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On the development of a method for the material balance of metamorphic processes in the transformation of coal seams

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Abstract

Establishing the material balance of metamorphic processes, in addition to solving general scientific geological problems, also has purely practical significance for successfully solving problems related to the safe conduct of mining operations. These problems are associated with the potential manifestation of hazardous properties of coal seams, which are caused by increased release of flammable and explosive gases, sudden outbursts of coal, rock, and gas, the occurrence of endogenous fires, and other adverse phenomena that complicate mining operations. For these reasons, scientific research related to establishing the material balance at different stages of metamorphic transformations of coal seams is relevant both for geological science as a whole and for solving local practical problems related to the safe development of coal deposits. The aim of this work is to develop a methodology for determining the material balance of metamorphic processes, taking into account the individual characteristics of each coal seam. The study resulted in conclusions and recommendations on the feasibility of applying the developed method to assessing the hazardous properties of coal seams and ensuring safe mining operations.

Keywords: Coal seams, Elemental composition, Balance, Hazardous properties, Metamorphic processes, Safety.

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Contents

Contribution of this paper to the literature

Analyzed existing methods for assessing gaseous metamorphic products, established empirical relationships for changes in the elemental composition of coals, and determined quantitative parameters for coal seam transformation based on experimental data on the elemental composition of coals, mathematical modeling of metamorphic processes, and an analysis of the relationships between the main components of organic matter.

1. Introduction

A reliable quantitative assessment of changes in the material balance during the coalification of the original organic matter is necessary for the scientific substantiation of carbon reserves in volatile products formed as a result of the metamorphism of the entire mass of organic matter buried in sedimentary rocks. This enormous amount of continuously generated gases plays a significant role in the formation of groundwater chemistry and in the overall carbon cycle in the Earth's crust [1].

Establishing the material balance of metamorphic processes, in addition to solving general scientific geological problems, also has purely practical significance for the successful resolution of problems related to the safe conduct of mining operations.

These problems are associated with the potential manifestation of hazardous properties of mine seams, which are caused by increased release of flammable and explosive gases, sudden outbursts of coal, rock, and gas, the occurrence of endogenous fires, and several other negative phenomena that complicate mining operations. For the above reasons, scientific research related to establishing the material balance at different stages of metamorphic transformations of coal seams is relevant both for geological science in general and for solving local practical problems of safe development of coal deposits.

A previously developed method for the quantitative assessment of gaseous products released during the coalification of humic organic matter [1] does not provide a strict, consistent sequence in the series of values, but only establishes a general trend in the change in the content of organic matter components and removed volatile products during the metamorphic transformation of coals. An additional disadvantage of the method under consideration is the division, without scientific justification, of all metamorphic processes into equal stages based on the release of coke into organic matter.

With this approach, a similar course of processes of transformation of the initial organic matter at different stages of coalification is assumed in advance. The absence of such uniformity in the course of processes is confirmed by data on the different temperatures of geological transformations of rocks during sedimentation, surface burial, diagenesis, metamorphism, partial melting and magma formation, as well as during thermal decomposition and ashing of fossil coals [2].

One of the possible reasons for the deviations of the calculated values from a certain pattern, according to the author himself [1], is the incomplete identity of the source material for each of the stages of metamorphism under consideration.

According to experimental data [3, 4], in all cases, the material of each coal seam is represented by an identical list of the main components of the elemental composition. This list, based on the sum of the elemental composition of the original substance, is represented by carbon, hydrogen, nitrogen, sulfur, and oxygen, almost 100%. Distinctive features between individual coal seams in terms of elemental composition lie only in the different ratios of the main components. These ratios determine the practical uniqueness and individuality of the material balance for each separately considered coal seam during the process of metamorphic transformations.

2. Materials and Methods

The idea is to partially eliminate the influence of different individual combinations between the main components of organic matter (carbon, hydrogen, nitrogen, sulfur and oxygen) on the formation of stages of metamorphic transformations of separately considered coal seams, which differ slightly from each other in the ratio of the average values of the main components at these stages, for the establishment of which the intersection points of the averaging curves of the dependences of each main component on the carbon content are used.

The objective is to establish the general quantitative direction of the change in the ratio between the main natural elemental components of the source material (carbon, hydrogen, nitrogen, sulfur, oxygen) and their average values at each stage of metamorphic transformations, which will allow for individual adjustments to the material balance of metamorphic processes for each specific coal seam under consideration, taking into account the actual established ratios between the main components of the elemental composition.

The methodology is based on experimental data on the elemental composition of the combustible portion of fossil coals from the Donetsk and Lviv-Volyn basins, available in reference documents [3, 4]. The composition of the products formed during the geologically long-term alteration of buried organic matter can only be partially characterized in this case.

