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UDC 532.529
DOI: 10.15587/2312-8372.2018.141302

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INFLUENCE OF EXTERNAL FACTORS ON THE PROCESS OF HYDRATES DEVELOPMENT IN LABORATORY CONDITIONS

Об'єктом досліджень є вплив різноманітних факторів на процес синтезу гідрату пропану у лабораторних умовах. Відомо, що синтезований газовий гідрат може містити значну кількість льоду, яка знижує його газовміст. На якість гідрату впливають: тиск газу, температура води, час проведення досліду та концентрація поверхнево-активних речовин.

Для дослідження комплексного впливу цих факторів на якість отриманого гідрату була розроблена експериментальна установка. Після синтезу газового гідрату його газовміст визначався за допомогою спеціально розробленого стенду. В ході досліджень використовувалися різноманітні вимірювальні прилади: манометри, термометри, мірний посуд, електронні ваги та ін., які дозволили отримати достовірну інформацію про теплофізичні характеристики процесу синтезу та дисоціації газового гідрату.

В результаті виконання багатофакторного експерименту отримано масив даних для аналізу методами математичної статистики. Визначено коефіцієнти кореляції і встановлено, що домінуючими факторами є тиск газу і концентрація поверхнево-активних речовин. Температура води повинна бути в межах робочого діапазону 1–5 °С. Час утворення гідрату у барботажному режимі у межах 0,5–5 год теж не здійснює істотного впливу на якість отриманого гідрату. Для усіх факторів побудовано регресійні залежності та графіки. Встановлено, що для стандартних регресійних залежностей (лінійна, експоненційна, логарифмічна та поліноміальна) коефіцієнти множинної кореляції знаходяться в межах 0,19–0,46. Це означає, що стандартні регресійні залежності не дозволяють урахувати усі особливості отриманих результатів. Тому підбір оптимальної залежності виконано методом варіації коефіцієнтів та типів функціональних залежностей і отримано апроксимаційну формулу для визначення прогнозованого газовмісту гідрату.

Результати досліджень показали, що завдяки комплексному урахуванню різних факторів можна визначити діапазон оптимальних значень тиску, температури та концентрації поверхнево-активних речовин, що дозволяє швидко отримувати гідрат високої якості.

Ключові слова: *газові гідрати, газовміст гідрату, зовнішні фактори, статистичний аналіз.*

1. Introduction

Gas hydrates are the only non-cryogenic source of natural gas on Earth, which can constitute a real competition to traditional deposits. Significant potential gas resources in hydrate deposits for a long time will provide humanity with high-quality energy raw materials.

The development of gas hydrate deposits requires the development of new technologies for exploration, production, transportation and storage of gas, more efficient than existing ones. With their help, traditional gas fields can be used, as well as those which development is now unprofitable. Extraction of gas from hydrate deposits can very quickly change the situation in the gas market, which can affect the export opportunities of Ukraine.

Due to the thermodynamic properties of gas hydrates, the possibility to realize the processes of their formation and decomposition for relatively low pressures and temperatures makes it possible to perform a number of technological processes with greater efficiency than existing

ISSN 2226-3780

industrial technologies. In this direction, it is promising to compress natural or other hydrate-forming gases, by transferring them to the hydrate state. Also an urgent issue is the study of the separation of gas mixtures in the process of hydrate formation, storage and transportation of gas in the gas hydrate state.

2. The object of research and its technological audit

The object of research is the influence of various factors on the process of hydrate synthesis. Usually hydrate synthesis occurs under conditions of intensive mixing of gas-hydrate former and water at the appropriate temperature and pressure. A necessary condition for the hydrate formation process is the constant removal of heat from the reaction region.

It is known that a synthesized gas hydrate can contain a significant amount of ice, reduces its gas content. The quality of the hydrate obtained is influenced by: gas pressure, water temperature, time of the experiment and concentration of surfactants. This work is devoted to a laboratory study of the effect of these parameters on the quality of propane hydrate.

