



## **Thermophysical-based Effect of Gas Hydrates Self-Preservation**

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### **1. Introduction**

The industrial use of gas hydrates (GH) can play a large role in supplying humanity with energy. Gas hydrates are considered to be the best alternative fuel in many countries (Koh 2012). However, effective technologies for the gas hydrates production, storage and transportation are still under development (Kvenvolden 1994, Pavlenko et al. 2017). The production of methane from natural gas hydrates is closely related to studies of its dissociation under various conditions. At atmospheric pressure and positive ambient temperature, gas hydrates gradually dissociate into gas and water. The study of the GH dissociation process enables to determine the optimal conditions for their transportation and storage, to minimize gas losses.

An important feature of gas hydrates is the possibility of a long stay in nonequilibrium conditions – the effect of self-preservation. It enables to simplify and reduce the cost of their storage and transportation significantly. However, under nonequilibrium conditions, hydrate dissociation, although it slows down, does not totally stop. Despite the fact that natural gas is a flammable and “greenhouse” gas, the problem of its release during transportation and storage is very important. That is why, the tasks associated with the gas hydrates dissociation are of great practical importance.

### **2. Review of the latest research sources and publications**

Two ways of supplying heat for GH dissociation are possible: surface and bulk. In (Misyura 2016), methane hydrate dissociation at negative temperatures in porous medium was considered. The transportation hydrodynamics of a dissociating hydrate mixture with a liquid was studied in (Zerpa 2013). Several authors

investigated the behavior of natural gas hydrates under water (Sabodh K. Garg et al, 2008). The hydrate dissociation effect on the soil elastic properties is considered in (Jeffrey et al. 2011). In (Thomas et al. 2010), it was proposed to synthesize and store hydrate in sealed containers under pressure. Considering that the minimum pressure for the methane hydrate formation is 35-40 bar, such tanks have too much metal consumption. The basic design for the hydrates storage facilities in the form of a hemisphere has been studied in (Hailu & Thor 2010). The storage temperature regime depends on the thermal characteristics of its tent and base and determines hydrate shelf life and its dissociation intensity.

The study of heat and mass transfer during gas hydrates self-preservation is important not only on the practical side, but also from a theoretical perspective. dissociation of gas hydrates thermophysical parameters The problems of non-stationary thermal conductivity with moving boundary conditions are called Stefan problems. Ice melting, metal solidification, wood maturing are just some examples of such tasks application. Analytical solutions of two-dimensional problems of nonlinear heat conduction are known only for separate, relatively simple cases (Pavlenko 2018, Pavlenko & Szkarowski 2018). The most universal way to solve them is the use of digital methods (Pavlenko 2020, Pavlenko et al. 2014), which can be easily implemented using computer technology. However, digital methods impose their limitations (Pavlenko 2019): the need to solve a system of a large number of linear algebraic equations is a small time step, which leads to cost increase of computer time for calculation (Dąbek et al. 2018). Using the method of grids dynamical adjustment (Dąbek et al. 2016) requires a complex algorithm to implement other problems.

Thus, an analysis of literature indicates an insufficient amount of information regarding the temperature regime of gas hydrate dissociation under nonequilibrium conditions and the absence of a mathematical model for its calculation.

### **3. The purpose and objectives of the study**

The aim of this work is to study the thermophysical features of the gas hydrates dissociation under nonequilibrium conditions. For a quantitative assessment of the dissociating GH temperature regime, a number of experimental studies should be carried out. For this, a number of tasks have to be solved:

1. To develop an experimental setup for performing GH studies of the temperature regime of array dissociation.
2. To conduct a series of field experiments with propane hydrate of various concentrations to determine the temperature regime of dissociating GH under long-term storage.

3. To develop a mathematical model of thermal processes occurring during the gas hydrates dissociation.

#### **4. Experimental studies of GH dissociation with surface heat input**

The dissociation rate of gas hydrate depends on the heat flow brought to its surface. Therefore, the temperature on the gas hydrate surface is an important factor that needs to be clarified. To determine the temperature regime on the surface and in the deeper layers of a dissociating gas hydrate, experimental research methods are presented. In field experiments, propane gas hydrate is used. It has thermophysical characteristics similar to methane GH, however it does not require high pressures to get it.

