https://doi.org/10.33271/nvngu/2024-4/018

K. A. Bezruchko*, orcid.org/0000-0002-3818-5624, O. V. Burchak, orcid.org/0000-0001-9114-8585, L. I. Pymonenko, orcid.org/0000-0002-5598-6722, V. V. Chelkan, orcid.org/0000-0002-0733-8739 Institute of Geotechnical Mechanics named by N. Poljakov of National Academy of Sciences of Ukraine, Dnipro, Ukraine * Corresponding author e-mail: gvrvg@meta.ua

SORPTION CAPACITY AND NATURAL GAS CONTENT OF COAL BEDS OF DONBAS

Purpose. To determine the general patterns in the formation of the sorption capacity of coal and the natural regional methane content of coal beds using a quantitative indicator - the relative gradient of gas content.

Methodology. To determine the sorption capacity of coal, the authors used the well-known "volumetric method" and the method of EPR-spectroscopy (electron paramagnetic resonance). We used the results obtained during geological exploration to analyze the natural gas content of coal beds. The determination of natural gas content was carried out using special gas-core-samplers and formation testers. Methods of mathematical statistics were used to process experimental data.

Findings. According to experimental data regarding the sorption methane-bearing capacity and natural gas content of Donbas coal beds, we carried out the analysis, statistical processing, and generalization of the obtained results. A regular change in the relative gradient of coal beds' methane content has been established for the entire Donbas as a whole. The values of relative gradients for coals of various grades naturally decrease with increasing stratification depth and also naturally decrease in each depth interval from low-metamorphosed (grade D) coal to highly-metamorphosed one. Based on the approximation results, the corresponding equations were obtained for each grade of coal metamorphism from gas to-anthracite (A).

Originality. New indicators have been proposed – the relative gradient of sorption methane-bearing capacity and the relative gradient of gas content, which allow comparison of these characteristics measured in absolute values $(cm^3/g, m^3/t)$ for individual coal beds of different grades of metamorphism, at different depths and lying in different geological conditions. It has been proven that the sorption capacity of coal matter determines the natural regional (background) methane content of coal beds, and naturally, according to a hyperbolic dependence, decreases with increasing stratification depth and also naturally decreases in each of the depth intervals from low-metamorphosed coal (grade D) to highly metamorphosed one (grade A), with a relative gradient that asymptotically approaches to 1 at pressures above 6 MPa.

Practical values. The obtained dependences of the relative gradient of gas content on depth and gas pressure for various grades of metamorphism can be used to predict the natural regional (background) gas content of coal beds by determining the maximum sorption capacity and calculating the desired depth or pressure.

Keywords: Donbas, coal beds, sorption capacity, gas content

Introduction. The need to solve the problem of mine methane in coal and gas fields defines the possibilities for the stable operation in the coal industry and consists of its removal from the subsoil to create safe conditions for coal mining, reduce the amount of harmful emissions of greenhouse gases into the atmosphere, and subsequent methane utilization as an energy and chemical raw material.

The closure of mines due to the access restriction to areas of fields that were previously developed, is associated with the closure of the drainage system, or the introduction of changes or restrictions on the scope of its operation. After all, such measures are linked with an increase in the water level and possible complete flooding of the mine [1].

Operating mines scheduled for final decommissioning should work with mines that were previously closed or are being closed. Closing a mine means decommissioning the drainage system and flooding its mine workings and surrounding rocks with a natural water influx [1]. Significant water reservoirs are formed in closed mines, which can be used as a carrier of thermal or mechanical energy. Mine gases accumulate in the above-water spaces of reservoirs created in mining workings. This is most often methane, which is pushed by the groundwater level to the Earth's surface [1]. Another powerful factor is added to the range of common ones of methane emissions associated with coal mining.

Methane is the main component of gases in coal fields, and since almost the entire coal-bearing strata of rocks are saturated with methane, the production of coal in coal mines, during their operation and after the completion of stopings, is constantly accompanied by methane emission. And, regardless of what extent the coal mining industry will work in the future, or if it works at all, the methane emission from the coal-bearing strata under underground water pressure will continue for decades. Given this, the problem of methane from coal and gas deposits will not lose its relevance in the future.

The results of scientific research and practical experience in the development of gas-saturated coal beds testify to the imperfection of the existing ideas regarding the formation and existence of the coal-gas system under normal natural conditions, and even more so under the impact of technogenic factors. First of all, this is due to the insufficient study of the processes that take place in the system under the impact of several factors – geomechanical, technogenic, and some others. The lack of the necessary adequate information reduces the accuracy in evaluating the properties and geomechanical state of the coal bed, as a result of which there is an increase in the volume and cost of measures necessary to increase the safety of mining operations, i. e. a significant increase in the price of energy production.

All the available results, obtained over a long time, testify to the uneven distribution of gases in the coal rock massif and a large number of deviations of the gas content and gas saturation in the coal-bearing strata from the general regional patterns, which in turn are also influenced by many local geological factors. The content and composition of gases in coal-bearing formations are primarily determined by such factors as the degree of metamorphism of coal beds and post-diagenetic transfor-

[©] Bezruchko K. A., Burchak O. V., Pymonenko L. I., Chelkan V. V., 2024

mations of host rocks, to which tectonics, coal content, the lithological composition of rocks, the thickness of covering deposits, hydrogeological conditions, immersion depth, and modern stratification depth are added. The degree of influence of each of the factors, except for metamorphism, is different in various gas zones, so the distribution of gases in the coal-bearing strata is characterized by diversity, as a result of the mutual influence and interaction between the above factors.

The influence of a large number of factors on the natural gas content of coal beds determines the difficulty of comparing the gas content measured in absolute values for individual coal beds, which differ in the degree of metamorphism, stratification depth, and various geological conditions. Separately, it should be noted that against the background of general trends that form the natural regional background gas content, various kinds of deviations caused by various anomalies are superimposed. These can be positive anomalies (increasing the gas content), which contribute to the improvement of gas content and form gas accumulations, and even cause gas-dynamic phenomena in coal mines. We also have negative anomalies, which cause a decrease in gas content through lateral migration or degassing up the rise of rocks, and subsequently from the Earth's surface into the atmosphere.

