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STUDY OF ADSORPTION PROCESSES INFLUENCE ON DEVELOPMENT OF NATURAL GAS FIELDS WITH LOW-PERMEABILITY RESERVOIRS

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Розробка нетрадиційних родовищ природних газів все ще перебуває на стадії свого становлення. Тому на даному етапі надзвичайно актуальними залишаються питання вдосконалення технології підвищення ступеня вилучення газу вже на початковій стадії розробки. Враховуючи світовий досвід у даному напрямку, дослідження, пов'язані з вивченням адсорбційно-десорбційних процесів у низькопористих низькопроникних родовищах природних газів, можуть стати ключовими для забезпечення високих коефіцієнтів вуглеводневилучення та поточних відборів газу. У даній роботі представлено аналіз вітчизняних та закордонних літературних публікацій, а також результати експериментальних досліджень на насипних моделях низькопроникних пластів щодо визначення їх адсорбційних параметрів. Зокрема вперше досліджено та проаналізовано адсорбційні процеси в ущільнених пісковиках, встановлено залежності між кількістю адсорбованого газу, проникністю, тиском та температурою. Результати лабораторних експериментів були піддані статистичній обробці та побудовано модель за методом нейронних сіток, яка дозволяє оперативно оцінити залежність між кількістю адсорбованого газу та термобаричними умовами і проникністю. Обґрунтовано напрямки подальших досліджень та зроблено відповідні висновки.

Ключові слова: природний газ, низькопроникні колектори, нетрадиційні родовища, адсорбція, десорбція.

Разработка нетрадиционных месторождений природных газов все еще находится на стадии своего становления. Поэтому на данном этапе чрезвычайно актуальными остаются вопросы совершенствования технологии повышения степени извлечения газа уже на начальной стадии разработки. Учитывая мировой опыт в данном направлении, исследования, связанные с изучением адсорбционно-десорбцийных процессов в низкопористых слабопроницаемых месторождениях природных газов, могут стать ключевыми для обеспечения высоких коэффициентов углеводородоотдачи и текущих отборов газа. В данной статье представлен анализ отечественных и зарубежных литературных публикаций, результаты экспериментальных исследований на насыпных моделях слабопроницаемых пластов по определению их адсорбционных параметров. В том числе впервые исследованы и проанализированы адсорбционные процессы в уплотненных песчаниках, установлены зависимости между количеством адсорбированного газа, проницаемостью, давлением и температурой. Результаты лабораторных экспериментов были подвергнуты статистической обработке и построена модель по методу нейронных сетей, которая позволяет оперативно оценить зависимость между количеством адсорбированного газа в зависимости от термобарических условий и проницаемости. Обоснованы направления дальнейших исследований и сделаны соответствующие выводы.

Ключевые слова: природный газ, слабопроницаемые коллектора, нетрадиционные месторождения, адсорбция, десорбция.

Development of unconventional natural gas fields is still in the stage of its formation. Therefore, the issues of improving the techniques for increasing gas recovery rate at the initial stage of development are extremely topical. Taking into account the international experience in this field, the studies related to the study of adsorption-desorption processes in low-porosity low-permeability natural gas fields may become the key factor for providing high rates of hydrocarbons recovery and current gas extraction. This paper presents the analysis of domestic and foreign scientific publications and results of experimental studies on the sand packed models of low-permeability reservoirs in order to determine their adsorption parameters. In particular, adsorption processes in tight sandstones were studied and analyzed for the first time and the dependences between the amount of adsorbed gas, permeability, *pressure and temperature were determined. The laboratory experiments results were subjected to statistical analysis and the model, which allows to estimate the dependence between the amount of adsorbed gas and thermobaric conditions and permeability, was developed with the help of the neural networks method. Directions for further studies were grounded and correspondent conclusions were made.*

Keywords: natural gas, low-permeability reservoirs, unconventional fields, adsorption, desorption.

Introduction

Growth of global trends of natural gas consumption on the background of the existing conventional fields depletion was a prerequisite of raising natural gas prices, which led to the development of new and improvement of existing technologies for its production. The dominant role of natural gas as the main source of energy will continue for the next decade. One of the main sources of additional gas production is unconventional natural gas deposits, which include deposits with low-permeable low-porous reservoirs [1].

According to the Energy Information Agency report (EIA, July 2013) technologically recoverable resources of shale gas in Ukraine equal 3.62 trln.m^3 (1.75% of world reserves), and including resources of tight gas reach 7 trln.m^3. Proven natural gas reserves of conventional deposits equal 1 trln.m^3 [3]. Previously (in 2011) US Energy Information Administration estimated the technically recoverable resources of Ukrainian shale gas at 1.2 trln.m^3 (0.6% of the estimated world reserves), and total – at around 5.6 trln.m^3. According to the Dixi Group report, shale gas resources in Ukraine vary and range from 5 to 8 trln.m^3, with technically recoverable 1-1.5 trln.m^3 [2].

The main prospective areas in Ukraine are the Lviv-Lublin basin in the west region (recoverable resources estimated at 1.47 trln.m^3) and the Dnieper-Donetsk Basin in the east region (recoverable resources of approximately 2.15 trln.m^3) [2].

Oleska area, which includes shale and natural gas, coalbed methane, oil and hydrocarbon condensate, is located within the Lviv and Ivano-Frankivsk regions and takes about 6.5 th. km^2. State Service of Geology and Mineral Resources of Ukraine had estimated resources of Oleska area in 2.98 trln.m^3 and Yuzivska area in 7.886 th. km^2 – 4.054 trln.m^3 of different types of gas (including tight gas) [4].