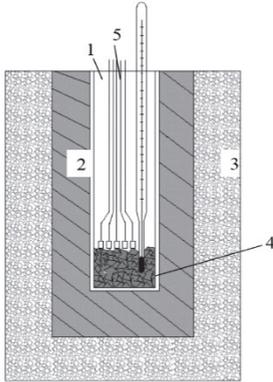
To carry out experimental studies to determine the temperature on the dissociating gas hydrate surface, an experimental setup was constructed. Its scheme and general view are shown in Fig. 1. To simulate a large array of GH, it is necessary to avoid significant heat loss to the environment in all directions except one. Therefore, GH was inside the Dewar vessel, which was placed in 2 layers of thermal insulation (mineral wool 2 cm thick, and polystyrene foam 4 cm thick). For temperature measurement, ds18b20 electronic sensors with a measurement accuracy of  $\pm 0.1^\circ\text{C}$  and a control thermometer with a division value of  $0.1^\circ\text{C}$  were used.

Propane hydrate was loaded into the Dewar flask, electronic temperature sensors and a mercury thermometer were placed. To reduce the effect of external air circulation, the Dewar flask was covered with a polystyrene foam lid. Periodically, the position of the sensors relative to the hydrate was changed by rotation around a vertical axis.

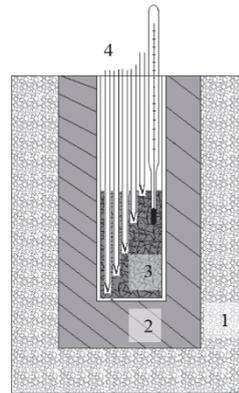
As a result of the experimental studies analysis, it was found that due to the hydrate structure heterogeneity, its temperature in different parts of the surface can differ significantly. The results of the studies show that the temperature on the surface of the “dry” propane hydrate is in the range  $0.3\text{--}3.0^\circ\text{C}$ . Its average value is  $+1.1^\circ\text{C}$ . After partial dissociation, an ice crust forms on the hydrate surface and its melting temperature becomes equal to  $0^\circ\text{C}$ . To reduce the effect of hydrate heterogeneity in measuring average temperatures on the hydrate surface and in its depth, in the next series of experiments, sensors with a small surface area and high thermal conductivity, in particular thermocouples, were used. Unlike electronic sensors, thermocouples are not “afraid” of moisture and, after calibration, can measure temperature with an accuracy of  $\pm 0.05^\circ\text{C}$ . For measuring thermocouples EMF, a F136 microvoltammeter was used.

The aim of the second series of studies was to clarify the temperature distribution in the inner layers and on the surface of a large mass of propane hydrate. Thus, all temperature sensors (thermocouples) were located inside the GH

at different depths and only one was on its surface. For research, “dry” GH was used with a gas capacity of 40 ml of gas / ml of water. The scheme and appearance of the research facility are shown in Fig. 2.

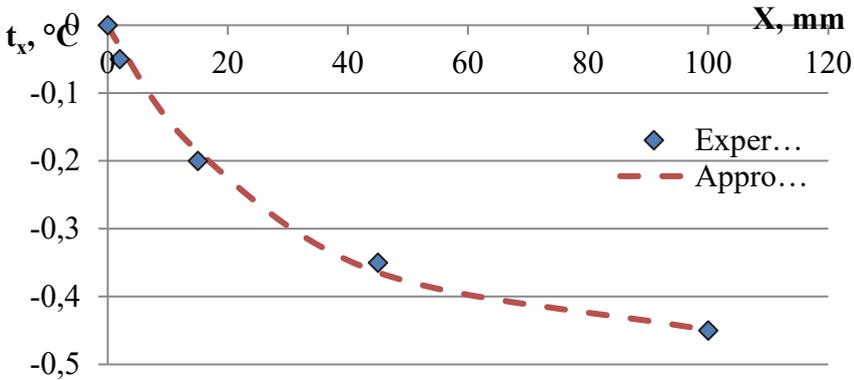


**Fig. 1.** Scheme of the research facility: 1 – Dewar vessel; 2 – layer of mineral wool; 3 – thermal insulation layer of expanded polystyrene; 4 – gas hydrate; 5 – electronic temperature sensors



**Fig. 2.** Scheme of the pilot plant: 1 – a layer of expanded polystyrene; 2 – layer of mineral wool; 3 – gas hydrate inside the Dewar vessel; 4 – thermocouples