3. The aim and objectives of research

The aim of research is establishment how the gas content of the hydrate depends on time, temperature, pressure and concentration of surfactants. To achieve this aim, it is necessary to perform the following tasks:

1. To develop a laboratory unit for studying the influence of various factors on the gas content of the obtained hydrate.

2. To conduct an experiment and obtain reliable data in a wide range of thermobaric conditions.

3. To perform statistical processing of the obtained data in order to reveal the quantitative relationships between the gas content of the hydrate and the factors of influence.

4. Research of existing solutions of the problem

Gas hydrates began to be considered in geological literature relatively recently. Most natural gas components (except for H_2, He, Ne, n-C_4H_{10} and heavy alkanes) are capable of forming individual hydrates. Water molecules form a polyhedric frame (polyhedral) in hydrates, with cavities in which gas molecules can be located. The equilibrium parameters of hydrates of different compositions differ, but for the formation of any hydrate at a higher temperature, a higher equilibrium concentration (pressure) of the hydrate-forming gas is required [1, 2].

Relatively low temperature at a sufficiently high hydrostatic pressure on the seabed, with water depths ranging from 300-400 m and more, predetermines the possibility of the existence of gas hydrates in the upper part of the bottom section. These conditions caused a special interest of geologists to submarine hydrates immediately after registration in the Soviet Union in 1969. The discovery of the properties of natural gases is in the earth's crust in a solid state and forms gas hydrate deposits [3, 4]. Excessive attention to submarine gas hydrates is due, first of all, to the fact that they are considered as a reserve of hydrocarbon raw materials. The study of the formation of carbon dioxide hydrate in a schistose medium was considered in [5]. The method of substitution of methane in hydrate for hydrogen sulphide was considered in [6]. Economic analysis of the application of gas hydrate technologies in comparison with traditional methods of gas supply was carried out in [7]. However, it should be noted that the approximating functions in which the hydrate formation processes are described are not given in the known papers. The struggle with hydrates by various methods is also investigated by many scientists, for example, scientists [8], but they remain either very energy-consuming, or environmentally harmful. It should be noted that scientists of the Poltava National Technical Yuri Kondratyuk University (Ukraine) also carried out theoretical studies in this direction [9, 10]. In particular, the hydrodynamic regime of a single gas bubble is considered in [9], which is under hydrate formation conditions. In [10], calculations of phase-reversal processes in the liquid surrounding the gas bubble were performed. However, the unresolved question remained the comparison of theoretical calculations with experimental data. In [11], for enclosing structures and in [12] for interstitial materials, separate experimental studies are presented, in which methods of mathematical statistics are widely used. These methods can be used to analyze the results of field experiments on the synthesis of gas hydrates.

Thus, the results of the analysis of literature data show that the mechanisms of complex influence of such factors as temperature, pressure, time, the presence of surfactants on the synthesis of gas hydrates are insufficiently studied.

Methods of research

The laboratory unit (Fig. 1) consists of a transparent bulb, which is used to observe the hydrate formation process. It allows to take photos and videos of formation and dissociation from any angle. A gas cylinder with a gas volume of 20 liters was used for the gas supply of the unit. To stabilize the gas pressure, a gas reducer and a pressure gauge with gas pressure measurement limits from 0 to 6 atm are used. To obtain the gas hydrate, the gas is fed to a transparent flask through the interstitial nozzle, which is located at the bottom of the bulb.

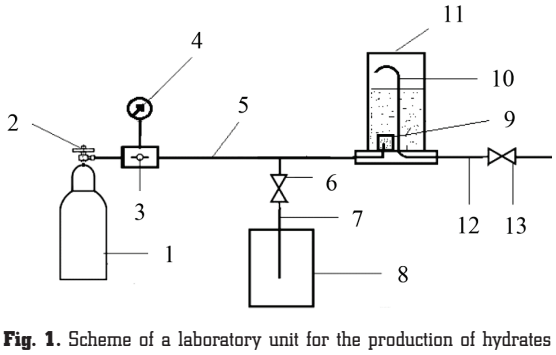

Fig. 1. Scheme of a laboratory unit for the production of hydrates in laboratory conditions: 1 – cylinder with gas; 2 – valve 3 – auxiliary gas reducer; 4 – gauge of excess pressure; 5 – gas hose; 6 – ball valve 7 – water drain hose; 8 – water tank; 9 – porous nozzle; 10 – copper tube; 11 – transparent bulb; 12 – gas outlet; 13 – valve for regulation and release of gas

The exhaust gas is removed from the top of the flask using a copper tube. To regulate the exhaust gas flow, a crane is provided.