The possibility of a correct quantitative comparison can be solved with the help of a universal quantitative indicator, which is relative and makes it possible to compare the absolute values of the gas content of coal beds in different geological conditions. The use of such an indicator is aimed at elucidating the general patterns in the formation of the natural regional (background) methane content of coal beds.

The research of criteria allowing us to reliably describe the sorption process of coal from methane, as well as to quantitatively assess the gas content of coal beds, will provide an opportunity to solve several urgent tasks related to the prediction of mining and geological conditions and processes in a coal rock massif.

It is appropriate to note that since the article is devoted to the methane problem of coal fields, therefore, the term "gas content" in the text should be understood as "methane content".

The purpose of the paper is to define the general patterns in the formation of the sorption capacity of coal and the natural regional methane content in coal beds using a quantitative indicator — the relative gradient of gas content.

Theoretical part. Despite a large number of existing sorption theories, the question concerning the nature of the methane-coal interaction remains open, primarily because methane is genetically related to hard coal. In addition, due to the great heterogeneity of the coal matter, its polycomponentity, heterogeneity, and metastability, the sorption interaction in the coal-gas system must be considered in the context of the change in the state of the sorbent (coal).

Studying the distribution conditions of methane in the coal-bearing strata determines the possibilities for solving the problem of methane in coal fields [2]. The sorption properties of coal and the state of the coal-gas system are closely connected. They characterize the forms of occurrence and determine the parameters and amount of gases of coal-and-gas fields, namely: natural gas content, gas capacity, gas saturation, outburst hazard of coal beds, and others.

According to the requirements of regulatory documents in force in Ukraine, when determining the volume of coal gases while hard coal mining, it is obligatory to define the sorption capacity of the coal matter [3, 4].

An important basis for the simultaneous extraction of coal and gas is the understanding of the mechanism for extracting methane from coal beds and knowledge of the gas sorption characteristics of coal.

The increase in gas concentration at the interface of two phases – gaseous (sorbate) and solid (sorbent) – occurs under the action of intermolecular forces that are unbalanced on the surface of the sorbent. A field of sorption interaction is formed at the phase interface. It is believed that most of the gas in coal beds is in the adsorbed state.

Experimental study of gas sorption provides a scientific foundation for the practical assessment of methane emissions in coal beds (CBM) and deposition of CO_2 in the coal bed [5]. In particular, the adsorption characteristics of CH_4 , CO_2 and H_2O play an important role in predicting CBM yield and geological sequestration potential of CO_2 in the research fields of CO_2 -rich methane extraction and CO_2 sequestration [6].

The research on gas sorption properties of coal is conducted in all coal-mining basins of the world, in particular in the USA, Canada [7], China [8, 9], Australia [7, 10], India [5], Indonesia [11], South Africa [12], Poland [13, 14], and other countries.

It has been established that the adsorption capacity of gas in coal beds is determined by the degree of coal metamorphism, the chemical composition of the adsorbent, the structure of the pore space (pore size distribution), and the porosity value, as well as pressure, temperature, and moisture [8]. The volume of micropores correlates with the adsorption capacity of coal – micropores have the largest specific surface area, which is the main regulator of the volume of adsorbed gas [9].

The paper [5] also notes that the stratification depth, the molecular size of gases, the affinity of gas to coal, density, porosity, grade of coal, etc. are the main factors determining the adsorption capacity of coal. The paper [7] highlights that the most important factors affecting the sorption capacity are also the type and content of mineral substances (ash content), while the maceral composition of the coal matter is of limited importance.

Some researchers note precisely the leading role of the degree of carbonization in the coal substance on its sorption capacity. The dominant impact of the degree of coalification on the sorption capacity in work [15] was estimated at 89%, in comparison with other factors. The leading role of metamorphism, in work [16], using the multiple regression model, was recorded in 86 % of cases.

Thus, theoretical and experimental research suggests that the ability of coal to absorb methane mainly depends on 3 main factors: the physicochemical properties of coal; the pressure under which the coal is located; and sorbent temperature. So far, the following three main provisions have been quite accurately defined, which quantitatively characterize the sorption capacity of coal:

- the sorption capacity in the metamorphic series of coal from long-flame coal to anthracite, characterized by various physical and chemical properties, continuously increases, but does not exceed 35-40 cm³ in 1 g of coal matter;

- with an increase in gas pressure to 5-6 MPa, the quantity of sorbed gas grows. The saturation limit practically occurs at these pressures, and a further increase in pressure causes only a slight increase in sorption (the increase in sorption in the pressure range of 5-10 MPa does not exceed 5-10 %);

An increase in temperature reduces the sorption capacity of coal matter. It has been proven that as the temperature increases from 0 to 42 °C, the amount of sorbed methane decreases by 68 %. Obviously, the sorption process stops at temperatures of 150–200 °C.

It is believed that methane can be in a free, sorbed state or in the form of a solid solution in a coal rock massif. That is, there are three forms of gas-solid phase interaction: adsorption (intermolecular interaction), absorption (penetration of gas molecules into a solid without chemical interaction with the formation of a solid solution), and chemisorption (chemical interaction between a gas and a substance).

The natural gas content of coal beds depends mainly on the sorption capacity of the coal matter. Gas in coal beds is mainly in a bound, sorbed state, which is due to the high sorption capacity of coal.

The fraction of free gas in coal beds is small and, according to most researchers, does not exceed 5-15 %. The main volume of gas is in clathrate or sorbed forms, although views on the ratio of various forms are not constant. Some believe that

the sorbed form contains up to 40% methane, chemically unbound methane of solid solutions (pseudo clathrate forms) — from 5 to 20–30 %, and molecularly bound methane -5-10 % of the total amount of physically bound methane in coal is 60–70 %. According to other researchers, methane in coal is contained mainly in the form of a solid solution. Most authors, as emphasized above, think that the fraction of free in coal beds is insignificant and does not exceed 5–15 %. The natural gas content of coal varies widely – from several hundred to 35–40 m³/t, according to some data, up to 35–45 m³/t [17].

For a long time, the main attention of researchers of gases in coal fields was focused mainly on the study of the distribution of gases in coal, so the patterns of methane distribution in coal beds are more researched. The most well-known and generally accepted is the dependence of the methane content of a coal bed on the degree of coal metamorphism.