Skifska area is located on the continental shelf of the Black Sea and its area is 16 698.2 km^2, and the depth at the site reaches 300-2000 m. The potential of natural gas at the area was estimated at 5.10 billion m^3 per year, and total recoverable resources are within the range of 200-250 billion m^3.

Another promising site is Slobozhanska area, located in the Kharkiv region, with area of about 6000 km^2. Technologically recoverable resources of shale gas and tight gas are estimated at 50-70 billion m^3, of hydrocarbon condensate – 2 mln. tons [4].

The main coal bed methane resources are concentrated in Ukraine Donetsk and Lviv-Volyn coal basins. Total coalbed methane resources in Ukraine equal 12-13 trln.m^3, of which technology available resources are 3-3.5 trln.m^3. Long-term production of coal bed methane is estimated at 2.12 billion m^3 per year. Expected cost of coal bed methane production, according to Baker Tilly Company, is 2300-3300 UAH/thn.m^3 [2].

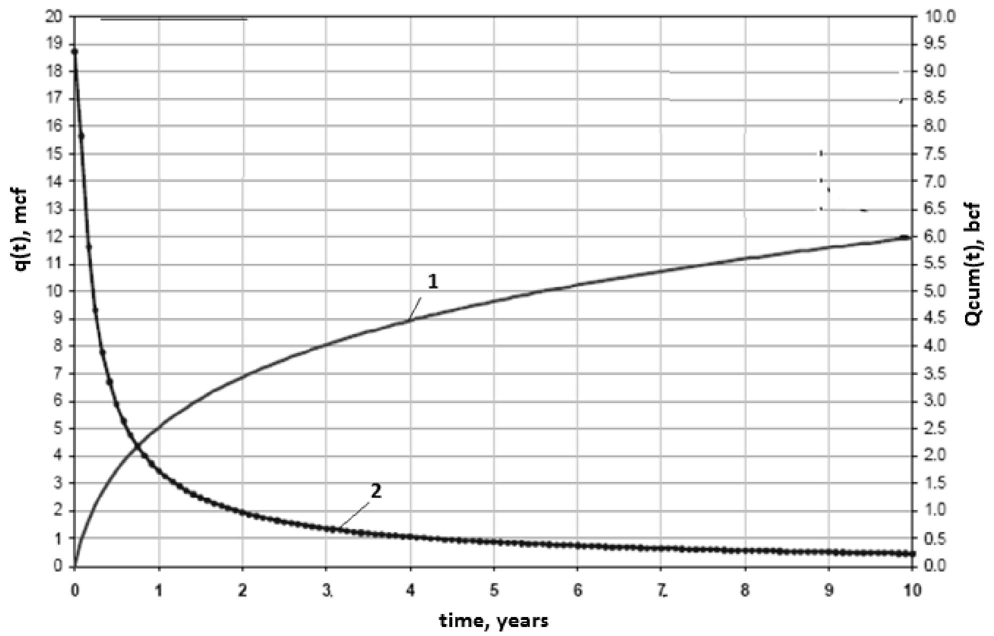
Critical literature review

One of the main differences between the development of conventional and unconventional natural gas fields is the presence of the respective stages of production. In conventional gas fields development there are following periods of gas production: increasing gas production, constant gas production and production gas declining, and after that they proceed to completion and field abandonment [5]. In case of unconventional fields drop-down production is observed from the beginning of their development [6]. For example, production decline curves for Heynesville shale gas field are given below (see. Fig. 1), were estimated by Chesapeake Energy Company [7, 8].

As it can be seen from Figure 1, the flow rate drop for production wells is quite fast. During the first year well production rate may decrease to 65-80%, during the second year - 35-45%, in the third - 20-30%. After a sharp decline of production rate relatively stable plato at the site is observed, which is called the "tail" of development. During the final stage of development the percentage production decline is reduced and in average it can range 5-7% of the previous year. This "tail" can last for decades, but it is limited to cost-effective production rate (minimum reservoir pressure).

The foregoing features of unconventional natural gas fields development could be explained by the peculiarities of gas occurrence mechanism in low-porous low-permeable reservoirs. Natural gas contained in shale deposits and coal seams is in the free state in the pores of the rock matrix and in the adsorbed state on the surface of pores space [8, 11, 12]. As it was established according to the field data of unconventional natural gas deposits development the amount of adsorbed gas may reach 40-50% of the initial reserves. Consideration of adsorption processes in predicting development strategies will allow engineers to more accurately determine the reserves and predict the final gas recovery factor [10]. Therefore, special attention should be given to the development of new methods for forecasting the indices of unconventional natural gas field development that will consider the peculiarities of unconventional natural gas fields and will give reliable results.

Accumulated international experience testifies that the development of low-porous low-permeable reservoirs with economically profitable production rate can be achieved by the infill drilling of horizontal wells with further intensification of gas inflow. Currently, high-performance and factually the only method of intensification is multistage



1 - cumulative gas production; 2 – well flow rate

Figure 1 – Production decline curves for Heynesville shale

hydraulic fracturing [13]. The main parameters that affect the performance of wells by using this technology is the length of the horizontal section of the wellbore, the number of perforated intervals, the number of HF fractures, their length, density and permeability.

Meyer et al. (2010) suggested considering the volume of the reservoir is limited by transverse fractures as simulated (drained) reservoir volume (SRV) [14]. The authors identified the main factors that influence on the size of SRV: formation thickness, the length of the horizontal section of the wellbore, the distribution of stresses in the layer, the presence of natural fractures. It was determined that in order to increase the stimulated reservoir volume should consider the increase fractures density, wells perforating parameters, wells horizontal section orientation, open hole well completion and others.