To discharge water from the bulb a hose and a ball valve are used. The waste water is drained into a separate tank.

During the laboratory studies, parameters such as water temperature, pressure, surfactant concentration, time, gas volume were changed, and a relationship was established between these parameters.

After the hydrate was obtained, a comparative analysis of the amount of gas in it was carried out with the aid of an apparatus, a diagram and a general view of which are shown in Fig. 2, 3 respectively.

To determine the gas content of the hydrate, the following procedure was used. Let's fill the flask 9 and the container 11 with water, attach the gas hose 5 to it, and then fill the tube 3 with the suspended hydrate (m_{gg}), close it tightly with a cover 4. In the test tubes, attach the gas hose 5 and pour warm water 2 into the container 1 to accelerate the decomposition process hydrate. After a while, as a result of the dissociation of the hydrate, a gas begins to escape from the test tube, which enters the volumetric flask 9 through the gas hose, while displacing the water that is in the flask. Using the scale on the flask, determine the amount of gas (V_g). The gas content of the hydrate was determined by the formula, l/kg:

$$Q = \frac{V_g}{m_{gg}}. \quad (1)$$

The results are listed in Table 1.

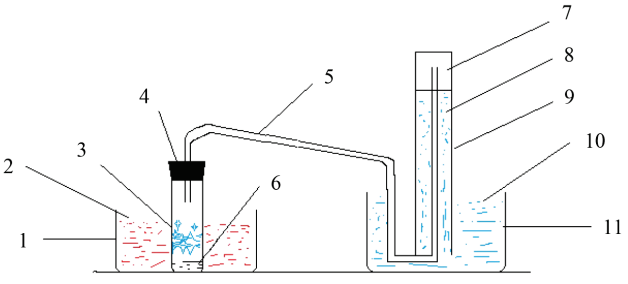

Fig. 2. Scheme of a laboratory unit for determining the amount of gas in the formed hydrate: 1 – container for water; 2 – warm water; 3 – test tube; 4 – cover; 5– gas hose; 6 – hydrate; 7 – volume of gas in the flask; 8 – amount of water in the flask; 9 – flask; 10 – water; 11 – water container

