E.O. Maksymova, Ph.D. (Geol.-Miner.)
P.B. Saik, Ph.D. (Tech.)
V.H. Lozynskyi, Ph.D. (Tech.)
Dnipro University of Technology

On the issue of creating a laboratory setup to study the gas hydrate dissociation processes

This paper deals with the problems emerging during the research into gas hydrate production processes, in particular, their dissociation in natural conditions. The advantages and disadvantages of known research methods have been analysed based on domestic and world research-and-practice experience of the conditions for hydrate formation and dissociation. Engineering tasks set for the development of a laboratory setup for gas hydrates dissociation have been performed using the analysis of analytical, experimental studies presented in scientific papers, publications, research projects and patents.

It has been determined that the «gas hydrate reservoir-massif» thermodynamic system is multicomponent and complex. Therefore, to transfer the research results from a model to full-scale object, it is necessary to use appropriate coefficients that take into account changes in the geometric parameters and physical properties of the modelled object. They are set on the basis of the principles of static, kinematic and dynamic similarities. It is convenient and economically feasible to conduct experimental research on analogue test models, as it makes possible to determine the high convergence of results.

An algorithm is proposed for conducting research on gas hydrates decomposition, which includes the stages of constructing a model of gas hydrate deposit depending on its lithological composition. In perspective, it is planned, when performing a series of theoretical and experimental research, to study the influence of variable indicators of a gas hydrate deposit lithological composition (clayed silt, sand-silty clay, sand, methane hydrate) on methane gas output depending on the heat-transfer medium temperature.

Keywords: gas hydrate; experimental setup; heat exchange; model; similarity coefficient.

Setting of a problem. In modern conditions of energy consumption intensity and the rapid development of research-engineering progress, it is obvious that mankind is approaching the depletion of traditional natural gas reserves. That is why, the study of alternative fuels, the search for additional sources of traditional and non-traditional energy resources is still the actual applied research task. The introduction of radical technologies for the coal mines reserves development [1–3] and technologies for the gas hydrate deposits development [4–6] are the promising methods for obtaining a substitute for natural gas. The development of coal mines reserves is based on the formation of energy-chemical complexes and an underground coal gasification base [7–9]. An important stage in the formation of these complexes is the synergistic approach to coal gasification technology and methane degassing in coal mines [10, 11]. At the same time, the problems with the introduction of technological solutions for the gas hydrates development in the offshore area of Ukraine have not been studied thoroughly and still require improvement. First of all, it is necessary to experimentally study the conditions for gas hydrates dissociation depending on the depth, lithological variety of the gas hydrate deposit and its physical-and-mechanical properties.

Thus, improving the technology for gas hydrate deposits development and its implementation is a significant leap in the fuel and energy sector development in industrialized countries. As for Ukraine, this is an opportunity to refuse importing natural gas.

Review of recent research and papers. Over the past decade, the research related to the formation and production of gas hydrates has become widespread. Such famous scientists as Yu.F. Makogon, K.Kvenvolden, S.Dallimore, Y.Okuda, E.F. Shnyukov, A.P. Ziborov, V.P. Kobolev, E.A. Bondarev have revealed the occurrence of gas hydrates in the Earth's mainland area, as well as their wide distribution in the shelf bowels and the World Ocean deposits. They have determined the conditions of their formation, the problems of genesis, geological and structural position, lithology, geomorphology and tectonics [12–16].

The authors [17, 18] argue that gas hydrates reserves ($2{\cdot}10^{14} - 2{\cdot}10^{16}$ m^3) are commensurate with the amount of oxygen in the Earth's atmosphere ($8{\cdot}10^{17} - 7{,}6{\cdot}10^{18}$ m^3). Taking into account the high specific concentration of gas in natural hydrates (up to 160 m^3 of gas in 1 m^3 of gas hydrate), their relatively shallow occurrence (under the seabed, starting from water depths of 300-500 m) [15], bottom gas hydrates are considered by the world scientific community as a real alternative to gas produced by traditional methods. At present, all industrialized countries, such as Great Britain, Germany, Canada, China, USA, Norway and Japan are involved in the

development of gas hydrate production technologies. Until recently, Japanese and Canadian researchers have had a successful experience in developing gas hydrate deposits. In Canada, gas has been obtained from the gas hydrate field of the permafrost zone. Canadian developers managed to maintain a stable gas output only for six days. This has been ascertained by the scientific community as a real breakthrough in the field of "blue fuel" production, since Canada in its Arctic part has gas hydrate reserves sufficient to meet the needs of its domestic market for several centuries.