Gaseous and liquid products of carbonaceous substance metamorphism, given the specific features of their physical state, are largely removed, with only solid residual products remaining completely intact [1]. For this reason, precise quantitative characterization is only possible for residual products—various types of carbonaceous substances. The balance of the total amount of substances and the characteristics of the removed gaseous and liquid products can only be established indirectly. The reliability of such determination depends, first and foremost, on the accuracy of quantitative determination of the elemental composition of the solid residual product at different stages of metamorphism.

This is the subject of our research. They are based on a specially developed new methodology, the scientific basis of which differs fundamentally from the known provisions [5] used in the development of the modern industrial classification [6]. The need to develop a new methodology for determining the stages of metamorphic transformations of coal seams is due to the fact that the issues of determining the consumer properties of fuel and the hazardous properties of coal seams are inherently different problems. Their solution requires completely different classification indicators and a scientific justification for their application. To establish the consumer properties of coals, according to Donetsk Research Coal Institute (DonUGI) [5], four areas of characteristics are distinguished.

- Source plant material.
- Quantitative petrographic composition by type of substance.
- Type by restoration.
- Degree of metamorphism.

These directions of change of characteristics in various combinations determine the entire diversity of consumer properties of coals established by the standard [6].

3. Results and Discussion

To determine the hazardous properties of coal seams, it is absolutely necessary to use the first two sets of features characterizing genetic parameters. According to Donetsk Research Coal Institute (DonUGI) [5] and GOST 25543–2013 [6], the yield of volatile matter during thermal destruction of coal is accepted as one of the main indicators of the degree of metamorphism, as it sufficiently characterizes many consumer properties of the fuel. At the same time, the yield of volatile matter, by its very nature, does not correspond to the generally accepted definition of metamorphism. According to the officially accepted definition [7], metamorphism is understood as the sequential transformation of brown coal into hard coal and anthracite as a result of changes in the chemical composition, structure, and physical properties of coal in the subsurface, primarily under the influence of elevated pressure and temperature.

More generally, such transformations apply not only to coal seams, but to all metamorphic rocks. They consist of changes in the mineral composition or the size and texture of grain aggregates, without significant changes in the chemical composition (except for the content of H_2O and CO_2) under the influence of fluids, temperature and pressure [8].

Metamorphic rocks are formed by the transformation of igneous or sedimentary rocks under the influence of high pressure, temperature, and hot gas-water solutions. As physical bodies, they are characterized by density, elasticity, strength, thermal, electrical, magnetic, radiation, and other properties, which primarily depend on their mineral composition and microstructure (structural-textural features) [9].

The volatile matter yield does not directly correspond to any formulation of the definition of metamorphism [7-9]. For the most part, it is a criterion for the quality of fossil coals, which largely determines their consumer properties.

According to generally accepted definitions of metamorphic processes [7-9], to fully characterize the transformation of mine seams, it is necessary to use not just one, but several dozen indicators. These indicators are divided into two fundamentally distinct categories. The first includes indicators that directly determine the elemental composition of organic matter, while the second includes indicators that comprehensively characterize its physical and mechanical properties.

The material balance of solid residual products of metamorphic processes is fully and uniquely determined for each seam (up to 100%) by the sum of the five main components of the elemental composition: carbon, hydrogen, nitrogen, sulfur, and oxygen. As the influence of metamorphic processes intensifies, the ratio of the elemental content of each of these components changes.

It has been established [1, 5] that this process leads to a unilateral increase in the carbon content and a decrease in the remaining components. Given this circumstance and the proximity of the sum of the main components to 100%, the interdependence between the carbon content and the sum of the remaining components will be, in practical terms, functionally described by the equation:

$$\sum H_o, N_o, S_o, O_o = 100 - C_o, \quad (1)$$

Where C_o, H_o, N_o, S_o, O_o are the individual elemental content of carbon, hydrogen, nitrogen, sulfur and oxygen in the organic mass for each mine seam, respectively, %.

When the condition of functional equality (1) between the carbon content and the sum of the remaining main components is met, for different coal seams, there are, to a significant extent, far from unambiguous dependencies of individual components on the carbon content (Figure 1).

In the case under consideration, experimental data on the content of the main components in the combustible part of the fuel were accepted for analysis [3]. This information was obtained on the basis of the analysis of enriched coal samples (ash yield less than 10%), which led to a minimal difference between the content of the main components in the organic matter and the combustible part of the fuel, respectively. The existing deviations of the experimental data from the averaging curves (Figure 1) indicate a different effect of the original plant material and its petrographic composition on the ratio between the main components with intensification of metamorphism (an increase in the carbon content to 97%).