In [15], the authors describe the specific features of gas production from tight sands. In particular, they include:

1. Identification of the most promising areas (sweet spot) in the productive layers which are areas with high porosity, permeability, high reservoir pressure increased compared to the rest of the reservoir, the presence of natural cracks. In deposits developing gas flows from remote areas to the most promising zones where wells have been drilled. If there are no such zones, commercial gas production is not possible without hydraulic fracturing.

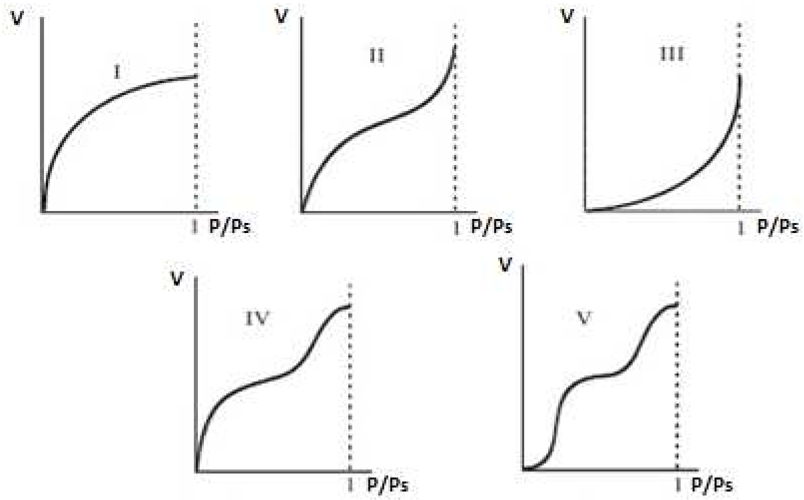
2. The increase of stimulated reservoir volume.

3. Low-permeable formation pollution and overlapping the channels for gas inflow by drilling fluids, HF liquids etc.

As it was established by the results of field research works and gained field experience of gas production from unconventional deposits, some amount of gas is in adsorbed state [16, 17, 18]. One of the possible and main ways of gas production increase from low-porous low-permeable reservoirs is gas desorption intensification from the surface of the pore channels.

The nature of adsorption forces is very different. In general, the adsorption can be divided into physical and chemical (chemisorption). With much appearance of van der Waals forces, the adsorption is called physical, but when the forces are valent, i.e. it is when the adsorption is accompanied by the formation of surface chemical compounds it is called chemical. In physical adsorption equilibrium is established quite quickly and is reversed. Physical adsorption can be caused by electrostatic forces; while adsorption is determined by the chemical nature of the adsorbate molecules. Chemisorption can be both fast and slow. It differs from physical adsorption in that it is more sensitive to the chemical nature of the adsorbent and adsorbate. [19]. Another distinguishing feature of chemisorption is its irreversibility and high thermal effects (hundreds of kJ/mol). Between physical and chemical adsorption, there are many intermediate cases (e.g., adsorption, due to the formation of hydrogen bonds). Adsorption processes in shale formations were described by Langmuir law [10, 20].

Monomolecular Langmuir adsorption theory is based on certain assumptions. In particular, according to the theory of Langmuir adsorption occur not on the entire surface of the adsorbent, but on the active centers, which are projections or depressions on the surface of the adsorbent; adsorption is local and is caused by the forces closed to chemical ones; active centers are considered to be independent and identical; each active center is able to interact with only one molecule of adsorbate; adsorption process is reversible and equilibrium; as a result of adsorption monomolecular layer is formed.



V – adsorbed gas content, m^3*/t; P/Ps – ratio of pressure to vapor saturation pressure*

Figure 2 – Types of adsorption isotherms

Taking into account these assumptions in real conditions the character of adsorption isotherms often differes from Langmuir once. In particular there are 5 types of adsorption isotherms, shown in Figure 2.

Existing of such isotherms is explained by the fact that the Langmuir theory does not consider the interaction between adsorbed molecules, the real structure of the surface of the adsorbent and also the possibility of further adsorption of multiple layers.

Type I isotherms reflect monomolecular adsorption. Isotherms of types II and III are usually associated with the formation of the multimolecular adsorption. Isotherms of types IV and V differ in that they are characterized by finite adsorption in approaching the vapor pressure to the saturation pressure Ps. Isotherms of types II and III are characteristic for the adsorption on non-porous adsorbent, and types IV and V for porous solids. All five types of adsorption isotherms are described by the theory of multimolecular adsorption "BET", named so by the initial letters of the authors (Brunauer, Emmett, Teller).

In the BET theory supplementary assumption was adopted that each molecule of the previous layer is a possible active center for the next layer adsorption. Therefore multimolecular adsorption isotherm has S-shaped character.

To describe the isotherms of types 4 and 5 M. Polyani first proposed the theory of multimolecular adsorption based on completely different ideas than the Langmuir theory [21, 22]. In particular, this theory is based on assumptions about the potential field of solid body surface on which adsorbate molecules fall. Using this approach adsorbed layer resembles the atmosphere which is compressed near the surface, and is sparse in the outer layers. According to the Polyani theory the character of adsorption isotherm for a particular adsorbate doesn't dependent from temperature. Therefore, if the combination of adsorption isotherms at different temperatures can get one curve, and if the results of experiments represented as $\lg(\vartheta/\vartheta_m)=f(RT\cdot\ln(P/P_S)$ as a result can be obtained curve, the shape of which is independent from temperature. This curve called the adsorption characteristic curve.