In 2013, Japan began to work on one of the largest and priority national programs in the world with huge budget financing, aimed at developing offshore gas hydrate deposits in the Nankai spur in the Pacific coastal strip of the Japanese island of Honshu from a depth of 950 m [13]. For the first time in the history of mankind, Japanese gas enterprises have managed to extract gas out of gas hydrates from the ocean bottom.

The history of the discovery of natural gas hydrates and modern studies on the existence and identification of the natural gas hydrates are comprehensively covered in the work [19]. This work has been performed in the framework of cooperation between three higher educational institutions specializing in the extraction and processing of energy resources, namely: National TU Dnipro Polytechnic, Ivano-Frankivsk National Technical University of Oil and Gas, and National University «Yuri Kondratyuk Poltava Polytechnic».

According to the research analysis results, it has been revealed that most of the studies are devoted to the conditions for gas hydrates formation by changing the thermodynamic and thermobaric conditions for their obtaining [20, 21], methods of hydrate formation processes acceleration [22, 23] and developing the methods for their extraction and transportation, based on the developed mathematical models [24, 25]. This approach does not allow to study the conditions for gas hydrates dissociation in laboratory facilities due to the lack of a laboratory base.

Thus, the authors set an **objective** to formulate the basic principles that will be a foundation to the development of the design implementation of a setup for studying the gas hydrate deposits dissociation processes with the possibility of reproducing their geological conditions of occurrence on full-scale ones.

Methodology of research. The development of the design implementation of a setup for research into gas hydrate dissociation process is based on the analysis of engineering and research-and-practice solutions when studying the hydrate formation and dissociation parameters, which are reflected in scientific papers and publications, research projects, patents and the like. The main task of the authors of the work, when developing the setup, was to ensure the similarity of the «reservoir-strip» thermodynamic system, given the reliability of obtaining experimental results relative to the full-scale object.

Statement of basic materials. When developing an experimental setup for studying gas hydrate dissociation, a crucial factor is to ensure the borderline conditions with respect to the full-scale ones. The design implementation of the dissociation setup is based on the characteristics of its thermal regime change over time. For specified conditions, the most rational is the calculated thermodynamic system «gas hydrate reservoir-massif» with the temperature distribution boundaries. In this case, provided that the temperatures are parallel, the thermal regime takes a quantitative characteristic. This makes possible to solve one-dimensional variants of heat exchange and heat transfer processes in mine rocks, ground and surface waters, which is described by the second order differential equations with partial derivatives of an elliptic type [26]:

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = 0 \, ;$$

parabolical

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = \frac{1}{\alpha} \cdot \frac{\partial T}{\partial t} \, ;$$

and hyperbolical types

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = \frac{1}{\alpha} \cdot \frac{\partial^2 T}{\partial t^2} \, ,$$

where T – temperature, °C, α – thermal diffusivity (characterizes the thermal anisotropy of the simulated massif), m^2/s, x, y, z – spatial coordinates, m, t – time, s.

When setting such a thermodynamic system, in the process of dissociation, the phase equilibrium will displace in the gas hydrate deposit, which is characterized by the processes of the deposit decomposition into gas and water with a constant gas movement towards its unloading.

To confirm the results obtained, according to analytical studies of both domestic and foreign scientists, the authors of the work have developed a technological scheme of a complex for studying the hydrate formation parameters and gas hydrates dissociation. The scientific-practical research into the methane hydrates formation in the laboratory of gas hydrate technologies at Dnipro University of Technology are the basis for the construction of this scheme [27]. The technological scheme with layout of the main nodes of the complex for research into parameters of hydrate formation and dissociation is shown in Figure 1.
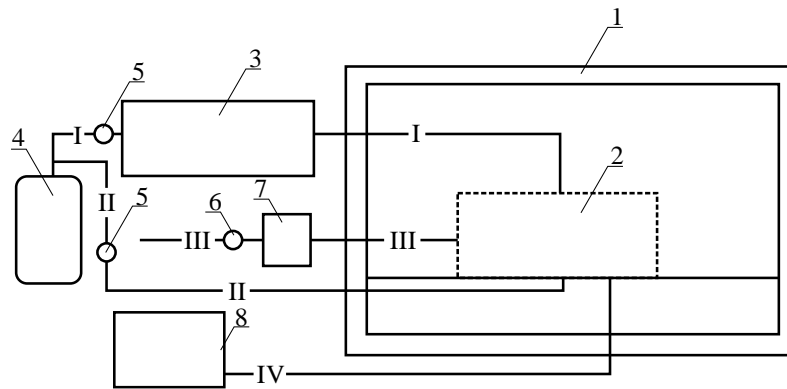
Fig. 1. Technological scheme of the complex for research into the hydrate formation parameters and gas hydrates dissociation: 1 – climatic temperature chamber, 2 – modulus of hydrate formation or dissociation, 3 – high pressure compressor, 4 – gas cylinder, 5 – gas-flow rate meters, 6 – water counter, 7 – high pressure pump, 8 – gas storage tank, I – high pressure gas supply pipeline (8–10 MPa), II – gas supply pipeline at bubbling, III – water supply pipeline (6–8 MPa), IV – pipeline for discharging the gas obtained in gas hydrates dissociation