The essence of this dependence is a natural increase in the methane content of coal beds with an increase in the degree of metamorphism from 1D to 10-11A. The maximum methane content corresponds to the yield of volatile substances of about 7 %. After that, its sharp decrease to zero values is observed in anthracites of groups 11-13A.

The stratification depth of coal beds is another factor whose effect on the gas content of the coal beds is proven. For a long time, the opinion of specialists was the only one: with increasing depth, the methane content only increases. Such unanimity was probably due to the availability of actual data that came from small depths of coal bed development. The maps of both natural methane content and forecast maps, built based on geological survey data, were systems of parallel isolines, in general, repeating the hypsometry of coal beds. That is, they were a function of the current coal depth. At that time, there were only disagreements in assessing the nature of the increase in gas content. As the depth of development increased, the number of direct determinations of methane content during geological exploration works increased, the data on the determination of methane content in mine workings accumulated, and ideas about changes in the methane content of beds at great depths were corrected.

Currently, there are three viewpoints on this issue. Most researchers believe that at a depth of 800 m, the increase in methane content has practically stopped and the methane content has practically stabilized. Some believe that it is not methane content that has stabilized as a whole, but its growth. There is also a viewpoint that below a certain depth the methane content should begin to decrease. There is information that the gas content of coal increases with depth, then stabilizes, and with an increase in depth of more than 1,000 m it begins to decrease [17]. This can be explained by a decrease in the sorption capacity of coal as the temperature increases.

The analysis of actual data shows that below the zone of gas weathering, there is an intensive increase in the methane content of coal beds, after which the gradient of its growth decreases sharply. There is a conclusion that the gas content of coal beds below the zone of gas weathering increases, after which it stabilizes. It is also believed that an increase in the stratification depth leads to a corresponding constant increase in the methane content due to an increase in the quantity of free-phase gas in the coal. According to another opinion, the change in the methane content of coal beds corresponds to a hyperbolic dependence. That is, at first the methane content of its growth begins to decrease, after which the methane content stabilizes at depths of up to 1.700 m.

Later, they concluded that the stabilization of the methane content should be attributed to the depth interval of 800–900 m, i.e., greater than other authors previously considered. Further, based on the dependences obtained from literature data, they prove not the stabilization of methane content at great depths, but the achievement of maximum values, after which it decreases [17]. Without overestimating the importance of statistical processing of the actual data and being aware of the conditionality of any extrapolations, it is emphasized that the further decrease in methane content with depth is not affirmative, but rather permissible. The boundary for starting this extrapolation is indicated by a depth of 1,200 m, as the greatest depth at which, at that time, the maximum methane content of coal was measured. Finally, the question of the depth impact on gas content still remains unexplained.

If we assume that the natural gas content of coal is mainly defined by its sorption capacity, and the sorption capacity reaches a maximum and stabilizes at pressures of 6 MPa, then based on the conclusion regarding the maximum value of the sorption capacity of coal, which is reached at these pressures (over 6 MPa), one should expect a change in the background regional natural gas content according to a hyperbolic dependence, reaching an asymptote almost precisely at these pressures. And based on the fact that the gas pressure of a coal rock massif is known to be 0.8-0.9 hydrostatic [17], this should occur at depths of about 660–750 m. This coincides well with numerous data on the practical measurement of natural gas content – stabilization of methane content is assigned to the depth interval of 800–900 m.

Another important factor that determines the gas content of a coal-bearing strata (both coal beds and host rocks), especially at the local level, is the tectonic factor, namely: the presence of disturbances, their type, amplitude, and angle of incidence, orientation in space [17]. It is worth noting that we are talking about both disjunctive and plicative disturbances. At the same time, disturbances play a dual role in the redistribution of gases in the coal-bearing strata: on the one hand, they can be gas traps that form methane accumulation zones and, on the other hand, be channels that contribute to degassing.

It is believed that if the ruptures are formed under conditions of stretching of the earth's crust (mainly faults), then they increase the permeability of coal-bearing sediments in the zone adjacent to the disturbance and generally contribute to the degassing of the rock massif. Raptures formed under conditions of compression (thrusts and upthrow faults) mainly hinder degassing, they are screens, and create conditions for the formation of methane accumulations. Also, the orientation of the raptures relative to the bedding of rocks can be significant. It is believed that raptures oriented along in the crossstrike direction of rocks reduce the gas content of coal-bearing sediments, and if the direction of the rapture coincides with the strike of rocks, then, in this case, the rapture plays as a screen and contributes to the methane accumulation. As for the role of diagonal raptures, the question remains debatable. Diagonal raptures in some cases play a degassing role; in others, on the contrary, they contribute to the formation of methane accumulations [17].

The hydrogeological regime also affects the methane content of coal beds. Active circulation of groundwater contributes to the degassing of the coal-bearing strata as a whole. That is, the presence of well-permeable sandstones in the section of aquifers determines the release and migration of methane in the zone of active water exchange, reducing the gas content. On the other hand, coal beds associated with hydrous rocks in the stagnant regime zone have a more significant methane content [17].

It is worth noting that the last two factors – tectonics and hydrogeological regime – define local deviations (both positive and negative) of the quantitative content of gas in the coalbearing strata from the regional (background gas content of coal beds). And the main task of this research is to define the patterns in forming exactly the natural regional (background gas capacity).

Thus, summing up, one can conclude that both the sorption properties of coal matter and the gas content of coal beds at the regional level are generally determined by common factors and are characterized by general patterns

Methodology. The authors applied the well-known "volumetric method" and the EPR-spectroscopy (electron paramagnetic resonance) method to determine the sorption capacity of coal. We used the results obtained in the course of geological exploration for the analysis of the natural gas content in coal beds. Determination of natural gas content was carried out with the help of special gas core samplers and formation testers. The methods of mathematical statistics were used to process the experimental data.

The object of research. Beds of all grades of hard coal were studied – from long-flame coal to anthracite within the Donets coal basin.

Results. Following the experimental data regarding the sorption methane-bearing capacity and natural gas content in the coal beds of Donbas, we carried out the analysis, statistical processing, and generalization of the obtained results.