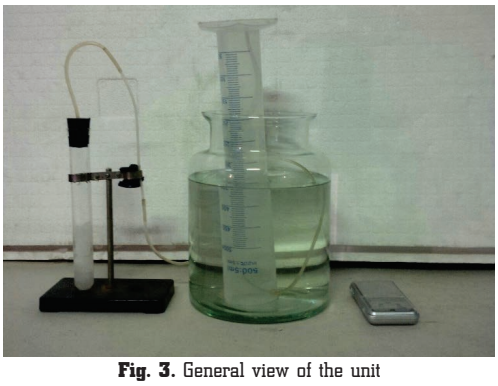

Fig. 3. General view of the unit

Table 1

Results of experimental studies

№ of test	Water volume, ml	Temperature, °C	Excessive pressure, bar	Time, h	Mass of hydrate, g	Gas volume, ml	*Q* gas content, l/kg	Surfactant, ml
1	500	0	4	2	16.81	3	0.17	0
2	500	0	3.25	3.5	188.4	49	0.26	0
3	500	0	4	3	106.4	15	0.14	0
4	500	0	3	2	20	0	0	0
5	500	0	0.3	1	40.18	40	0.9	0
6	500	0.2	5	1	22	0	0	0
7	500	0.3	2.75	2.5	87.77	850	6.61	0
8	500	0.3	5	2	18	0	0	0
9	500	0.5	2.5	3	95.75	270	2.81	0
10	500	3	3.75	2	21	0	0	0
11	500	4	4	2	19	0	0	0
12	500	4.3	2.5	2	50.85	75	1.47	0
13	500	0	3.5	4	170.4	210	2.96	0.5
14	500	0	3.5	4.5	41.88	230	5.49	0.5
15	500	0	3.5	5	68.7	1000	14.56	0.5
16	500	2.2	2.5	2	172.67	560	3.24	0.5
17	500	2.6	2.5	2	163.05	270	1.66	0.5
18	500	1	2.5	2	19.5	0	0	0.5
19	500	0.2	2.75	4	110	1095	9.95	1
20	500	0.8	2	0.7	20.93	1450	69.28	1
21	500	1	2.75	2.5	24	150	6.25	1
22	500	1	1.5	2	56.5	3900	69.03	1
23	500	1.4	0.5	2	16.16	220	13.61	1
24	500	1.5	1.5	2	9.14	600	65.64	1
25	500	2	1.5	2	46.85	1450	30.95	1
26	500	2.3	2.75	2	54.22	1450	26.74	1
27	500	5	1.5	1	23.24	400	17.21	1
28	500	1.3	1.5	1.5	19.51	70	0.28	5
29	500	0	1.5	1	12.17	230	18.9	5

ISSN 2226-3780

The water temperature varied between 0÷4.3 °C, an overpressure of 0.3÷5 bar, an experimental time of 0.7÷4.5 h, a surfactant concentration of 0–5 ml/500 ml (0÷1 %) of water. Thus, an array of initial data is obtained for subsequent analysis by mathematical statistics methods.

6. Research results

Linear, logarithmic, exponential and polynomial regressions are used as standard methods of regression analysis. After the regression dependence is determined, the significance of both the equation as a whole and its individual parameters is evaluated. Estimation of the significance of the regression equation as a whole can be carried out using various criteria.

Let's divide the whole set of data into two groups: x_i factors – include such factors as pressure, time, temperature and the surfactant amount, and the resultant attribute that is being studied is the gas content of hydrate Y.

Determination of correlation coefficients. For statistical calculation, it is decided to use multivariate analysis using the computer program «Statistics 6.0». So, there are 29 observations and a significance level $a=0.05$.

The initial data for the statistical analysis are given in Table 2.

Table 2

Initial data for statistical analysis

No.	Time, X_1	Pressure, X_2	Temperature, X_3	Surfactant, X_4	Gas content Q, Y
1	2	4	0	0	0.17
2	3.5	3.25	0	0	0.26
3	3	4	0	0	0.14
4	2	3	0	0	0
5	1	0.3	0	0	0.9
6	1	5	0.2	0	0
7	2.5	2.75	0.3	0	6.61
8	2	5	0.3	0	0
9	3	2.5	0.5	0	2.81
10	2	3.75	3	0	0
11	2	4	4	0	0
12	2	2.5	4.3	0	1.47
13	4	3.5	0	0.5	2.96
14	4.5	3.5	0	0.5	5.49
15	5	3.5	0	0.5	14.56
16	2	2.5	2.2	0.5	3.24
17	2	2.5	2.6	0.5	1.66
18	2	2.5	1	0.5	0
19	4	2.75	0.2	1	9.95
20	0.7	2	0.8	1	69.28
21	2.5	2.75	1	1	6.25
22	2	1.5	1	1	69.03
23	2	0.5	1.4	1	13.61
24	2	1.5	1.5	1	65.64
25	2	1.5	2	1	30.95
26	2	2.75	2.3	1	26.74
27	1	1.5	5	1	17.21
28	1.5	1.5	1.3	5	0.28
29	1	1.5	0	5	18.9