**Research progress.** Thermocouples were placed at different heights in the Dewar vessel, Fig. 2. After that, the propane hydrate was poured and rammed. The measured temperatures were recorded in a table. The results of temperature measurements are shown in Fig. 3. An analysis of the data obtained indicates that, at hydrate low gas capacity, the temperature curve always starts at 0°C (ice), and ends with the hydrate stable storage temperature, Fig. 3. The obtained results can be explained with the following physical explanation: on the hydrate surface there is ice that melts at a temperature of 0°C. Due to the elevated temperature, hydrate layers that come into contact with the surface dissociate faster. Gas is released through slots and cools hydrate inner layers. At a certain depth, the hydrate temperature stabilizes and the process of dissociation slows down.



**Fig. 3.** Low-gas hydrate temperature regime

The obtained experimental data show that the temperature distribution in the surface layer of propane hydrate is well approximated by the dependence:

$$t_x = -0,47 + (0 - (-0,47)) \ell^{-35x}, \quad (1)$$

where:  $-0,47$  – stable temperature of the deep layers, °C;  $0$  – temperature on the surface of the array, °C;  $35$  – approximation coefficient,  $m^{-1}$ ;  $x$  is the distance from the array, m.

A number of experiments were conducted with hydrate, which has a high gas content (57 ml of gas / ml of water). The analysis of the obtained data indicates that the top layer of the “dry” not covered with ice crust, propane hydrate is in the range of positive temperatures  $0,5-0,8^\circ\text{C}$ . The deepest layer is in conditions close to thermodynamic stability at a temperature of  $-0,9^\circ\text{C}$ . There is a transition region approximately 50 mm thick, where a significant temperature gradient is observed, Fig. 4. The temperature distribution in the surface layer of propane hydrate is well approximated by the dependence:

$$t_x = -0,9 + (0,7 - (-0,9)) \ell^{-50x}, \quad (2)$$

where:  $-0,9$  – stable temperature of the deep layers, °C;  $+0,7$  – temperature on the array surface, °C;  $50$  – approximation coefficient,  $m^{-1}$ .

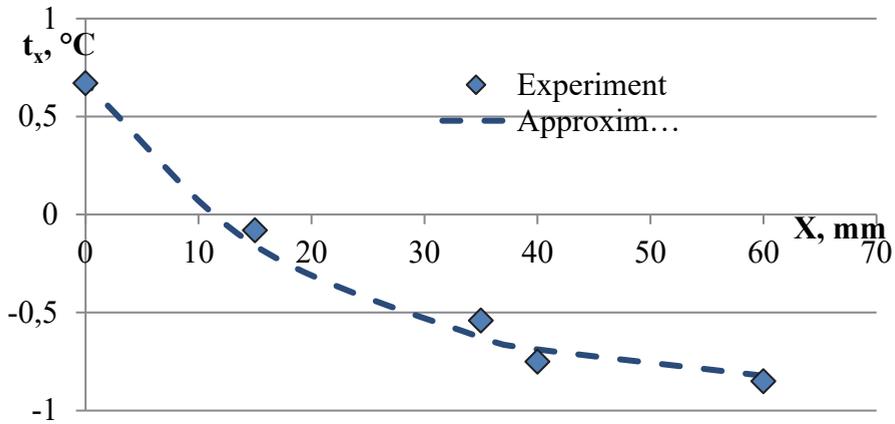


Fig. 4. Temperature distribution at the hydrate surface

Thus, the results of experimental studies show that the temperature distribution in the depth of the slowly dissociating GH is well approximated by the dependence:

$$t_x = t_\infty + (t_0 - t_\infty) e^{-kx}, \quad (3)$$

where:  $t_\infty$  – the temperature of the thermodynamically stable layer of the hydrate, °C;  $t_0$  – temperature on the hydrate surface, °C;  $k$  – is the approximation coefficient,  $m^{-1}$ . The coefficient  $k$  reflects a change in the intensity of internal heat sources in the depth of GH.