Table 1 shows the gradients of the sorption methane-bearing capacity of coal beds, calculated on the base of data on the sorption methane-bearing capacity of coal in the Pavlohrad-Petropavlisk district in the pressure range of 1-5 MPa. The gradients of the sorption methane-bearing capacity of coal beds are the difference between the next (for higher pressure) and the previous (for lower pressure) values of the sorption methane-bearing capacity in increments of 1 MPa. By its essence, it is an indicator of the change in the sorption methanebearing capacity measured in absolute units $-m^3 per 1$ ton of

Table 1

Gradients of sorption methane-bearing capacity of coal in the Pavlohrad-Petropavlivsk district in the pressure range of 0.1-5 MPa at a temperature of 20 °C

Area/bad	Gradients of sorption methane-bearing capacity of coal at different pressures, m ³ /t d.a.m.				
Alea/deu	0.1–1.0 MPa	1.0-2.0 MPa	2.0-3.0 MPa	3.0-4.0 MPa	4.0–5.0 MPa
Brahynivska/ $c_6^1 - c_6$	$\frac{3.10-8.40}{5.30}$	<u>8.40–10.60</u> 2.20	<u>10.60–12.30</u> 1.70	<u>12.30–13.10</u> 0.80	<u>13.10–13.70</u> 0.60
<i>c</i> ₅	$\frac{3.50-8.50}{3.00}$	<u>8.50–11.40</u> 2.90	<u>11.40–13.00</u> 1.60	<u>13.00–14.00</u> 1.00	$\frac{14.00-14.40}{0.40}$
<i>c</i> ₂	<u>2.40–9.60</u> 7.20	<u>9.60–12.60</u> 3.00	<u>12.60–14.50</u> 1.90	<u>14.50–15.60</u> 1.10	<u>15.60–16.60</u> 1.00
c_2^1	<u>2.30–7.30</u> 5.00	$\frac{7.30-10.70}{3.40}$	$\frac{10.70-12.70}{2.00}$	<u>12.70–13.80</u> 1.10	<u>13.80–14.60</u> 0.80
$c_2 - c_2^1$	$\frac{2.30-7.80}{2.50}$	$\frac{7.80-11.20}{3.40}$	<u>11.20–13.10</u> 1.90	<u>13.10–14.30</u> 1.20	<u>14.30–15.10</u> 0.80
Petropavlivska-Hlyboka/ c_{10}^b	$\frac{2.90-8.10}{5.20}$	<u>8.10–10.70</u> 2.60	<u>10.70–12.30</u> 1.60	<u>12.30–13.20</u> 0.90	<u>13.20–13.60</u> 0.40
c_8^b	<u>3.00–9.10</u> 6.10	<u>9.10–11.00</u> 1.90	<u>11.00–14.00</u> 3.00	<u>14.00–15.50</u> 1.50	<u>15.50–16.70</u> 1.20
c_5^b	$\frac{1.40-9.80}{8.40}$	<u>9.80–12.50</u> 2.70	$\frac{12.50-14.20}{1.70}$	<u>14.20–15.20</u> 1.00	<u>15.20–16.20</u> 1.00
<i>c</i> ₂	<u>2.40–9.60</u> 7.20	<u>9.60–12.60</u> 3.00	<u>12.60–14.50</u> 1.90	<u>14.50–15.60</u> 1.10	<u>15.60–16.70</u> 1.10
<i>c</i> ₁	<u>2.10–9.10</u> 7.00	<u>9.10–11.20</u> 2.10	<u>11.20–13.10</u> 1.90	<u>13.10–14.40</u> 1.30	$\frac{14.40-15.00}{0.60}$
Svydivska $/c_8^u$, c_8^l	$\frac{3.00-8.40}{5.40}$	<u>8.40–11.70</u> 3.30	<u>11.70–14.00</u> 2.30	<u>14.00–15.10</u> 1.10	<u>15.10–16.20</u> 1.10
C ^u ₇	<u>3.00–9.00</u> 6.00	<u>9.00–13.00</u> 4.00	$\frac{13.00-15.40}{2.40}$	<u>15.40–17.30</u> 1.90	<u>17.30–18.50</u> 1.20
c_6^u	$\frac{3.00-8.60}{5.60}$	<u>8.60–11.80</u> 3.20	<u>11.80–13.60</u> 1.80	<u>13.60–15.40</u> 1.80	<u>15.40–16.20</u> 0.80
<i>c</i> ₅	$\frac{2.90-8.90}{6.00}$	<u>8.90–12.20</u> 3.30	$\frac{12.20-14.50}{2.30}$	<u>14.50–16.10</u> 1.60	<u>16.10–17.20</u> 1.10
c_2^u	<u>3.10–6.20</u> 3.10	<u>6.20–11.00</u> 4.80	$\frac{11.00-14.50}{3.50}$	<u>14.50–15.20</u> 0.70	<u>15.20–16.40</u> 1.20
<i>c</i> ₁	$\frac{2.90-8.50}{5.60}$	<u>8.50–12.00</u> 3.50	$\frac{12.00-14.50}{2.50}$	$\frac{14.50-15.90}{1.40}$	<u>15.90–17.00</u> 1.10
Mine "Yuvileina"/ c_6	$\frac{2.34-6.60}{4.26}$	<u>6.60–8.55</u> 1.95	<u>8.55–9.49</u> 0.94	<u>9.49–9.98</u> 0.49	<u>9.98–10.06</u> 0.08
c_2^1	<u>3.27–7.91</u> 4.64	<u>7.91–10.23</u> 2.32	<u>10.23–11.83</u> 1.60	<u>11.83–13.26</u> 1.43	<u>13.26–13.75</u> 0.49
C2	<u>2.60–8.58</u> 5.98	<u>8.58–10.90</u> 2.32	<u>10.90–12.15</u> 1.25	<u>12.15–12.71</u> 0.56	<u>12.71–13.75</u> 1.04
c_2^{l}	<u>3.56–8.66</u> 5.10	<u>8.66–10.88</u> 2.22	<u>10.88–11.84</u> 0.96	<u>11.84–12.44</u> 0.60	<u>12.44–13.02</u> 0.58
Average	5.43	2.90	1.94	1.13	0.83