In addition, it is advisable to speculate that the monomolecular adsorption is only a partial case of multimolecular adsorption, which can be described by BET isotherms. The process of the multimolecular layer formation may be accompanied by phase transitions of gas-liquid [19].

One of the first methods for the determination of adsorption parameters is so-called direct method, firstly proposed by Bertard (1970) and later improved by Kissell (1973). The essence of this method is to measure the amount of desorbed gas from the pore space surface of the sample species, obtained from the experimental cell and getting into a measuring flask displaces the same volume of fluid [23, 24, 25].

More accurate method was developed by the US Bureau of Mining for the determination of even small amounts of desorbed gas (Schatzel, 1987), which, unlike the previous method, based on the additional measurement of the pressure in the tank and expenses during the gas releasing, and then use the equation of state to determine the desorbed gas volume, which consists of a pressure vessel (desorption canister) pressure gauge, the input and output valves and gas flow meter. The specificity of such experiments is very low rate of working agents at the model output that need to be fixed, and the need for special sensors to determine the instantaneous concentration of different gases or surfactants. This method is characterized by accuracy and reliability of the results and is easy of implementation. That is why it have become a significant spread in the world.

In [26] presented the results of studies of gas adsorption on the surface of the pore space for shale rocks (so-called Devonian shale). Studies were conducted using laboratory setup consisting of the reservoir model, additional capacitance manometers, thermometers and valves. It should be noted that in this work for the first time an analysis of errors during the measuring of the amount of adsorbed gas, which can be caused by inaccurate

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determination of temperature, pressure or empirical equations of state errors. According to the results total error during adsorption-desorption experiments equals in average 1.1%. Possible error of amount of methane adsorbed measuring on illite was grounded, a method for reducing errors in determining the temperature and pressure was developed.

Investigation of the intensity of methane desorption from coal from the initial equilibrium pressure is given in [27]. The research was conducted using volumetric method. It was found that at low pressures (3 MPa), this dependence is exponential, and when more pressure is converted into a quadratic. The effect is due to the presence of the transition phase desorption, which changes the priority of the mechanisms leading role of methane - filtration and diffusion. The results of experimental studies show that between the initial and final phases of the methane desorption from coal there is a long transition phase, during which a change in the leading role of methane flow mechanisms – from filtration to diffusion. For researchers engaged in practical use of scientific developments, it is important to a violation of proportionality between the methane content in coal and the intensity of its allocation in the department of pieces of coal from the seam.

In the desorption kinetics there are two phases: initial and final. In [28] it was found that in the initial phase of desorption is adsorbed and free methane of from coal models. A specific feature of the initial phase is gas filtration in the open pores. In this phase, the pressure in the pores is reduced from several megapascals (initial equilibrium pressure) to several kilopascal. The duration of the initial phase depends on the coal models size and can last from fractions of seconds to a few tens of seconds, and the amount of released gas is approximately 30% of the total content. Final phase is characterized by the long duration and low intensity of gas discharge. Lack of or poor display of transient processes makes the final phase of an affordable and convenient for the desorption study.

Investigation of gas flow in coal seams is presented in [29]. The movement of gas in the reservoir initiates a number of processes: molecular diffusion (predominance caused by collisions between molecules), Knudsen diffusion (predominance caused by collisions of molecules with the walls of the pores) and surface diffusion (movement of gas in the adsorbed layer). The consequences of these processes are in-sity displacement of methane molecules. This method consists of three phases: gas flow in fractures, gas diffusion and rock matrix and matrix adsorption effects (mainly in the micropores).

To increase coal bed methane production non-hydrocarbon gas injection is widely used in order to reduce the methane partial pressure in the reservoir. In the same time reservoir pressure does not decrease, and may even increase. It allows maintaining a constant well flow rate without lowering the deposit energy potential. Injected CO_2 mainly adsorbed on the surface of the pore space, displacing CH_4 from coal. Displacement ratio of CH_4: CO_2 ranges from 2: 1 to 10: 1. In case of N_2 injection methane desorption increase not only due to substitution and nitrogen adsorption and by reducing the methane partial pressure. Reducing the CH_4 partial pressure provides the driving force for desorption. Implemented pilot projects of CO_2 and N_2 injection in order to improve gas recovery of coal deposits have shown successful results. Increasing coal bed methane recovery by carbon dioxide injection was considered in [30]. CO_2 injection in coal seams will not only increase the ratio gas recovery but also reduce the amount of greenhouse gases in the atmosphere through their underground sequestration. An analysis of the project in San Juan coal basin (USA) using well pattern with consist from 4 injection and 7 production wells shows its economic and technological effectiveness. In particular, injection of 56.6 million m^3 of carbon dioxide increase natural gas production by 150%, without CO_2 breakthrough to the production wells. For the successful usage of this technology the coal seam should have limited size, relatively high permeability and lithological irregularities and lack of significant natural fractures. Injection wells should be completed unstimulated, while production wells can be cavitated or hydraulically stimulated. In addition to CO_2 injection another possible method of increasing gas recovery is methane displacement by nitrogen. Using this method may be achieved final recovery factor near 90%. Sources of CO_2 for injection may become natural deposits. However, to improve the ecological situation a rational option could become it transportation from large factories, plants, etc., which is extremely expensive. Therefore, at the design stage of this method economic parameters should to be considered.

