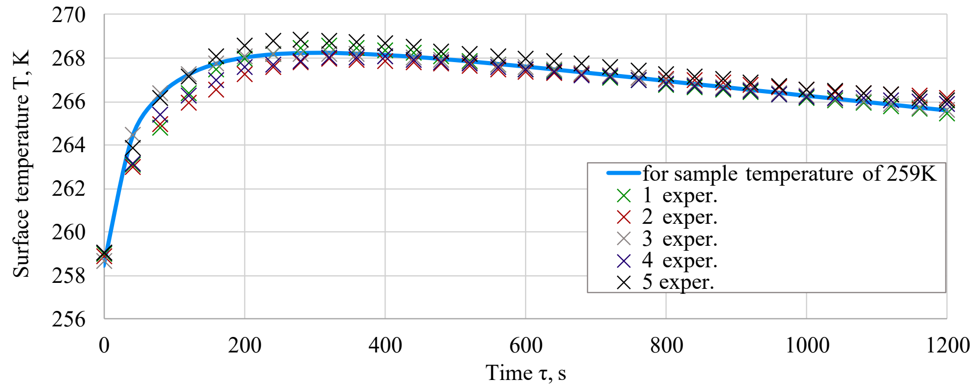


Fig. 2. Dependence of change in the surface temperature of a gas hydrate sample during the crystallization of deposited water on time (initial temperature 259 K, correlation coefficient $r = 0.92$)

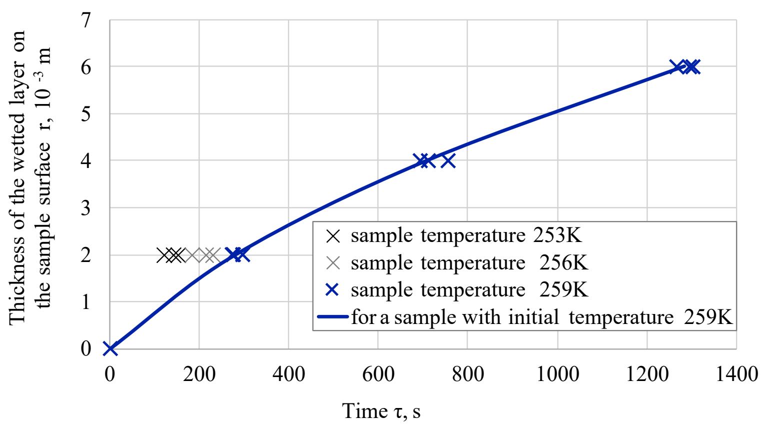


Fig. 3. Dependence of the thickness of the formed ice layer on the surface of a gas hydrate sample on the crystallization time of applied water

During statistical treatment, a three-factor regression analysis of the criterion equation (14) was performed. The data for establishing the empirical coefficient of the criterion equation and powers for dimensionless complexes are summarized in Table 4.

Table 4

Data for establishing the empirical coefficient of the criterion equation and the powers for dimensionless complexes

Initial sample temperature T_1, s	Ice layer thickness r, m	$Fo = X_1$	$K_\varepsilon = X_2$	$Ko = X_3$	Θ_2/Θ_1
259	0.0019	15.37	1.672	1.1771	1.023
259	0.0021	17.11	1.672	1.1771	1.027
259	0.0020	16.10	1.672	1.3733	1.023
256	0.0020	12.96	1.672	1.1771	1.027
256	0.0019	10.32	1.672	1.1771	1.027
256	0.0021	12.06	1.672	1.3733	1.023
253	0.0022	8.64	1.672	1.0300	1.032
253	0.0021	7.96	1.672	1.1771	1.028
253	0.0023	6.79	1.672	1.3733	1.024
259	0.0038	8.92	1.670	1.0986	1.035
259	0.0040	8.32	1.670	1.2360	1.031
259	0.0039	7.97	1.670	1.0986	1.035
259	0.0058	5.97	1.667	1.0594	1.039
259	0.0059	6.41	1.667	0.9631	1.042
259	0.0061	6.22	1.667	1.0594	1.039