Over the entire range of metamorphic transformations of coals and anthracites, with an increase in the carbon content, close correlations are observed between the decrease in oxygen (O_r) and hydrogen (H_r). The determination coefficients (R^2) in both cases are equal to 0.87, and the standard deviations (σ) are 1.11 and 0.46%, respectively (Figure 1, a and b). This indicates a deliberate decrease in the average O_2 and H_2 contents as the carbon content increases. Such clearly expressed changes, as the influence of metamorphic processes increases, are not observed for the dependences of the elemental content of nitrogen (Figure 1, c) and sulfur (Figure 1, d).

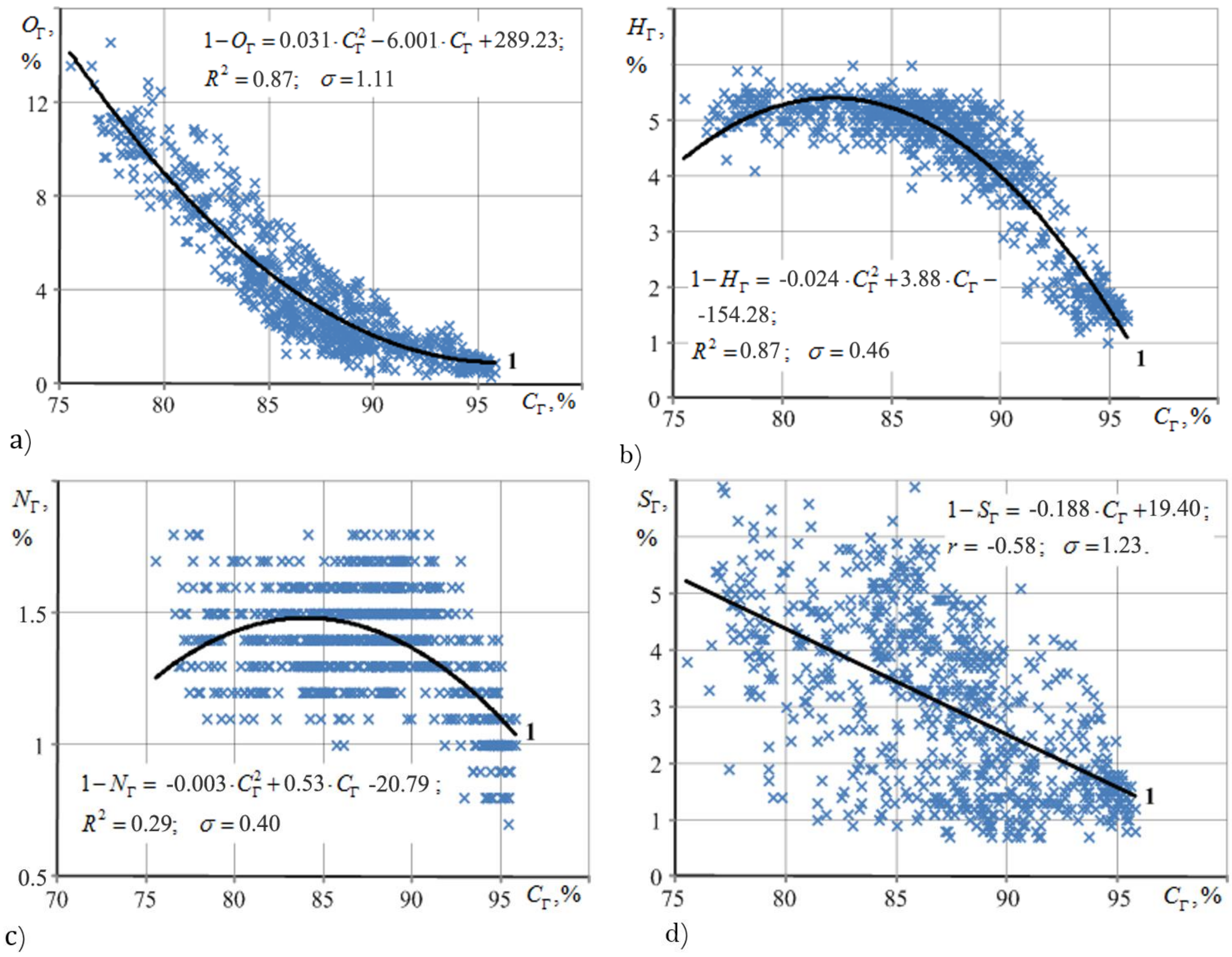

Figure 1. Dependence of the content of oxygen (a), hydrogen (b), nitrogen (c) and sulfur (d) in the combustible part of the fuel on the carbon content (C_Γ) for coal seams and anthracites.