Mining of coalbed methane at depletion mode is relatively simple and cheap method. But it is produced only up to 50% of initial gas reserves [31]. In this situation, the authors examined the possibility of increasing gas recovery using nitrogen and helium. The physical essence of the process is that the pumping of non-hydrocarbon gas decreases the methane partial pressure, which in turn initiates its desorption without reservoir pressure reducing. As it was found in the result of experiments amount of adsorbed methane on the rock surface depends not only on temperature and pressure under which it is located, but also on its concentration in the gas. This conclusion is based on the results of the experiment on the methane and helium adsorption with different concentrations of different concentration on coal models surface. To investigate the influence of methane partial pressure on desorption, a series of experiments that included methane adsorption on the surface of coal was conducted. After reaching pressure equilibrium helium was injected to the model inlet while methane was produced from the model outlet into additional cell. The process was carried out in stages with maintaining constant pressure in the model (7MPa). After a certain period of time the model was closed on both sides and pressure values have been recorded. Methane desorption was occurred in the model at this time, as was evidenced by pressure increasing. Knowing the amount of injected helium, methane, gas concentration at the model outlet and thermobaric parameters based on the material balance conditions the amount of additional extracted methane was determine. However, as helium is relatively expensive gas for industrial usage, a number of studies related to the nitrogen injection in order to intensify methane desorption by the foregoing method was conducted. It should be noted that the nitrogen adsorption capacity is 40% lower than methane. Amount of injected nitrogen equals about 3 pore volumes. In this case all free and about 80% of the adsorbed gas was produced. Laboratory experiments were conducted on sand packed and core models, and showed great efficiency of nitrogen injection in order to enhance gas recovery from coal bed methane fields.

In [32] the question of methane, nitrogen and carbon dioxide adsorption on coal samples from other deposits USA was studied. Before the experiments coal samples were crashed, purified and screened. After that 25 cm in length and 4,25 cm in diameter cylindrical container was filled by coal. Model porosity and permeability was measured using helium because it is not adsorbed on the coal surface. As it was measured porosity equal 37%, permeability - 31mD. All studies were conducted at 22°C using gravimetric method. According to the experimental results, it was found that the coal from the field is absorbed three times more CO_2 than methane. Nitrogen adsorption capacity is lower than that of methane. An interesting fact is also that during desorption hysteresis was observed. And it was the largest for methane and CO_2. Some researchers attribute this phenomenon for measurement errors.

In order to determine the methane displacement ability by CO_2 and nitrogen the experiments were carried out at pressures of 2.9 and 4.1 MPa. For these experiments gases of different compositions were used (pure nitrogen, pure carbon dioxide and mixtures thereof). Thus displacement agent was injected at a constant flow rate. Based on the experiments results it was established that CO_2 breakthrough occurs after injection of 1.2 pore volume. In this case, the highest rate methane recovery is achieved after 1.5-1.8 pore volume of CO_2 injected. With regards to nitrogen, it breaks much sooner after injection of 0.5 pore volume. The maximum gas recovery reached after 2-2.5 pore volumes injection. When using a mixture of gases to methane displacement, regardless of the concentration of individual components of the first to exit the model breaks nitrogen, displacing of methane. However, the higher the concentration of nitrogen in the mixture, the sooner it breaks. Then CO_2 was break. Thus before CO_2breakthrough jump in methane production was observed.

The hydrodynamic model of depleted shale gas with two horizontal wells with transverse multistage fracturing has been used to analyze the influence of parameters of adsorption of CO_2 and CH_4 on the accumulated gas production, total volume injected CO_2 and CO_2 breakthrough time in [31]. To determine the relative adsorption capacity of CH_4 and CO_2 was used replacement rate of methane with carbon dioxide or CO_2-CH_4 relative adsorption capacity is defined as:

$$\alpha_{CO_2-CH_4} = \frac{V_{L-CO_2} \cdot P_{L-CH_4}}{V_{L-CH_4} \cdot P_{L-CO_2}}, \tag{1}$$

V_{L-CO_2}, V_{L-CH_4} – Langmuir volume for CO_2 and CH_4, m^3/t;

P_{L-CO_2}, P_{L-CH_4} – Langmuir pressure for CO_2 and CH_4, MPa.

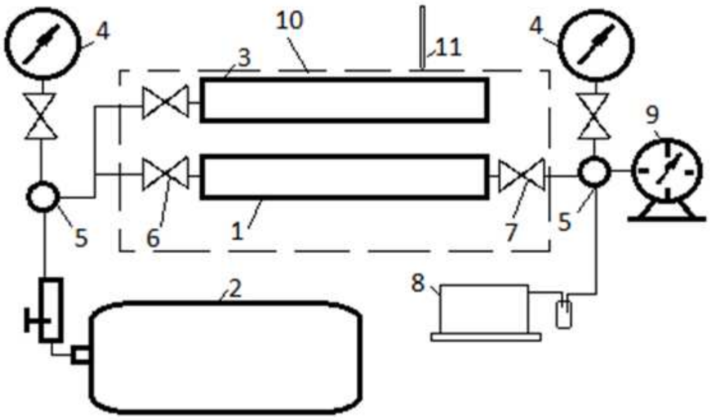
To calculate the baseline pressure and volume Langmuir methane amounted to 2 m^3/t and 5 MPa; for carbon dioxide under 3.4 m^3/t and 2.7 MPa. In order to determine the effect of these parameters on the extraction of gas from deposits held by individual launches hydrodynamic calculations stimulator for CO_2 and CH_4 for the basic version, and the parameters of the Langmuir 50% higher and lower than their value for the base case. By increasing the amount of methane Langmuir 50% final rate gas recovery increased by 3.4%, the amount of injected CO_2 is reduced by 12% and decreases the breakthrough of CO_2 and 8% decreases its production. The increase for CO_2 Langmuir 50% have no effect on the ratio of the final gas recovery, but can increase by 18.5% volume of injected CO_2 and reduce its production by 68%. Increased pressure Langmuir methane by 50% can increase gas production by 1.25% and the amount of injected CO_2 by 3.5%. However, it dramatically (by 51%) reduced time to breakthrough of CO_2 producing wells. An increase in the Langmuir pressure for CO_2 by 50% does not affect the increase gas recovery, but reduces the amount of CO_2 injected 5% significantly increases its production (69%).