To establish empirical coefficients for dimensionless complexes, by using mathematical transformations and substitutions:

$$Y = \ln\frac{\Theta_2}{\Theta_1};\ d_0 = \ln A;\ d_1X_1 = \ln Fo^{\alpha};$$

$$d_2X_2 = \ln K_\varepsilon^{\beta};\ d_3X_3 = \ln Ko^{\gamma}, \quad (18)$$

and the least squares method, the following regression coefficients and linear dependence were obtained:

$$d_0 = -1.87;\ d_1 = -0.02;\ d_1 = -0.02;$$

$$d_2 = 3.91;\ d_3 = -0.01, \quad (19)$$

$$Y = -0.02X_1 + 3.91X_2 - 0.01X_3 - 0.187. \quad (20)$$

The influence of each of the defined dimensionless criteria on the resulting dimensionless temperature criterion Θ_2/Θ_1 was established by the natural elasticity coefficients:

$$E_{x1} = d_1\frac{x_1}{Y} = -0.82;$$

$$E_{x2} = d_2\frac{x_2}{Y} = 36.2;$$

$$E_{x3} = d_3\frac{x_3}{Y} = -0.3, \quad (21)$$

where X_i – average values of the studied factors and the resulting characteristic.

According to the elasticity coefficients, the maximum influence is exerted by K_ε, which characterizes the relative thermal conductivity of bodies. When the temperature changes by 1%, the Fourier criterion Fo and the Kosovich criterion Ko change, respectively, only by 0.8% and 0.3% (inverse dependence). However, the criterion characterizing the ratio of thermal activity of both selected parts changes directly proportionally by 36.2%.

To confirm the main hypothesis of the study – the course of the process of preservation of the gas hydrate structure by a layer of ice depends on a number of interconnected physical parameters and their ratios – the coefficient of determination was established. The quality of approximation of the studied dependences by the regression equation and the adequacy of the established empirical coefficients and powers of the criterion equation was $R^2 = 0.95$.

The regression equation, which consists of similarity criteria Fo, K_ε and Ko, controls about 95% of change in the dimensionless temperature criterion Θ_2/Θ_1 and confirms the accepted hypothesis and takes the following form

$$\frac{\Theta_2}{\Theta_1} = 0.154 \cdot Fo^{-0.02} \cdot K_\varepsilon^{3.91} \cdot Ko^{-0.01}, \quad (22)$$

where A', α, β, γ are the empirical coefficients and powers of dimensionless complexes, respectively. $A' = 0.154$, $\alpha = -0.02$, $\beta = 3.91$, and $\gamma = -0.01$.

Fig. 4 shows the dependence of temperature ratio in the water-wetted surface layer of a gas hydrate sample on its thickness in the coordinates $\Theta_2/\Theta_1(Fo)$.

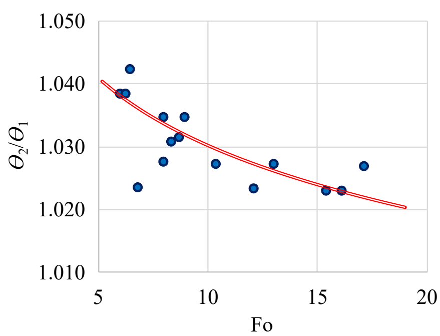


Fig. 4. Dependence of temperature ratio in the water-wetted surface layer of a gas hydrate sample on its thickness in the coordinates Θ_2/Θ_1(Fo)

6. Discussion of results based on determining the criteria and parameters for the production of gas hydrate structures

Based on our analysis of the mathematical model and the results of experimental studies on the process of conserving gas hydrate structures, a number of parameters were established on which the possibility of the process of water crystallization in the surface layer depends: (1) to (5). Theoretically, using the π-theorem and the method of sequential elimination of dimensions, dimensionless complexes Fo, K_ε and Ko were obtained from the set of derived parameters. The complexes were used to describe heat exchange in the wetted surface layer and the adjacent layer of dry gas hydrate, which "controls" the process of conservation of the gas hydrate structure by an ice layer regardless of its size (14).

Correctness of the resulting dimensionless complexes for describing the studied process was confirmed during statistical processing and analysis of experimental data. The regression equation (22) confirms the main hypothesis of our study about the mutual connection and correlation between the parameters of the process of formation of a cemented layer on the surface of a gas hydrate structure. In this case, the similarity criteria Fo, K_ε and Ko control up to 95% of change in the dimensionless temperature criterion.

The Kosovich criterion, calculated from equation (15), takes into account the porosity of gas hydrates and may vary within wide limits. Regardless of the shape of the formed gas hydrate structures, with a sufficiently low porosity (0.1–0.15), the energy of the wetted and adjacent dry parts of the gas hydrate will be sufficient to neutralize the heat of the exothermic process of water crystallization. For example, for a porosity of 0.11 and a thickness of the cemented surface layer up to 0.006 m, the Kosovich criterion (Ko) is less than 1.