Note: 1 - averaging curves; × - experimental data [3]; R^2 and σ - respectively, the coefficients of determination and the standard deviations from the averaging curves.

Only the tendencies towards decreasing content of the components under consideration can be noted. The coefficients of determination for these components are 0.29 and 0.35, respectively, and the standard deviations are 0.40 and 1.23%. Despite the existing significant deviations of the experimental data from the averaging curves, the individual sum of the content of the main components (C_Γ, H_Γ, N_Γ, S_Γ and O_Γ) for a separately considered seam remains practically constant and equal to 100%. If, with an increase in carbon content, the sum of the remaining components, according to Equation 1, is inversely proportional and functionally reduced, then with a general tendency towards reduction of the remaining components (Figure 1), equality (1) must be observed for each individually considered mine seam. Such equality in the process of intensification of metamorphism and targeted reduction of oxygen and hydrogen (Figure 1, a and b) is possible only under the condition that the decrease in the content of O_Γ and H_Γ is compensated for by a corresponding change in the content of nitrogen and sulfur. Compensation is possible due to the existing deviations of the individual values of the main components from the averaging curves towards a decrease or increase. This is confirmed by the approximate equality of the sum of the standard deviations from the averaging curves of oxygen and hydrogen content (1.11 + 0.46 = 1.57%) and the sum of the standard deviations of nitrogen and sulfur content (0.40 + 1.23 = 1.63%). With such ratios of the standard deviations of individual values from the averaging curves, the balance of the sum of the main components being equal to 100% is practically maintained.

If we proceed from the reliably established facts that in the process of intensification of metamorphic processes the carbon content increases unilaterally, and the sum of the remaining components decreases [1, 5] and at the same time, according to Donetsk Research Coal Institute [3] and Donetsk Research Coal Institute [4] equality (1) is observed, then we can draw conclusions that are important for geological science and justify the research methodology.

- The ratios between the main components of the organic (combustible) mass for each coal seam were initially formed and established during the early stages of coal formation. They individually depend on the original plant material and its petrographic composition.
- At the stages of metamorphic processes, some change in such ratios occurred, associated with a one-sided increase in the carbon content and an individual decrease in the content of each of the other main components with varying intensity.
- With the intensification of metamorphic processes and the general tendency toward a decrease in the average values of oxygen, hydrogen, nitrogen, and sulfur, it follows that at individual stages of metamorphism, there was no significant increase in the content of these components. Possible variations in their individual values for individual seams occurred within the limits of standard deviations from the averaging curves of the dependences on carbon. The sum of the contents of the five main components of organic matter, including the carbon content, remained approximately equal to 100 percent.
- Consideration of the averaging curves of the dependences of the main components on the carbon content makes it possible to partially eliminate the influence of the initial plant material and petrographic composition on the change in the ratio between them.

The intersection points of the averaged curves of the dependences of the main components on the carbon content represent the approximate boundaries of the stages of metamorphic transformation of the seams, characterized by changes in the ratios between the main components. This provides the opportunity to monitor the quantitative direction of changes in the material balance of solid products at each stage of the metamorphic processes.

Based on the statistical processing of experimental data [3], empirical dependencies of the content of the main components of the combustible part of the fuel (oxygen, hydrogen, nitrogen, sulfur) on the carbon content were established (Figure 1). In all cases, these dependencies were presented as second-degree polynomials (square trinomials). This made it possible to determine the intersection points of the averaging curves of the dependence of the main components on the carbon content. The coordinates of these points are the approximate boundaries of the stages of metamorphic transformations of the coal seams both in terms of the carbon content factor and the ratio between the remaining components of the combustible (organic) part of the fuel. The coordinates of the points were determined from the condition that at the intersection point of two curves describing the change in the content of two components (y_1, y_2), the carbon content (x) should be the same for each of them (Figure 2).