## 5. The mathematical model of quasi-stationary gas hydrates dissociation

In a dissociating hydrate, the process of phase transition and heat removal with gas heating both occur simultaneously. Most of the heat flow, which is supplied to the inner layer by heat conduction, is absorbed by dissociating hydrate, and a smaller part is removed by heating the gas, which moves from the cold internal (deep) layers of the hydrate to the outside. The system of equations describing these processes is a mathematical model of the process of gas hydrates dissociation. For the most accurate reproduction of thermophysical processes in mathematical modeling, it should be focused on the experimental data as for the temperature distribution inside a dissociating gas hydrate.

It is supposed that there is a sufficiently GH large array so that the temperature field in it can be considered one-dimensional. Heat is supplied to the surface of this hydrate by heat exchange. To calculate the hydrate storage

conditions, the dissociation process can be considered long enough to establish a stationary temperature distribution in the hydrate mass.

The mathematical model of the hydrate dissociation thermal regime is based on the nonlinear Fourier heat equation with heat specific volume sources (or sinks). Considering the conditions for conducting experimental studies, it can be assumed that the temperature field varies along only one coordinate axis. The one-dimensional Fourier heat equation with volumetric heat sources (Lysak 2010) has the form:

$$\frac{\partial t_g}{\partial \tau} = \frac{\lambda_g}{c_g \rho_g} \frac{\partial^2 t_g}{\partial x^2} + \frac{q_{V(x)}}{c_g \rho_g}, \quad (4)$$

where:  $t_g$  – temperature, °C;  $\lambda_g$  – thermal conductivity of hydrate, W/(m·°C);  $c_g$  – its heat capacity, J/(kg·°C);  $\rho_g$  – hydrate density, kg/m<sup>3</sup>;  $q_v$  – volumetric heat sources, W/m<sup>3</sup>.

At the boundary of the hydrate mass, the boundary conditions of the third kind are valid:

$$-\lambda_g \frac{dt_{g(0)}}{dx} = \alpha_3 (t_3 - t_{g(0)}), \quad (5)$$

where:  $\alpha_3$  – GH surface heat transfer coefficient, W/(m<sup>2</sup>·°C);  $t_3$  – ambient air temperature, °C;  $t_{g(0)}$  – GH surface temperature, °C.

Since the problem is stationary, the partial derivative of temperature with respect to time is zero, and the partial derivative of temperature with respect to spatial coordinate can be replaced by the full derivative. Thus, the capacity of volumetric heat sinks can be defined as a function of coordinate, W/m<sup>3</sup>:

$$q_{V(x)} = -\lambda_g \frac{d^2 t_g}{dx^2}. \quad (6)$$

Substituting the second derivative of equation (3) into (6), it is obtained the distribution of the volume runoff of heat in hydrate, W/m<sup>3</sup>.

$$q_{V(x)} = -\lambda_g k^2 (t_0 - t_\infty) \ell^{-kx}. \quad (7)$$

For experimental conditions: thermal conductivity of propane hydrate  $\lambda_g = 0.5$  (W/(m·°C)), surface temperature of hydrate  $t_0 = 0.7$ °C, stable temperature of deep layers of hydrate  $t_\infty = -0.9$ °C. Therefore, after substitution of the original data in equation (7), it is obtained the values of the volumetric heat sinks, W/m<sup>3</sup>:

$$q_{V(x)} = -2000 \ell^{-50x}. \quad (8)$$

Equation (8) shows that heat sinks with a power of  $2000 \text{ W/m}^3$  act on the surface of a dissociating propane hydrate. In the deep layers, their power decreases rapidly and at a depth of  $50 \text{ mm}$  it is only  $164 \text{ W/m}^3$ .

The temperature at the surface of the dissociating GH ( $t_0$ ) plays a key role, since it determines how dissociation occurs. At  $t_0 < 0^\circ\text{C}$  – hydrate recrystallization in ice occurs and the effect of self-preservation is observed, at  $t_0 > 0^\circ\text{C}$  – the hydrate immediately dissociates into gas and water. The surface temperature of the dissociating GH is the result of the heat balance of the surface with the environment and can be found from the boundary conditions (5), where the first derivative from equation (3) with respect to  $x$ :

$$\frac{dt_0}{dx} = -k(t_0 - t_\infty) e^{-kx}. \quad (9)$$

Substitution of the obtained expression (9) into equation (5) enables to find the temperature on the hydrate surface:

$$t_0 = \frac{\lambda k t_\infty + \alpha_3 t_3}{\lambda k + \alpha_3}. \quad (10)$$