Linear approximation of the gas content from time is shown in Fig. 4, and is described by the equation:

$$Y=22.6778-4.3738X_1. \quad (2)$$

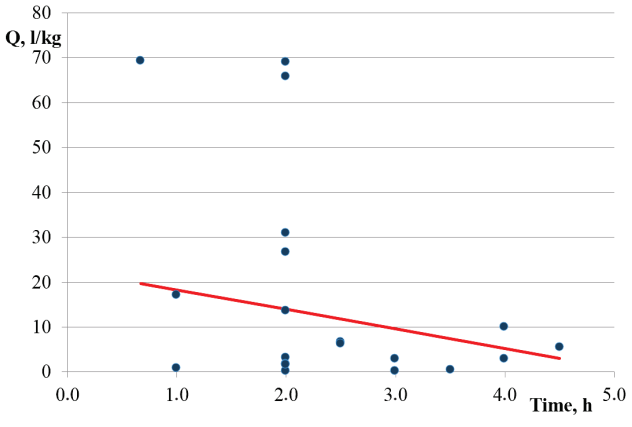

Fig. 4. Graph of gas content dependence on time

The correlation coefficient for the dependence (2) $r=0.22$. The Fisher criterion F_p is 1.41 and the value of $F_{crit}=2.20$. Since $F_p \le F_{crit}$, obtained regression equation is assumed to be statistically insignificant.

The hypothesis of the adequacy of the model was not confirmed.

As it is possible to see, the correlation coefficient is insignificant, hence the probability of this factor is only 22 %.

The dependence of the gas content of the hydrate on pressure is shown graphically in Fig. 5, which is described by the equation:

$$Y=33.1302-7.6178X_2. \quad (3)$$

The correlation coefficient for the dependence (3) $r=0.43$. The Fisher criterion is 6.16 and the value of $F_{crit}=6.16$. Since $F_p=F_{crit}$, obtained regression equation is assumed to be statistically significant.

The hypothesis of the adequacy of the model was confirmed.

As it is possible to see, the correlation coefficient is 43 %. This indicates that the gas content is inversely proportional to the pressure. As the pressure increases, the gas content decreases.

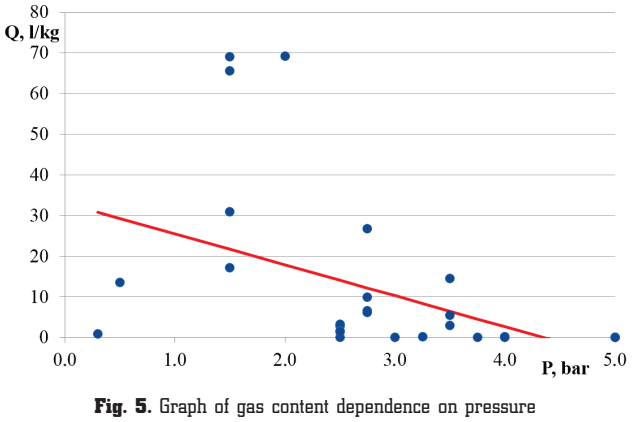

Fig. 5. Graph of gas content dependence on pressure

The dependence of the gas content on temperature is shown graphically in Fig. 6, which is described by the equation:

$$Y = 11.8935 + 0.6647X_3. \quad (4)$$

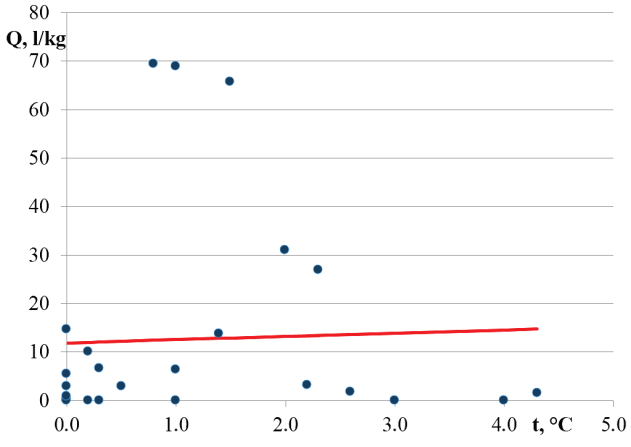

Fig. 6. Graph of gas content dependence on temperature

The correlation coefficient for the dependence (4) $r = 0.46$. The Fisher criterion is 0.06 and the value of $F_{crit} = 0.06$. Since $F_p = F_{crit}$, obtained regression equation is assumed to be statistically significant. The hypothesis of the adequacy of the model was confirmed.