Gradients of sorption methane-bearing capacity of coal
in the Pokrovsk district in the pressure interval of 1-5 MPa
at a temperature of 20 °C

	Gradients of sorption methane-				
Mine/bed	bearing capacity of coal at different				
Willie/ bed	pressures, m ³ /t d.a.m.				
	1-2 MPa	2-3 MPa	3-5 MPa		
Rodynska/ l_8^1	3.3	2.3	3.6		
l_7^l	3.3	2.1	2.4		
Chervonolymanska/l ₇	3.7	1.9	2.4		
<i>l</i> ₃	3.3	0.7	1.8		
k_5	3.0	1.9	2.9		
Kapitalna/l ₇	3.9	2.5	3.0		
l_3	2.2	1.3	1.7		
l_1	2.5	1.9	1.8		
k_3^b	2.3	1.4	2.4		
Tsentralna/l ₇	3.7	2.1	1.5		
l_3	2.6	1.4	1.5		
l_1	2.8	1.7	2.4		
Mine 5/6					
l_7	3.4	2.2	2.6		
	2.7	1.4	1.8		
k_3^u	2.4	1.4	1.6		
Named after	2.8	1.5	1.7		
T. H. Shevchenko/ f_1					
Average	3.0	1.7	2.2		

dry ash-free mass. It can be seen that for each of the pressure intervals, the gradients differ from each other and on average decrease from 5.43 (0.1–1 MPa) to 0.83 (4–5 MPa) m^3/t d.a.m. (dry ash-free mass).

Table 2 shows gradients of the sorption methane-bearing capacity of coal beds, calculated based on data on the sorption methane-bearing capacity of coal beds in the Pokrovsk district in the pressure range of 1-5 MPa. In this case, the gradients on average decrease from 3.0 (1–2 MPa) to 2.2 (3–5 MPa).

As might be expected, judging by the shape of the sorption methane-bearing capacity isotherms, which have the form of hyperbolas, the gradients of the sorption methane-bearing capacity of coal beds naturally decrease with increasing pressure. The values of the gradients of the sorption methane-bearing capacity in the corresponding pressure intervals for the coal of Pavlohrad-Petropavlivsk District and Pokrovsk District are quite close.

Similarly, the gradient of the natural methane content of coal beds is also changing, as determined from geological exploration data. Table 3 shows the results of the research on the natural methane content of coal beds at several mines in the Donets basin (mostly of the medium and high degree of metamorphism). According to these data, the methane content decreases on average from 2.3 to 0.6 m³/t d.a.m. in the depth range of 100–500 m.

Table 4 shows the data in general for the entire Donets Basin regarding coal beds of hard coal from low-metamorphosed (Grade G) to anthracite at various depths. The gradient of methane content decreases in the depth interval of 100 to 500 m from 3.8 to 0.5 m³/t d.a.m. The gradients of gas content

Table 3

Gradients of the natural methane content of coal beds for different depths in the areas of mines* in the Donets basin

Table 2

Minefields	Gradients of sorption methane content of coal at different depths, m ³ /t d.a.m.				
Willeneids	100–200 m	200–300 m	300–400 m	400–500 m	500-600 m
Krasnodonski (A)	20.0-23.0	23.0-24.4	24.4-25.5	25.5-26.0	26.0-26.6
	3.0	1.4	1.1	0.5	0.6
Bystrianski (PS)	<u>11.7–14.7</u>	<u>14.7–16.6</u>	<u>16.6–17.8</u>	<u>17.8–18.8</u>	<u>18.8–19.5</u>
	3.0	1.9	1.2	1.0	0.7
No. 5	12.2-13.7	<u>13.7–15.3</u>	<u>15.3–16.5</u>	<u>16.5–17.5</u>	<u>17.5–18.3</u>
Chornukhynska,	1.5	1.6	1.2	1.0	0.8
Named after S.V. Kosior (P)					
Nikanor,	<u>13.1–15.0</u>	<u>15.0–15.8</u>	<u>15.8–16.0</u>	<u>16.0–16.3</u>	<u>16.3–16.6</u>
Komysarivska (P)	1.9	0.8	0.2	0.3	0.3
Yasynivska-Hlyboka (K, P, PS)	<u>10.9–13.0</u>	<u>13.0–14.0</u>	14.0-14.5	14.5-15.0	<u>15.0–15.5</u>
	2.1	1.0	1.0	0.5	0.5
Average	2.3	1.3	0.8	0.7	0.6

* The names of the mines are given at the time of sampling and experimental work

Table 4

Gradients of the methane content of coal beds (grades G-A) at different depths

Cool grada	Gradients of the methane content of coal beds of different grades for the individual depth intervals, m ³ /t d.a.m.					
Coargrade	100-200 m	200–300 m	300–400 m	400–500 m	500-600 m	
G	<u>1.9–4.8</u>	4.8-6.7	<u>6.7–7.5</u>	7.5-8.0	8.0-8.5	
	2.9	1.9	0.8	0.5	0.5	
Zh	4.8-8.9	<u>8.9–11.0</u>	<u>11.0-12.0</u>	12.0-12.8	12.8-13.3	
	4.1	2.1	1.0	0.8	0.5	
K	<u>6.3–9.5</u>	<u>9.4–11.5</u>	<u>11.5–12.6</u>	12.6-13.4	13.4-14.0	
	3.2	2.1	1.1	0.8	0.6	
OS	<u>7.3–11.1</u>	<u>11.1–13.5</u>	13.5-14.9	14.9-15.7	<u>15.7–16.3</u>	
	3.8	2.4	1.4	0.8	0.6	
Т	9.6-14.3	14.3-16.4	<u>16.4–17.5</u>	<u>17.5–18.2</u>	18.2-18.6	
	4.7	2.1	1.1	0.7	0.4	
A	8.5-12.3	12.3-14.7	14.7-16.1	16.1-17.0	17.0-17.6	
	3.8	2.4	1.4	0.9	0.6	
Average	3.8	2.2	1.1	0.8	0.5	

are greater at shallow depths and greater for less metamorphosed coal.