During the experiment, the ambient temperature was  $t_3 = +11^\circ\text{C}$ . For a top-down heat flow in still air, the coefficient of heat exchange near the surface is  $\alpha_3 = 4 \text{ W/(m}^2\text{C)}$ . Substituting the experimental data, the temperature value on the surface of the dissociating hydrate can be obtained,  $^\circ\text{C}$ :

$$t_0 = \frac{0,5 \cdot 50 \cdot (-0,9) + 4 \cdot (+11)}{0,5 \cdot 50 + 4} = 0,74.$$

Since the temperature on the hydrate surface is above  $0^\circ\text{C}$ , the effect of self-preservation is not observed, and the hydrate dissociates into gas and water.

## 6. Hydrate self-preservation

Another important parameter that determines the heat transfer processes in the gas hydrate array is the coefficient  $k$ . It reflects the intensity of change in internal heat sinks in the hydrate. It is checked at which  $k$  the effect of self-preservation on the open surface in the room by the formula (10). It is observed:

$$k = \frac{\alpha_3(t_3 - t_0)}{\lambda(t_0 - t_\infty)}. \quad (11)$$

Substitute the value for propane hydrate at  $t_0 = 0^\circ\text{C}$ , room temperature  $+18^\circ\text{C}$ , heat transfer coefficient near the vertical surface  $\alpha_3 = 8.7 \text{ W}/(\text{m}^\circ\text{C})$ , it is obtained:

$$k = \frac{8,7(18-0)}{0,5(0-(-2,5))} = 125.$$

Thus, theoretically, the effect of self-preservation for propane hydrate in "room conditions" can be expected at values of 125. However, the experimentally determined maximum value of  $k$  for does not exceed 50.

Similarly, the  $k$  value for methane hydrate can be calculated. The temperature on the ice surface is  $-2^\circ\text{C}$ , and the equilibrium temperature inside the methane hydrate mass at atmospheric pressure is  $-33^\circ\text{C}$ . Substituting these values for the methane hydrate of formula (11) gives  $\text{m}^{-1}$

$$k = \frac{8,7(18-(-2))}{0,5((-2)-(-33))} = 11,2$$

Thus, for methane hydrate, self-preservation can be observed already at values  $k > 11.2$ . The intensity of internal heat sinks near the surface of GH,  $\text{W}/\text{m}^3$

$$q_{V(0)} = -0,5 \cdot 11,2^2 (-2 - (-33)) = 1944.$$

Similarly, this factor can be determined for other hydrating gases. The calculation results for the various gases are summarized in Table 1.

**Table 1.** Characteristics of hydrates under conditions of self-preservation

Gas - hydrating agent	$t_0, ^\circ\text{C}$	$t_\infty, ^\circ\text{C}$	$k, \text{W}$	$q_{V(0)}, \text{W}/\text{m}^3$
Nitrogen	-1	-49	6,9	1142
Methan	-2	-33	11,2	1944
Carbon Dioxide	-2	-22	17,4	3027
Ethan	-1	-13	27,6	4570
Propan (for self-presevation)	0	-2,5	125	19531
Propan (by experiment)	0,7	-0,9	50	2000

Analyzing the results, it can be noted that in propane hydrate, the intensity of volumetric heat sinks is about ten times lower than it is required to obtain the self-preservation effect. Thus, at relatively high ambient temperatures, the self-preservation of propane hydrate is usually not observed.

From the formula (11) it is possible to obtain a dependence for determining the temperature of the outside air below which the effect of self-preservation can be obtained, °C:

$$t_{3c} = t_0 + \frac{k \cdot \lambda (t_0 - t_\infty)}{\alpha_3}. \quad (12)$$

For example, for propane GH, a self-preservation phenomenon can be expected if the ambient temperature drops below °C.

$$t_{3c} = \frac{50 \cdot 0,5(0 - (-2,5))}{8,7} = 7,1.$$

Thus, formula (12) is of great practical importance because it enables to calculate the conditions under which long-term storage of hydrates in a nonequilibrium state can be achieved.

In some scientific papers, the authors note the complexity, and even the inability to obtain the self-preservation effect for small pieces of hydrate. The physical explanation of this phenomenon has not been discovered by the authors. The results of our studies show that the transition temperature range is 50-100 mm deep in the GH array. Thus, the lack of self-preservation effect for small pieces of gas hydrate is explained by the inability to obtain the required temperature decrease in their depth.