As it is possible to see, the correlation coefficient will be 46 %. This indicates that the gas content of the hydrate depends directly on the temperature change. As the temperature rises, the gas content increases.

The dependence of the gas content of gas hydrates on surface-active substances is shown graphically in Fig. 7, which is described by the equation:

$$Y = 10.282 + 3.1787X_4. \quad (5)$$

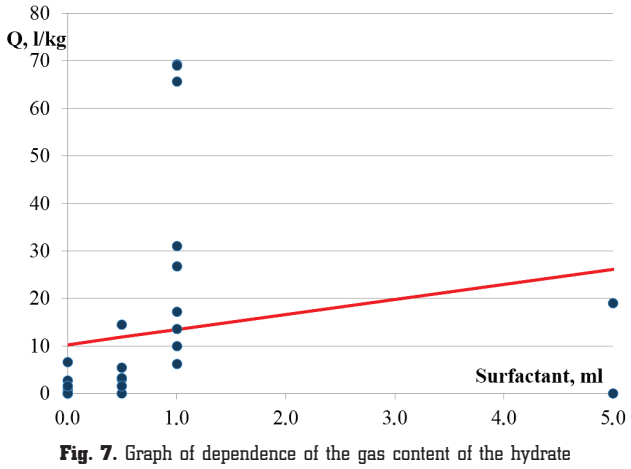

Fig. 7. Graph of dependence of the gas content of the hydrate on the number of surfactants

The correlation coefficient for the dependence (5) $r = 0.19$. As it is possible to see, the correlation coefficient is insignificant, hence the probability of this equation is only 19 %. The Fisher criterion is 1.012 and the value of $F_{crit} = 2.20$. Since $F_p \le F_{crit}$, the regression equation obtained is assumed to be statistically insignificant.

The obtained result of the experiment shows that the concentration of the surfactant has a sufficiently large effect on the gas content of the hydrate.

Using the computer program «Statistics 6.0», the coefficients of the regression equation (6) are determined:

$$Y = 38.796 - 0.4X_4 - 0.99X_3 - 7.36X_2 - 2.13X_1. \quad (6)$$

The correlation coefficient for the dependence (6) $r = 0.45$. The obtained value of the correlation coefficient indicates that the probability will be 45 %. The Fisher criterion is 1.46, and the value of $F_{crit} = 2.20$. Since $F_p \le F_{crit}$, the regression equation obtained is assumed to be statistically insignificant. The hypothesis of the adequacy of the model was not confirmed.

The obtained experimental studies show that the coefficients of multiple correlation according to equations (2)–(6) are 0.19–0.46. This means that linear regression does not allow taking into account all the features of the experimental data. In a similar scheme, with the help of «Statistics 6.0», coefficients of regression equations are calculated on the basis of logarithmic, exponential and polynomial functions. All of them give low correlation coefficients. Therefore, for the final determination of the approximating formula, the method of variation of coefficients and types of functional dependences of the regression equation is applied.

Based on the results of the experiments, the main factors affecting the formation of propane hydrate are the pressure and concentration of the surfactant. Fig. 8 shows the results of approximating the simultaneous action of two factors: amount of surfactant and pressure. Using the method of variation of the coefficients and types of functional dependencies, an approximation function of the form is obtained:

$$Q = 1.4\frac{1+36P^2}{1+0.018P^6}\left(1.039 - \ell^{-1.15z}\right), Q \le Q_T, \quad (19)$$

where P – excess gas pressure, bar; z – surfactant concentration; Q_T – the maximum theoretical value of the gas content of the hydrate. For propane hydrate, the maximum value of $Q_T = 73$ l/kg.