To solve the main task, namely, to determine the general patterns in the formation of the values of sorption capacity and natural methane content of coal by comparing individual coal beds. beds of different grades of metamorphism and at different depths, and lying in various geological conditions, it is proposed to use the indicator – the relative gradient of sorption capacity and relative gradient of gas content.

The relative gradient of the sorption methane-bearing capacity of coal beds is the ratio of the next (for a higher pressure) value of the sorption methane-bearing capacity to the previous one (for a lower pressure) in increments of 1 MPa. Similarly, the relative gradient of gas content of coal beds is the ratio of the next value of gas content to the previous one with the same step (1 MPa). The relative gradient of sorption methane-bearing capacity (gas content) is essentially a reduction rate of sorption capacity (gas content) with depth and, accordingly, with increase in pressure, and is a dimensionless quantity.

Table 5 shows the data on the relative gradients of the sorption methane-bearing capacity of coal in several beds (grade G) of the Pavlohradsk-Petropavlivsk district in the pressure range of 0.1-5 MPa at a temperature of 20 °C, calculated according to the data in Table 1. Average values of relative gradients naturally decrease from 3.22 (0.1-1 MPa) to 1.06 (4-5 MPa). In a

Table 5

Relative gradients of sorption methane-bearing capacity of coal beds in the Pavlohradsk-Petropavlivsk district in the pressure range of 0.1-5 MPa at a temperature of 20 °C

	Relative gradients of sorption methane-bearing capacity of coal at different pressure, d/l (dimensionless)				
Area/bed	0.1–1MPa	1–2 MPa	2-3 MPa	3-4 MPa	4-5 Mpa
Brahynivska/ $c_6^1 - c_6$	2.70	1.26	1.16	1.07	1.05
<i>c</i> ₅	2.43	1.34	1.14	1.08	1.03
<i>c</i> ₂	4.00	1.31	1.15	1.08	1.06
c_2^1	3.17	1.47	1.19	1.09	1.06
$c_2 - c_2^1$	3.39	1.44	1.17	1.09	1.06
Petropavlivska-Hlyboka/ c_{10}^b	2.79	1.32	1.15	1.07	1.03
c_8^b	3.03	1.21	1.27	1.11	1.08
c_5^b	7.00	1.28	1.14	1.07	1.07
<i>c</i> ₂	4.00	1.31	1.15	1.08	1.07
<i>C</i> ₁	4.33	1.23	1.17	1.10	1.04
Svydivska/ $c_8^u \cdot c_8^l$	2.80	1.39	1.20	1.08	1.07
c_7^b	3.00	1.44	1.18	1.12	1.07
c_6^b	2.87	1.37	1.15	1.13	1.05
c_5^b	3.07	1.37	1.19	1.11	1.07
c_2^b	2.00	1.77	1.32	1.05	1.08
<i>C</i> ₁	2.93	1.41	1.21	1.10	1.07
Mine "Yuvileina"/ c_6	2.82	1.30	1.11	1.05	1.01
	2.42	1.29	1.16	1.12	1.04
c ₂	3.30	1.27	1.11	1.05	1.08
c_2^1	2.43	1.26	1.09	1.05	1.05
Average by beds	3.22	1.35	1.17	1.09	1.06

Table 6

Relative gradients of natural methane content of coal beds on individual areas of mines in the Donets basin

Minefields	Relative gradients of the methane content of coal beds for the individual depth intervals, d/l				
(coal grade)	100–200 m	200–300 m	300–400 m	400–500 m	500–600 m
Krasnodonski (A)	1.15	1.06	1.05	1.02	1.02
Bystrianski (PS)	1.26	1.13	1.07	1.06	1.04
No. 5 Chornukhynska, Named after S. V. Kosior (P)	1.12	1.12	1.08	1.06	1.05
Nikanor, Komysarivska (P)	1.15	1.05	1.01	1.02	1.02
Yasynivska-Hlyboka (K, P, PS)	1.19	1.08	1.04	1.03	1.03
Average	1.17	1.09	1.05	1.04	1.03

* The names of the mines are given at the time of sampling and experimental work

The relative gradients of methane content of coal beds of different grades for individual depth intervals. d/l Coal grade 300-400 m 100-200 m 200-300 m 400-500 m 500-600 m G 2.53 1.40 1.12 1.07 1.06 Zh 1.85 1.09 1.07 1.04 1.24 Κ 1.51 1.22 1.10 1.06 1.04 PS 1.52 1.22 1.10 1.05 1.04 Р 1.04 1.02 1.49 1.15 1.07 1.45 1.20 1.10 1.06 1.04 А 1.04 Average 1.73 1.24 1.10 1.06





Fig. 1. Dependence of the average relative gradient of methane content on the occurrence depth of the coal bed



Fig. 2. Dependence of the average relative gradient of methane content on gas pressure

similar way, the values of the relative gradient of the natural methane content of coal beds in individual sections of the mines in the Donets basin change (Table 6), which have been calculated on the same areas, according to the data in Table 3



Fig. 3. Dependence of the relative gradient of methane content on the grade of coal for different depth intervals



Fig. 4. Dependence of the relative gradient of methane content on the occurrence depth for coal of different grades

and according to the grades of coal (Table 7 - calculations according to the data of Table 4).

The values of the relative gradient of methane content for coal beds of different grades naturally decrease with increasing the occurrence depth and also naturally decrease in each of the depth intervals from low-metamorphosed (grade D) to highly metamorphosed (grade A) coal. This is well illustrated by the graphs in Figs. 1–2. These are the dependences of the average relative gradient of methane content on the occurrence depth of the coal bed and the average relative gradient of methane content on gas pressure.