Problem formulation

Although there are a large number of studies, known technology for natural gas desorption intensification from shale gas deposits and coal bed methane fields using displacement agents, there are no studies specifically for tight low-porous low-permeable reservoirs. Also, it is not clearly established the dependence on porosity, permeability, pore size, rock grain size, rock surface area and its ability to adsorbed methane at different temperatures. Determination of these dependencies will improve current gas production and increase the final gas recovery possibly not only from unconventional deposits, but also from the conventional natural gas fields. Also, the impact of non-hydrocarbon gas injection pressure on the process of desorption intensification from models with different permeability is not fully investigated.

General description of experimental research

Additional studies were conducted, which provide the opportunity to establish regularities of adsorption-desorption processes in tight sands and develop methods (technologies) that leads to increase gas recovery coefficient from low-porous low-permeable reservoirs, and, perhaps, from conventional natural gas fields.



1 – model; 2 – source of gas; 3 – reference cell; 4 – pressure gauges; 5 – manifolds; 6 – input valve; 7 – output valve; 8 – вакуумний насос; 9 – gas meter;10 – thermo bath; 11 – temperature sensor

Figure 3 – Scheme and general view of experimental setup

To investigate the adsorption- desorption processes from low-permeable reservoirs the laboratory setup was developed. A schematic diagram and its general view are shown in Figure 3. The methodology of carrying out the experiments is as follows. Model is filled with the sand of selected fractions (0.125, 0.5, 1 and 2 mm). The porosity and absolute permeability, the volume of the model lines and additional cells are determined. Model is evacuated for some hours maintaining a constant temperature, thereby releasing pore space of the model from previously adsorbed gas (including air). The temperature is maintained closes to 100°C. Some authors in their studies for model degassing evacuated it for 12 hours at 50°C. For rock samples with high clay content it is recommended to maintain a temperature of about 200°C [34].

At the beginning of the experiment a constant temperature is set, which will be maintained throughout the whole period of its duration. The experiments were conducted at temperatures 22°C, 40°C and 60°C. The model is filled with methane at a pressure P_1. The volume of methane in the model is determined by the equation of state of the gas in the pore volume for a particular temperature and pressure conditions. Pressures at the inlet and outlet of the model were measured. The model withstands some period of time to stabilize the pressure in it. This process can take from 4 to 8 hours. Throughout all period of time pressure is measured. Methane adsorption on the surface of the pore space is fixed as a result of the pressure drop in the model. According to studies after 3 hours the pressure in the model varies slightly, so stabilized value of pressure was determined in 4,5 hours. Amount of adsorbed gas is determined using the equation of its state. Then the experiment is repeated for the other values of the initial pressure. The data of studies were processed according to the known method [35].

The result of the construction of graphical dependence is checked by using the pressure drop method. To do this, valve 7 is opened and free gas is released for 4-5s to receive atmospheric pressure at the model output. Then the valve at outlet 7 is closed and the liquid flow meter is attached. To determine the amount of desorbed gas valve 7 is slowly opened. The investigation continues for as long as the gas flow will not be less than 10 ml/d.

In the experiments the model with the length of 16,7 cm and the diameter of 2,6 cm was used. The experimental setup was pressure-tested to one half of the working pressure (20 MPa). The pressure in the experiments varied from 1 to 8,9 MPa, and the maximum value of 16 MPa was achieved. Pressure measurement during the experiments was carried out with pressure gauges with accuracy class 0,15 (date of calibration 2013). Studies were conducted using experimental design theory. As a source of methane gas cylinders were used. According to the passport of natural gas quality for compliance with GOST 27577:2005 methane content equals 97%, hexane+ 0,004%, non-hydrocarbon components of about 0,8%. Gas specific gravity is 0,574, dew point temperature – minus 35,5°C. To prevent uncontrolled gas leakage on laboratory setup alarm methane gas detector "Leleka" was installed, with operating boundary 0,75% of methane concentration in air, which corresponds to 5% of the lower limit of explosion (date of calibration 13.12.2013).

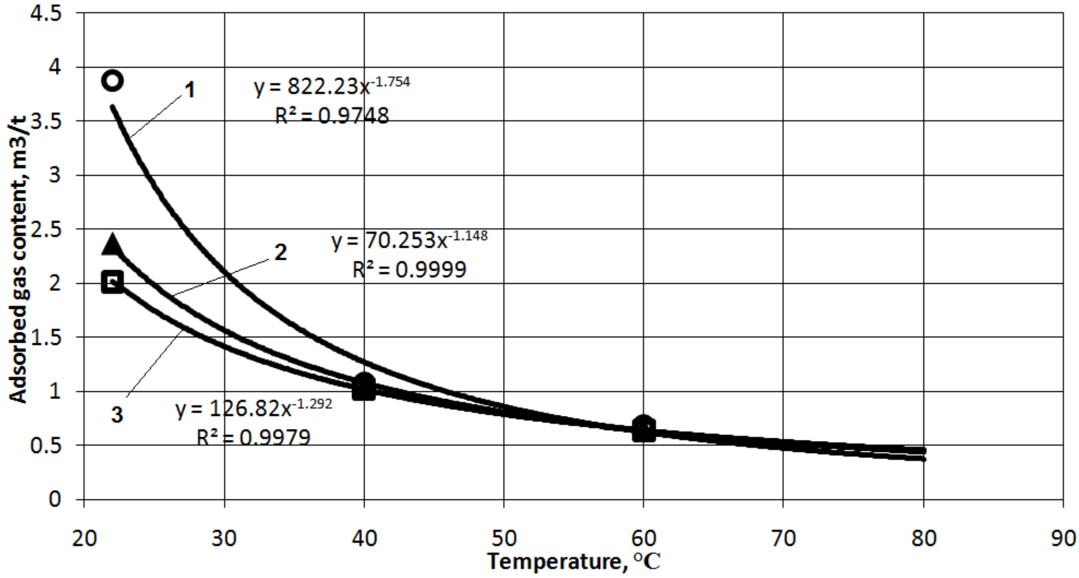
Experiments to determine the adsorption capacity of solid sandstone were conducted in three phases for different values of porosity and absolute permeability (Table 1).