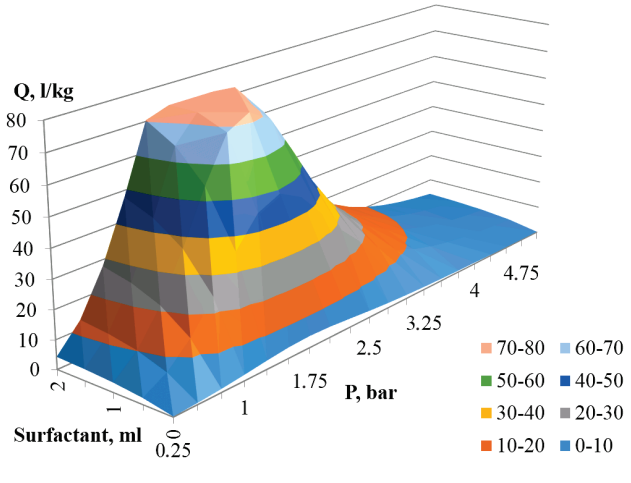

Fig. 8. Approximation of the gas content of propane gas hydrate as a function of pressure and concentration of surfactants

ISSN 2226-3780

Analysis of time and temperature requires further study. To do this, let's analyze the «residues» of the difference between the obtained values (from the results of the experiments) and the approximation function, the results of which are reflected in Fig. 9. From the obtained graph, it can be concluded that the horizontal trend line indicates the independence of the gas content of the hydrate from the time of the experiment. This is confirmed in separate experiments, when during the depressurization of the reactor the hydrate is formed almost immediately. This conclusion is of great importance for the industrial production of hydrate. It means that this technology allows to obtain hydrate very quickly.

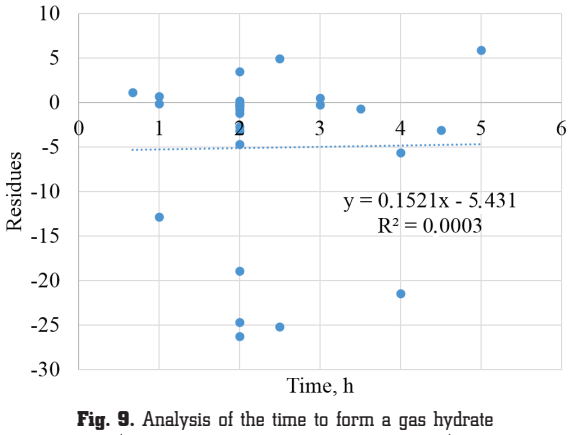

Fig. 9. Analysis of the time to form a gas hydrate (the trend line is shown as a dotted line)

To analyze the effect of water temperature, let's also construct a residue diagram with a trend line (Fig. 10). A small inclination angle of the trend line indicates a slight influence of the water temperature on the gas content of the hydrate.

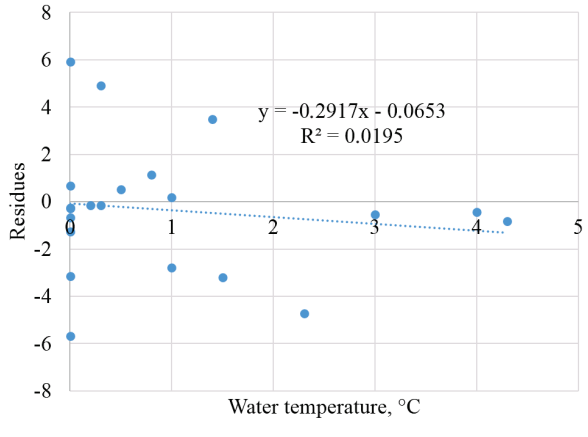

Fig. 10. Analysis of the effect of water temperature on the hydrate form (the trend line is shown as a dotted line)

So, it is possible to conclude that within the range of water temperatures from 0 to +5 °C, its effect on the gas content of the hydrate can be neglected.

7. SWOT analysis of research results

Strengths. The research results show that there is a range of pressures and concentrations of surfactants, in which high-quality hydrate of propane can be rapidly obtained. Application of these parameters in the technological process will increase the volume of production of gas hydrate and reduce its cost price.

Weaknesses. The hydrate is always preceded by a 10–15 minute stage of formation of microcrystals. For industrial production of large amounts of hydrate, this stage also requires intensification.