A constituent of the complex is a gas hydrate setup, the main block of which is the climatic chamber (1) with a replaceable module (2). One module is designed to study the hydrate formation processes, the other one – for dissociation.

The main module difference, proposed by the authors [27], is the introduction of additional pipelines (II, IV). Pipeline II is designed for additional intensification of the hydrate formation process by way of bubbling, which provides faster gas molecules ingress into water molecules according to the "guest-host" mechanism [28]. Pipeline IV is designed for obtained gas withdrawal as a result of dissociation process. Therefore, to research into the parameters of the gas deposit dissociation with account of changing lithological variety of the gas hydrate deposit, the authors have developed the second module of the gas hydrate setup. The module is developed on the basis of technological scheme for a gas hydrate deposit extraction with the use of injection and draining wells [29]. Figure 2 shows a laboratory setup for studying the processes of gas hydrates dissociation.
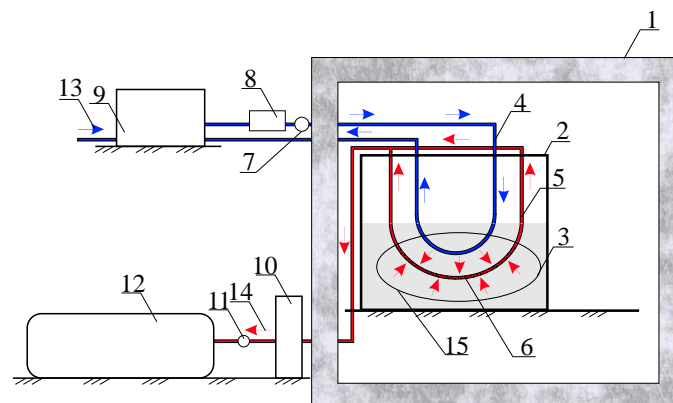

Fig. 2. Technological scheme of a laboratory setup for research into the gas hydrates dissociation parameters: 1 – climatic chamber, 2 – modulus of dissociation, 3 – gas hydrate deposit which is formed, 4 – polypropylene water supply pipeline, 5 – gas-outlet polypropylene pipeline, 6 – perforation hole of the gas-outlet polypropylene pipeline, 7 – water counters, 8 – water heater with an automated temperature control system, 9 – circulation pump, 10 – expansion tank, 11 – gas meter, 12 – gas storage tank, 13 – water movement direction, 14 – movement direction of gas, obtained by gas hydrate dissociation, 15 – dissociation zone

The reliability of the results obtained when conducting research on the gas hydrates dissociation should be ensured by similarity criteria under which the following conditions are met:

-   the natural materials in their physical properties correspond to the full-scale ones;
-   the geometrical parameters of the dissociation site are reproduced in the model, ensuring the principles of static similarity;
-   stratum lithological texture takes into account its change in area;

- hierarchical dependence of factors forming a stress-strain state of the rock massif;
- dissociation technological effectiveness is ensured by balancing the chemical reactions behavior and the physical velocities of the process.

To study dissociation processes, an important stage is the formation of a gas hydrate square pillar with a lithological variety (the presence of clayed silt, sand-silty clay and sand). The pillars are formed in the climatic chamber at a temperature of +1,0°C (Figs. 2, 1), which makes impossible their decomposition. The most rational is to conduct a series of experiments on 12 different physical models (Table 1).

Table 1

Parameters of the gas hydrate deposit models formation during the experimental studies

Model No. (pillar)	Lithological variety, %			
	Clayed silt	Sand-silty clay	Sand	Methane hydrate
1	20	10	–	70
2		25	5	50
3		45		30
4		65		10

To simulate the gas hydrates dissociation process in real conditions, it is necessary to set on a laboratory setup the scale factors and similarity conditions based on the general provisions of the mechanical similarity theory [30]:
- geometric similarity of full-scale objects and model;
- physical constants proportionality of full-scale object and model;
- identity of the initial system state of the full-scale object and model;
- compliance with the system borderline conditions.