A number of scientific papers by different authors (Tarko 2012) note the key role of the ice crust on the hydrate surface in obtaining the effect of self-preservation. To analyze the effect of the thermal resistance of the ice crust on the hydrate self-preservation, it is determine the effect that it can have on the heat exchange process on the GH surface. If the effect of self-preservation (as for methane) is observed, then the formed layer of ice on the surface can be considered as an additional resistance to heat transfer. The thickness of the ice crust on the surface of GH methane in the mode of self-preservation is approximately 0.3 mm. The calculation results show that such a thickness of the ice crust increases the thermal resistance by only 0.1% and is not able to significantly affect the thermal regime of GH. In such a case, the principle of ice crust application can only be to partially seal the GH and reduce the area of the heat exchange surface.

## 7. Conclusion

For the investigation of thermal modes of gas hydrate dissociation, an experimental setup has been developed, where a number of experimental studies with propane hydrate have been performed. Approximation dependences for determining the surface temperature and the depth of the dissociating gas hydrate

are obtained. Thus, it has been established that the temperature distribution in the depth of the hydrate is exponential. The results of the experimental studies show that hydrate dissociation occurs within a certain temperature range. Increasing the temperature leads to an intensification of this process.

The one-dimensional mathematical model of thermal processes occurring during the dissociation of a hydrate array is developed. It considers the functional dependence of heat sinks on the depth in the hydrate and enables to determine the temperature on the surface and in the depth of the gas hydrate array. The results of the studies show that the effect of the hydrate self-preservation is due to the decrease in the temperature of its deep layers due to partial dissociation. The freezing of supercooled water and the formation of an ice crust on the surface of the dissociating GH massif are only a consequence of this effect. The reason for the lack of self-preservation effect in small pieces of gas hydrate is established.

The practical significance of the research results is to determine the conditions under which the phenomenon of hydrate self-preservation is observed. The prospect of further scientific development in this direction is the study of the thermal insulation effect on the slowdown of GH dissociation, which is stored under nonequilibrium conditions.

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## Abstract

To improve gas hydrates dissociation technology, studies of heat transfer processes on the interfacial surface are significant. In the work, experimental and theoretical studies of the gas hydrates dissociation are presented.

The scientific novelty is in establishing quantitative characteristics that describe the gas hydrates thermophysical parameters thermophysical characteristics influence on the heat transfer processes intensity on the interphase surface under conditions of gas

hydrates dissociation. Based on the results of experimental studies, approximation dependences for determining the temperature in the depths of a dissociating gas hydrate array have been obtained. Gas hydrates dissociation mathematical model is presented.

The practical significance of the research results is in determining quantitative indicators of the heat transfer processes intensity under the conditions of propane hydrate dissociation. The results of the work can be applied to designing equipment for gas hydrates storage and dissociation.

**Keywords:**

gas hydrates, self-preservation effect, experimental studies, mathematical modeling, dissociation

**Termofizyczny efekt samozachowawczy hydratów gazowych****Streszczenie**

Badania procesów wymiany ciepła na powierzchni międzyfazowej mają ogromne znaczenie dla poprawy technologii dysocjacji hydratów gazowych. W pracy przedstawiono eksperymentalne i teoretyczne badania dysocjacji hydratów gazu.

Nowość polega na ustaleniu cech ilościowych, które opisują wpływ właściwości termofizycznych hydratów gazu na intensywność procesów wymiany ciepła na powierzchni międzyfazowej w warunkach dysocjacji hydratów gazu. Na podstawie wyników badań eksperymentalnych uzyskuje się zależności aproksymacyjne do określania temperatury na głębokościach dysocjującego układu hydratów gazu. Przedstawiono matematyczny model dysocjacji hydratów gazu.

Praktyczne znaczenie wyników badań polega na określeniu ilościowych wskaźników intensywności procesów wymiany ciepła w warunkach dysocjacji hydratu propanu. Wyniki można zastosować w projektowaniu urządzeń do magazynowania i dysocjacji hydratów gazu.

**Słowa kluczowe:**

hydraty gazu, efekt samozachowawczy, badania eksperymentalne, modelowanie matematyczne, dysocjacja