Results discussion

The maximum amount of adsorbed gas on the pore space surface with the temperature increase from 40°C to 60°C decreases by 1,5 times (from

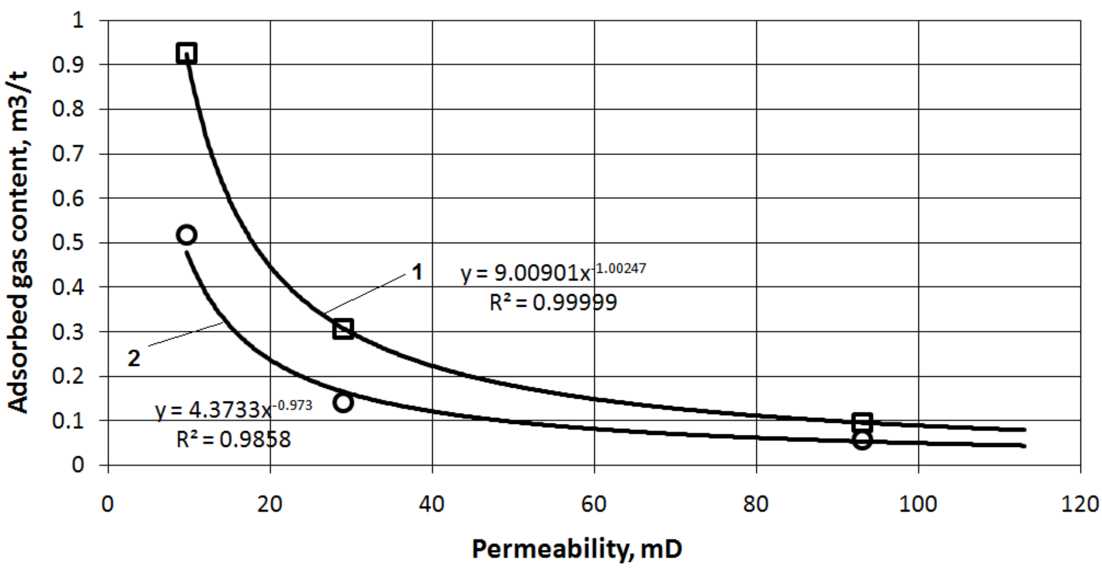
Table 1 – Experimental models description

Model	Rock mass in the model, g	Porosity, %	Permeability, mD	Pore radius, mm	Surface area of the model, m^2/m^3
1	172,39	21,01	9,7	0,001	$6{,}8 \cdot 10^5$
2	170,64	21,8	29	0,0017	$3{,}25 \cdot 10^5$
3	148,99	31,7	93	0,002	$4{,}097 \cdot 10^5$



1 – at the pressure 20MPa; 2 – at the pressure 10MPa; 3 – at the pressure 8MPa.

Figure 4 – Graphical dependences of adsorbed gas content from temperature (adsorption isobar) for different pressures (model 1 – 9,7mD)



1 – at the temperature 40 °C; 2 – at the temperature 60 °C.

Figure 5 – Graphical dependences of adsorbed gas content from model permeability for different temperatures at the pressure 3 MPa

1,2 m^3/t to 0,75 m^3/t) for model 1, by 1,2 times (from 0,43 to 0,35 m^3/t) for model 2 and by 1,5 times (from 0,25 to 0,15 m^3/t) for model 3. Moreover, at constant temperature with increasing the model permeability the amount of adsorbed gas reduces by about 80% (from 1,2m^3/t for model 1 to 0,22 m^3/t for model 3).

Analyzing the figure 4 it can be concluded that at constant pressure with increasing the temperature the amount of adsorbed gas decreases. However, when the temperature 80°C amount of adsorbed gas is weakly dependent on pressure, and an average equal 0,35-0,45 m^3/t for model 1. With the permeability increasing by 3 times the amount