The process of modeling a gas hydrate deposit provides for the determination of the necessary scale factor ($\overline{x_i}$), at which these parameters (gas hydrate deposit thickness, diameter of injection and draining well) are in relation of simple similarity and found by the following ratio [46]:

$$\overline{x_i} = k_i x_i, \quad i = 1, 2, ..., N,$$ (1)

where $\overline{x_i}$ – characteristic of the first parameter; x_i – characteristic of the second parameter, respectively; k_i – empirical coefficient.

Based on the function (1) conversion, any two parameters of the same nature can be put in the similarity relation. The required scale factor has been chosen with account of the ratio between the thicknesses of hydrate deposit, developed in-situ and in the model:

$$x_m = \frac{m_{n.h.}}{m_{m.h.}},$$

where $m_{n.h.}$ – the thickness of hydrate deposit in-situ, m; $m_{m.h.}$ – the thickness of hydrate deposit in model, m.

The kinematic similarity, that is, the similarity between the velocities in-situ and in the model is expressed by the correspondence of directions and the proportionality of all velocities:

$$\frac{v_{n.}}{v_{m.}} = x_v = const,$$

where $v_{n.}$, $v_{m.}$ – velocity of the dissociation zone propagation of full-scale object and model, m³/day.

Dynamic similarity provides for the fulfillment of the condition according to which at any point of the model, the forces act which are similar in nature and direction to the forces at the corresponding point [31].

A change in the lithological variety in the gas hydrate physical model is set on the basis of typical formations of gas hydrate deposits in natural conditions [32]. The process of creating models involves the primary laying the polypropylene pipes down in them to simulate injection and draining wells with a diameter of 16 mm and a wall thickness of 2 mm. In the lower part of the polypropylene gas-outlet pipe (see Fig. 2), the holes with a diameter of 6 mm are drilled symmetrically on both sides. This ensures efficient draining out of gas during the gas hydrates decomposition process. The formed gas hydrate pillars are placed in a square module of organic glass with dimensions of 0,4×0,4×0,3 m. The dissociation module (2) itself is being placed in a climatic chamber (1). The temperature in the chamber should be set at the level of + 9,0°C, which corresponds to the natural temperature of gas hydrates in the Black Sea offshore area. The movement of water is provided using a circulation pump (9), and the temperature is maintained by a water heater with an automated temperature control system. When the hydrate is decomposed, the obtained gas through the drilled perforation holes (6) and through

the gas-outlet polypropylene pipeline (5) enters into the expansion tank, after which it enters into the storage tank (12) through the gas meter (11).

Conclusions. The intensive rate of indicators increase in energy consumption is encouraging the search for alternative energy sources. Therefore, the gas hydrate deposits development is one of the advanced technologies that will allow to obtain high-calorific gas – methane. At the same time, an experimental testing under laboratory conditions is an integral part of the innovative technology for developing gas hydrates. This will allow to assess the main advantages and disadvantages of methods for disclosing, preparing and exploiting a gas hydrate deposit.

The proposed laboratory setup designed for research into the gas hydrates dissociation processes makes possible to investigate and correct theoretical studies relative to the gas hydrates extraction processes. The recommendations should be developed and the parameters should be substantiated for the gas hydrates development providing practical advice on their development in natural conditions.

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Максимова Елла Олександрівна – кандидат геолого-мінералогічних наук, доцент кафедри будівництва, геотехніки і геомеханіки Національного технічного університету «Дніпровська політехніка».

Наукові інтереси:
– інженерний захист територій;
– газогідратні технології.
http://orcid.org/0000-0003-1206-6181.
E-mail: elmaks28@gmail.com.

Саїк Павло Богданович – кандидат технічних наук, доцент кафедри гірничої інженерії та освіти Національного технічного університету «Дніпровська політехніка».

Наукові інтереси:
– геотехнології гірництва;
– підземна газифікація вугілля.
http://orcid.org/0000-0001-7758-1083.
E-mail: saik.nmu@gmail.com.

Лозинський Василь Григорович – кандидат технічних наук, доцент кафедри гірничої інженерії та освіти Національного технічного університету «Дніпровська політехніка».

Наукові інтереси:
– геотехнології гірництва;
– підземна газифікація вугілля.
http://orcid.org/0000-0002-9657-0635.
E-mail: lvg.nmu@gmail.com.