The defined criterion equation and dimensionless complexes could be used to establish relationships between the required parameters. For example, the dependence of change in the dimensionless temperature criterion of a gas hydrate structure (surface ice layer) on the Fourier criterion (Fig. 4).

One of the parameters for the Fourier criterion is the thickness of the cemented layer on the surface of a gas hydrate structure of arbitrary shape and different initial temperatures. According to [18], a cemented layer with a thickness of up to 0.002 m is sufficient to prevent gas losses. Therefore, experimental studies were carried out for a surface layer with a thickness of 0.002–0.006 m and an initial temperature within 253–259 K. These temperature limits, in accordance with the equilibrium conditions of natural gas hydrate formation, are appropriate for the industrial production of gas hydrate structures. The graphical dependence $\Theta_2/\Theta_1(Fo)$ in Fig. 4 includes the Fourier criterion calculated for the thickness of the cemented layer within 0.002–0.006 m and the specified initial temperatures.

Therefore, according to the plot (Fig. 4) and regardless of the initial temperature of a gas hydrate structure, the temperature ratio in the surface layer is within 0.02–0.04.

Determining the parameters for complex technological processes, for example, the forced conservation of gas hydrate structures, requires solving a system of differential equations. However, the obtained criterion equation and dimensionless complexes are a simplified version of establishing the dependence between the main parameters that regulate the process under study (conservation of the surface layer of the gas hydrate structure by a layer of ice). In this case, the defined dimensionless complexes control up to 95% of change in the temperature criterion. It is the established limits of change in the resulting criterion that will ensure adequate selection of parameters for the technological process and gas hydrate structures, regardless of their shape. This would prevent losses of natural gas during its transportation and storage.

However, the application of the method of dimensionless complexes for use in industrial production may be limited by the high porosity of the obtained gas hydrate structures. As shown above, the permissible porosity limits for high-quality conservation are 0.1–0.15. At the same time, the extreme limits of the dimensionless Kosovich criterion should be close to 1. That is, the cold reserve of the gas hydrate structure should be sufficient for the crystallization of the deposited water.

Despite the simplicity of the application of the established criteria, some simplification of the results is attributed to the disadvantages, but this does not affect the possibility of using the dependences for calculations in industrial production and the implementation of gas hydrate technology for transporting natural gas.

Further studies should be aimed at clarifying the limits of application of our method. They may include additional investigation of wider limits for the initial temperature, narrowing of the time limits and geometric dimensions of gas hydrate structures.

7. Conclusions

1. Based on the results from our analysis of the mathematical model of cementing the surface of a gas hydrate structure with a layer of ice, the parameters that affect the course of

this process have been established. These parameters are the initial and final temperatures of the surface layer, the amount of heat of crystallization of water in it, the amount of energy required to change the temperature of the surface layer to the final temperature.

2. The application of the dimensional analysis method (Buckingham's π-theorem and the method of sequential elimination of dimensions) has made it possible to establish dimensionless complexes to describe the process of cementing the surface of gas hydrate structures regardless of size, as well as derive a criterion equation. Among them are the Fourier similarity criterion Fo, the criterion characterizing the relative thermal conductivity of bodies K_ε, the Kosovich criterion Ko, and the dimensionless temperature criterion Θ_2/Θ_1. The established boundaries of the resulting criterion will make it possible to ensure an adequate selection of parameters for the technological process of producing gas hydrate structures regardless of their shape and size. This will prevent losses of natural gas during its transportation and storage in the form of gas hydrates.

3. To establish correctness of the defined criterion dependence and dimensionless complexes for describing the studied process, experimental data were used. The limits of the applied parameters of the stability of gas hydrate structures under non-equilibrium conditions, regardless of their shape and size, were experimentally established: porosity, 0.1–0.15; initial temperature, 253–259 K; thickness of the cemented surface layer, 0.002–0.004 m. For the specified parameters, the dimensionless criterion of temperature ratio in the cemented surface layer is within 0.02–0.04. Based on regression analysis, an approximation equation for dimensionless complexes with empirical coefficients and powers was derived. The determined criteria control up to 95% of change in the dimensionless temperature criterion for the wetted surface layer and the adjacent layer of dry gas hydrate.

Conflicts of interest

The authors declare that they have no conflicts of interest in relation to the current study, including financial, personal, authorship, or any other, that could affect the study, as well as the results reported in this paper.

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Data availability

All data are available, either in numerical or graphical form, in the main text of the manuscript.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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