Opportunities. Opportunities for further research are the determination of the influence of other factors on the rate of formation and the quality of the synthesized hydrate, such as the dimensions of the bubbles, the intensity of mixing, the use of attachments of different interstitial ability to the like. Further research aimed at optimizing the synthesis of hydrates of other gases or their mixtures, which are of great importance for introducing gas hydrate technologies into production.

Threats. To operate the gas hydrate synthesis plant on an industrial scale, recirculation of the exhaust gas must be used. The use of recirculation requires the use of an additional compressor and the corresponding energy consumption for its operation.

8. Conclusions

1. Laboratory unit for the synthesis of gas hydrate in bubbling mode with the possibility of specifying controlled external factors is constructed. Also, an additional unit for determining the gas content of the hydrate is collected. Thus, the integrated use of all equipment allows performing studies of the influence of various factors on the gas content of the resulting gas hydrate.

2. 29 full-scale experiments are conducted in which the influence of 4 factors on the gas content of propane hydrate: gas pressure, water temperature, surfactant concentration and the time of the experiment is studied.

3. With the help of statistical analysis it is established that the pressure and concentration of surfactants have the greatest influence on the quality of the synthesized hydrate. The obtained experimental studies show that the coefficients of multiple correlation are 0.19–0.46. This means that the standard regression relationships do not allow to take into account all the features of the obtained results. Therefore, the search for the optimal approximation dependence is performed using the method of variation of coefficients and types of functional dependencies. An approximate formula is obtained for determining the predicted gas content of the hydrate.

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UDC 621.316
DOI: 10.15587/2312-8372.2018.141391

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STUDY INTO ENERGY EFFICIENCY OF THE DRIVE OF ELECTRIC VEHICLES WITH AN INDEPENDENT POWER SUPPLY DEPENDING ON THE CONFIGURATION OF THE POWER SOURCE

Об'єктом даного дослідження є електричний транспортний засіб з автономним джерелом живлення. На сьогодні подібні транспортні засоби займають все більшу нішу на автомобільному ринку, витісняючи своїх конкурентів з двигуном внутрішнього згорання за рахунок вищої енергетичної ефективності. І хоча ця перевага над транспортом з двигуном внутрішнього згорання є очевидною, з точки зору електричних систем ККД електромобілів залишається доволі невисоким. Проблемним місцем, яке накладає ці обмеження, є джерело живлення – літій-іонний акумулятор – який має значний внутрішній опір.

Для усунення цього недоліку на прикладі приводу електроскутера з асинхронним двигуном з короткозамкненим ротором досліджувалась схема з підключенням паралельно до акумулятора батареї суперконденсаторів. Суперконденсатори мають значно менший внутрішній опір і тому беруть на себе основне миттєве навантаження при перехідних процесах: розгоні та гальмуванні, коли через джерело протікають найбільші струми.

Дослідження показали, що подібна конфігурація покращує енергетичну ефективність транспортних засобів. Причому існує оптимальне значення необхідної ємності суперконденсатора для досягнення найбільшої ефективності (найменшого споживання енергії). Це пов'язано з тим, що батарея суперконденсаторів є доволі габаритним об'єктом і суттєве збільшення ємності призводить до збільшення маси транспортного засобу і відповідно до збільшення споживання енергії. Додатково була досліджена покращена система живлення, в якій суперконденсатор пришвидшено заряджається під час пауз руху транспортного засобу. Вона дозволила покращити вже отримані результати, ще зменшивши споживання електричної енергії.

У порівнянні з проведеними раніше дослідженням було показано важливість правильного вибору ємності суперконденсаторів та системи контролю живлення. Була доведена наявність точки оптимуму та чисельно продемонстрована різниця показників споживання енергії в цій та в інших точках.

Ключові слова: *система приводу електроскутера, літій-іонний акумулятор, паралельне з'єднання суперконденсатора та акумулятора, міський їздовий цикл.*

1. Introduction

Current trend in the transportation industry demonstrates that electric automobiles that have rapidly developed recently are gradually displacing their analogs driven by the internal combustion engine as they are more energy efficient and environmentally friendly. Economic approach when selecting a vehicle shows that the higher