Fig. 3 shows the dependence of the relative gradient of methane content on the coal grade f or different depth intervals. Fig. 4 demonstrates the dependence of the relative gradi-

Table 8

The equation for the dependence of the relative gradients of methane content for coal beds of different degrees of metamorphism

Coal grade	Equation	Coefficient of determination, <i>R</i> ²
G	$y = -7 \cdot 10^{-8}x^3 + 8.992 \cdot 10^{-5}x^2 - 0.03813125x + 6.47970536$	0.9983
Zh	$y = -4 \cdot 10^{-8}x^3 + 5.08 \cdot 10^{-5}x^2 - 0.02130208x + 4.04015179$	0.9988
K	$y = -1 \cdot 10^{-8}x^3 + 1.68 \cdot 10^{-5}x^2 - 0.00836875x + 2.41211607$	0.9999
PS	$y = -1 \cdot 10^{-8}x^3 + 1.61 \cdot 10^{-5}x^2 - 0.00827083x + 2.43776786$	0.9994
Р	$y = -2 \cdot 10^{-8}x^3 + 2.637 \cdot 10^{-5}x^2 - 0.01144792x + 2.67199107$	0.9983
А	$y = -1 \cdot 10^{-8}x^3 + 1.464 \cdot 10^{-5}x^2 - 0.00717292x + 2.2222679$	0.9996

ent of methane content on the occurrence depth for coal of different grades. The graphs are built according to the data in Table 7 and are common for the coal beds of the entire Donbas.

In the latter case (Fig. 4), it is a bundle of curves that gradually converge and, subsequently, asymptotically approach 1. Thanks to the proposed indicator (relative gradient of methane content), it is possible to extrapolate the obtained dependences in the direction of higher pressures above 6 MPa, and respectively, greater depths.

Conspicuous is the fact that at greater depths the values of relative gradients for coal beds of all grades are leveled off (Table 7, Fig. 3). And at depths of 300-400 m they are 1.07-1.12, at depths of 400-500 m - 1.04-1.07, at depths of 500-600 m - 1.02-1.06. That is, the possible increase in gas content, due to an increase in depth, and, accordingly, pressure, can potentially amount to 2-6% if the value of the relative gradient is equal to 1. This is completely consistent with the above data when the increase in sorption in the pressure range of 5-10 MPa does not exceed 5-10%.

It is worth emphasizing that we are talking about general regional patterns of changes in methane content, which, as mentioned above, can be and are superimposed by local deviations, including anomalous ones, for example, such as sudden emissions of coal and gas in the mine workings. Anomalies can be different in nature. They can occur as a result of slow or sudden gas generation, or the emission of previously formed free methane. The obtained dependences are advisable to use for predicting the background natural regional gas content of coal beds.

Instead of expensive and time-consuming geological exploration fieldwork, the gas content of coal beds at different depths can be determined by the maximum sorption capacity of coal samples, in particular by the EPR method. This can be done by determining the limit sorption capacity of the coal and making a correction to the required depth (or pressure) using the appropriate relative gradient of the gas content for a particular grade.

Knowing the limit sorption capacity of hard coal, it is possible to calculate the value of methane content for different pressures based on the obtained dependencies for a separate grade of metamorphism. By correcting the actual gas pressure (0.8–0.9 hydrostatic) for a specific depth, it is possible to predict the natural gas content for that depth. Such calculations require the use of inverse formulas, in which the function is the relative gradient of gas content, and the argument is either gas pressure or depth. Although it is advisable to note that, according to the physical essence, the relative gradient of gas content depends on the depth and, accordingly, gas pressure, and not vice versa. That is why the graphs in Figs. 3–4 are built in exactly this way.

Table 8 shows the equations of the dependence of the relative gradients of methane content for coal beds of different degrees of metamorphism based on the results of approximation with the corresponding reliability coefficients.

Conclusions. The authors propose new indicators – the relative gradient of the sorption methane-bearing capacity and

the relative gradient of the methane content, which allows the comparison of these characteristics. measured in absolute values $(cm^3/g, m^3/t)$ for individual coal beds of different grades of metamorphism, at different occurrence depths and lying in various geological conditions

A regular change in the relative gradient of the methane content of coal beds for the entire Donbas as a whole has been established. The values of the relative gradients for coal of different grades naturally decrease with increasing occurrence depth and also naturally decrease in each of the depth intervals from low-metamorphosed (D grade) coal to highly-metamorphosed (A grade). This proves that the sorption capacity of the coal matter determines the natural regional (background) methane content of hard coal beds and naturally, according to a hyperbolic dependence decreases with increasing stratification depth and also decreases regularly in each of the depth intervals from low-metamorphosed (grade D) coal to highly metamorphosed (grade A), with a relative gradient that asymptotically approaches 1 at pressures above more than 6 MPa.

The obtained dependences of the relative gradient of gas content on depth and gas pressure for different grades of metamorphism can be used to predict the natural regional (background) gas content of coal beds by determining the limit sorption capacity and calculating the desired depth or pressure.

References.

1. Bukowska, M., & Bukowski, P. (2023). Investigation of Geomechanical Properties of Carboniferous Rocks for Evaluating the Possibility of Energetic Use of Water and Methane from Hard Coal Mines. *Archives of Mining Sciences*, *68*(2), 207-225. <u>https://doi.org/10.24425/</u> ams.2023.146176.

2. Zhang, L., Ye, Z., Li, M., Zhang, C., Bai, Q., & Wang, C. (2018). The binary gas sorption in the bituminous coal of the Huaibei Coalfield in China. *Adsorption Science & Technology*, *36*(9-10), 1612-1628. https://doi.org/10.1177/0263617418798125.

3. SOU 10.1.00174088.011-2005 Rules for conducting mining operations in strata prone to gas-dynamic phenomena (2005). Kyiv: Minvuhleprom Ukrainy. Retrieved from <u>https://issuu.com/mitc2/docs/026</u>.

4. Rudko, H. I., Bulat, A. F., & Kuznetsova, L. D. (2015). *Methodological recommendations for the geological study on the gas-bearing capacity of coal beds and host rocks for the calculation of reserves and the assessment of gas (methane) resources of underground coal deposits,* (pp. 106-112). Kyiv: Institute of Geotechnical Mechanics named by N. Poljakov of National Academy of Sciences of Ukraine. Retrieved from https:// www.nas.gov.ua/EN/Book/Pages/default.aspx?BookID=0000009846.

5. Harinandan, K., Mishra, M. K., & Mishra, S. (2019). Sorption capacity of Indian coal and its variation with rank parameters. *Journal of Petroleum Exploration and Production Technology*, *9*, 2175-2184. https://doi.org/10.1007/s13202-019-0621-1.

6. Wen, Z., Yang, Y., Wang, Q., & Yao, B. (2021). Mechanism and characteristics of $CH_4/CO_2/H_2O$ adsorption in lignite molecules. *Geofluids*, 2021. https://doi.org/10.1155/2021/5535321.