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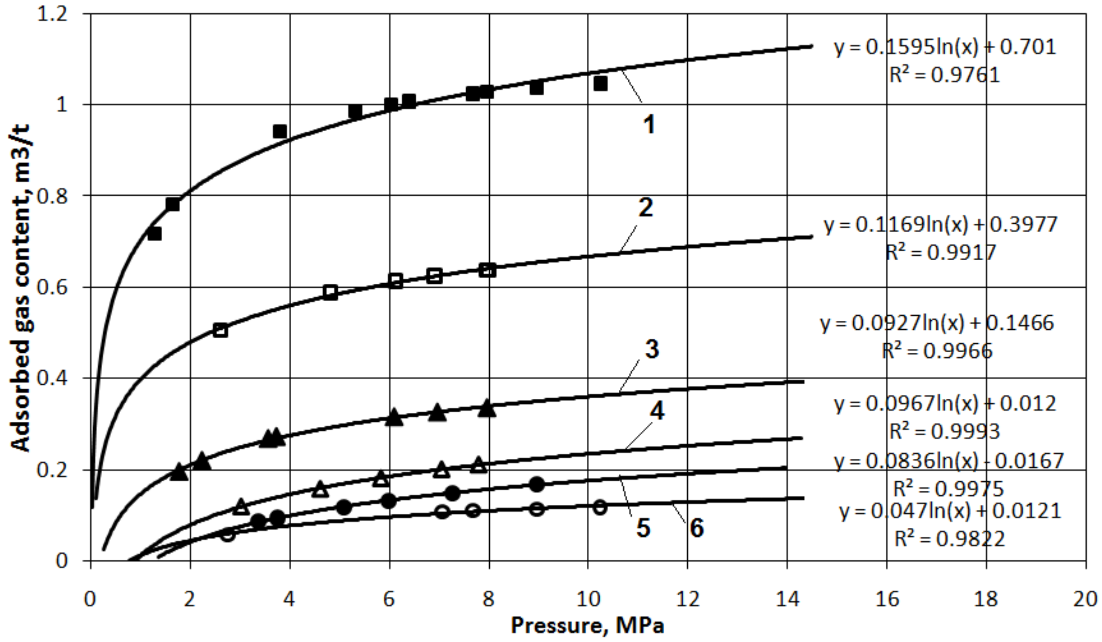

1, 2 – adsorption isotherms for the model with permeability of 9,7mD at temperature 40 °C and 60 °C respectively;
3, 4 – adsorption isotherms for the model with permeability of 29mD at temperature 40 °C and 60 °C respectively;
5, 6 – adsorption isotherms for the model with permeability of 93mD at temperature 40 °C and 60 °C respectively

Figure 6 – Graphical dependences of adsorbed gas content from pressure (adsorption isotherm) for models with different permeability at different temperatures

of adsorbed gas is reduced by about 2 times. This suggests that even in deep deposits with high reservoir temperatures adsorption processes occurs.

Figure 5 shows the dependence of the adsorbed gas content from the model permeability. These dependences are nonlinear. The highest correlation coefficient obtained when using a power function. However, even when the model permeability reach 100mD amount of adsorbed gas ranges 0,065-0,1m^3/t, and a further permeability increasing has virtually no effect on the adsorption capacity.

It is also worth noting that with permeability increasing the absolute dependence of the amount of adsorbed gas on temperature decreases. If for model 1 with permeability 9,7mD with increasing temperature from 40°C to 60°C the amount of adsorbed gas is reduced from 0,95 to 0,5 m^3/t (by 1,9 times), for model 3 the amount of adsorbed gas decreases from 0,1 to 0,065 m^3/t (by 1,5 times). Thus, with increasing permeability by 10 times the amount of adsorbed gas is reduced to about 7-8 times. In this regard, it can be concluded that natural gas is adsorbed on the surface of the pore space even in conventional highly permeable layers, but its amount is much less than in unconventional low-porous low-permeable reservoirs.

Figure 6 shows the dependence of the amount of adsorbed gas on the pressure for different temperatures and permeability of the model. Analyzing this dependence it should be noted that the amount of adsorbed gas increases with pressure rising. However, at high pressure values the amount of adsorbed gas does not increase with pressure growing up. This effect can be explained by the fact that all adsorption centers are occupied and further adsorption is not possible.

The results of experimental studies were subjected to statistical analysis in order to obtain empirical dependence, which would link together permeability, temperature, pressure and amount of adsorbed gas. However, it failed to achieve the required degree of accuracy in determining the amount of adsorbed gas using linear and nonlinear dependencies. For different dependences desired reproducibility was achieved only at a certain interval, while for the entire studied interval the coefficient of correlation ranged within 0.5-0.7, which testifies the non-linear of the process. Therefore, to obtain approximation dependencies of the above mentioned factors computerized neural networks of multilayer perceptron and radial basis function type were used. According to the analysis of the obtained results the highest coefficient of correlation was got for a model multilayer perceptron type MLP 3-7-1 (7 hidden neurons, in which there are three input values (permeability, temperature, pressure) and 1 output (the amount of adsorbed gas)), for which the coefficient of correlation for the test and control samples exceeds 0,999. The highest level of significance of input variables is permeability, and the lowest one - the pressure, which is consistent with experimental results. This model can be used for the rapid determination of the amount of adsorbed gas depending on the temperature and pressure conditions and reservoir permeability.

Conclusions

1. To remove the adsorbed gas just reservoir pressure lowering is not enough due to the nature of adsorption isotherms. Particularly at pressure decreasing by 8-10 times compared to initial reservoir pressure (Figure 6) only about 30-40% of the total amount of initially adsorbed gas is desorbed. And at considerable reservoir pressure reduction the further deposit development is not economically profitable.

2. In this situation it is necessary to implement other methods of gas desorption enhancement without reservoir pressure reduction. One of such methods is the use of non-hydrocarbon gases to replace the methane molecules on the surface of the pore space. Conducted studies have also shown that even in gas reservoirs with high permeability adsorption processes take place. Proof of this can serve field examples when at the final stage of the field development the reservoir pressure begins to rise due to gas desorption and the return of the well to production becomes expedient.

3. Therefore, the development and application of gas desorption intensification methods will enable to increase gas recovery factor not only from low-porous low-permeable reservoirs, but, also, from conventional gas fields, which in conditions of energy deficit is an extremely important issue.

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