7. Perera, M.S.A., Ronjith, P.G., Choi, S.K., Airey, D., & Weniger, P. (2012). Estimation of Gas Adsorption Capacity in Coal: A Review and an Analytical Study. *International Journal of Coal Preparation and Utilization*, *32*(1), 25-55. <u>https://doi org/10.1080/19392699.2011.</u> <u>614298</u>. **8.** Gao, D., Hong, L., Jiren Wang, J., & Zheng, D. (2019). Adsorption simulation of methane on coals with different metamorphic grades. *AIP Advances*, *9*, 095108. https://doi.org/10.1063/T5115457.

9. Kui Dong, Zhiwei Zhai, & Aijun Guo (2021). Effects of Pore Parameters and Functional Groups in Coal on CO_2/CH_4 Adsorption. *ACS Omega, 6*, 32395-32407. Retrieved from <u>http://pubs.acs.org/journal/acsodf</u>.

10. Ekundayo, J. M., & Rezaee, R. (2019). Volumetric measurements of methane-coal adsorption and desorption isotherms – effects of equations of state and implication for initial gas reserves. *Energies*, *12.* https://doi.org/10.3390/en12102022.

11. Raharjo, S., Bahagiarti, S., Purwanto, H.S., & Rahmad, B. (2018). The effect of coal petrology on the capacity of gas methane absorption in coal formation Tanjung Barito in Binuang Region. *South Kalimantan. Series: Earth and Environmental Science, 212*(2018), 012029. <u>https://doi.org/10.1088/1755-1315/212/1/012029</u>.

12. Okolo, G.N., Everson, R.C., Neomagus, H.W.J.P., Sakurovs, R., Grigore, M., & Bunt, J.R. (2019). Dataset on the carbon dioxide. methane and nitrogen high-pressure sorption properties of South African bituminous coals. *Elsevier*, *25*, 40-53. <u>https://doi.org/10.1016/j.dib.2019.104248</u>.

13. Czerw, K., Dudzińska, A., Baran, P., & Zarębska, K. (2019). Sorption of carbon dioxide on the lithotypes of low rank coal. *Adsorption*, *25*, 965-972. <u>https://doi.org/10.1007/s10450-019-00122-5</u>.

14. Wojtacha-Rychter, K., Howaniec, N., & Smoliński, A. (2020). Effect of porous structure of coal on propylene adsorption from gas mixtures. *Scientific reports, 10*, 11277. <u>https://doi.org/10.1038/s41598-020-67472-x</u>.

15. Barbara Dutka (2021). Effect of depth on the sorption capacity of coals affected by outburst hazard. *Energies, 306.* <u>https://doi.org/10.1016/j.fuel.2021.121611</u>.

16. Godyn, K., Dutka, B., Chuchro, M., & Młynarczuk, M. (2020). Synergy of Parameters Determining the Optimal Properties of Coal as a Natural Sorbent. *Energies*, *13*, 1967. <u>https://doi.org/10.3390/en13081967</u>.

17. Bulat, A.F., Lukinov, V.V., & Bezruchko, K.A. (2017). *Conditions of gas traps forming in carboniferous sediments*. Kyiv: Naukova Dumka. ISBN 978-966-00-1534-0.

Сорбційна здатність і природна газоносність вугільних пластів Донбасу

К. А. Безручко^{*}, О. В. Бурчак, Л. І. Пимоненко, В. В. Челкан

Інститут геотехнічної механіки імені М.С.Полякова НАН України, м. Дніпро, Україна

* Автор-кореспондент e-mail: <u>gvrvg@meta.ua</u>

Мета. Визначення загальних закономірностей формування сорбційної здатності вугілля та природної регіональної метаноносності вугільних пластів із використанням кількісного показника — відносного градієнта газоносності. Методика. Для визначення сорбційної здатності вугілля використовувалися загальновідомий «об'ємний метод» і метод ЕПР-спектроскопії (електронного парамагнітного резонансу). Для аналізу природної газоносності вугільних пластів використані результати, отримані у процесі ведення геолого-розвідувальних робіт. Визначення природної газоносності здійснювалося за допомогою спеціальних газокернонабірників і пластовипробувачів. Для обробки експериментальних даних застосовувалися методи математичної статистики.

Результати. За експериментальними даними щодо сорбційної метаноємності та природної газоносності вугільних пластів Донбасу проведені аналіз, статистична обробка та узагальнення отриманих результатів. Встановлена закономірна зміна відносного градієнту метаноносності вугільних пластів для всього Донбасу в цілому. Значення відносних градієнтів для вугілля різних марок закономірно зменшуються зі збільшенням глибини залягання й також закономірно зменшуються в кожному з інтервалів глибин від низькометаморфізованого (марка Д) вугілля до високометаморфізованого. За результатами апроксимації отримані відповідні рівняння для кожної марки метаморфізму вугілля від газового до напівантрацитів (А).

Наукова новизна. Запропоновані нові показники відносний градієнт сорбційної метаноємності й відносний градієнт газоносності, що дозволяють порівняння цих характеристик, виміряних в абсолютних величинах $(cm^3/r, m^3/r)$ для окремих вугільних пластів різних марок метаморфізму, на різних глибинах і залягаючих у різноманітних геологічних умовах. Доведено, що сорбційна здатність вугільної речовини визначає природну регіональну (фонову) метаноносність пластів кам'яного вугілля й закономірно, за гіперболічною залежністю, зменшується зі збільшенням глибини залягання й також закономірно зменшується в кожному з інтервалів глибин від низькометаморфізованого (марка Д) вугілля до високометаморфізованого (марка А) з відносним градієнтом, який асимптотично наближається до 1 за тиском понад 6 MПа.

Практична значимість. Отримані залежності відносного градієнту газоносності від глибини й газового тиску для різних марок метаморфізму можуть бути використані для прогнозування природної регіональної (фонової) газоносності вугільних пластів шляхом визначення граничної сорбційної здатності та розрахунку на шукану глибину або тиск.

Ключові слова: Донбас, вугільні пласти, сорбційна здатність, газоносність

The manuscript was submitted 08.03.24.