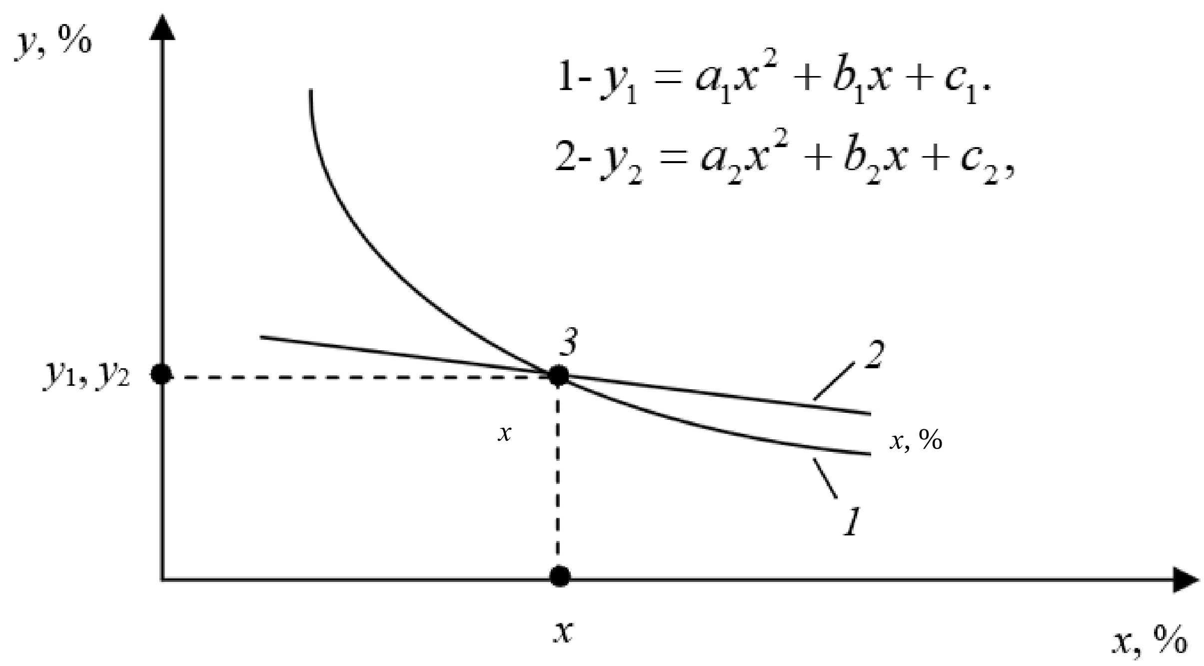

Figure 2. Scheme for determining the coordinates of the intersection point (3) of two averaging curves (1,2) characterizing the average content of the components under consideration in the combustible (organic) part of the fuel (y_1, y_2) depending on the carbon content x.

Note: 1,2 – averaging curves and empirical equations describing them; 3 – intersection point of curves 1 and 2.

The elemental content of one of the components is described by the equation.

$$y_1 = a_1x^2 + b_1x + c_1 \quad (2)$$

The second one has a similar dependence:

$$y_2 = a_2x^2 + b_2x + c_2 \quad (3)$$

Where $a_1, b_1, c_1$and a_2, b_2, c_2 are the empirical coefficients of Equations 2,3 established by the least squares method to describe the elemental content of components 1 and 2, respectively. Their quantitative values are shown in the graphs (Figure 1).

To determine the carbon content x at the intersection point of two curves (Figure 2), taking into account the equality of the elemental content of the two components (y_1, y_2), a new equation of a square trinomial with known coefficients is obtained: $a_1 - a_2, b_1 - b_2, c_1 - c_2$.

$$(a_1 - a_2)x^2 + (b_1 - b_2)x + (c_1 - c_2) = 0. \quad (4)$$

By solving Equation 4 for x, we found the quantitative value of the carbon content corresponding to the abscissa of point 3 of the intersection of two curves 1 and 2 (Figure 2).

In the process of processing the experimental data [3] using the developed methodology, ten characteristic stages of metamorphic transformations of coal seams were identified (Figure 3). They differ from each other in different ranges of changes in the content of both carbon and other main components of the combustible (organic) mass, which is fully consistent with the definition of the processes of metamorphic transformations of seams in terms of changes in the elemental content of the main components of the combustible (organic) mass of coals [5-7]. In addition, the obtained results allow us to analyze the intensity (rate) of changes in the main components at each stage of metamorphic transformations. For these purposes, a technique for determining the intensity of individual changes in the elemental composition of the main components at each stage of metamorphic processes of seam transformations has been specially developed. If we proceed from the possible types of averaging lines of change in the dependencies of the average values of the elemental content of each of the main components (Figure 3), then possible options for the direction of change in the average content of any main component at an isolated (separate) stage of metamorphic transformations can be presented in the form of graphs (Figure 4). Rectilinear segments of the averaging lines (1), parallel to the abscissa axis, indicate a constant (unchanging) value of the average component content within the range of carbon content variation (x_1 - x_2) (an isolated stage of metamorphic processes of strata transformation). More generally, a reduction in the average component content within the range of increasing carbon content from x_1 *to* x_2 can occur along curvilinear dependencies, which, in their nature, are changes similar to segments of lines 2 and 4.

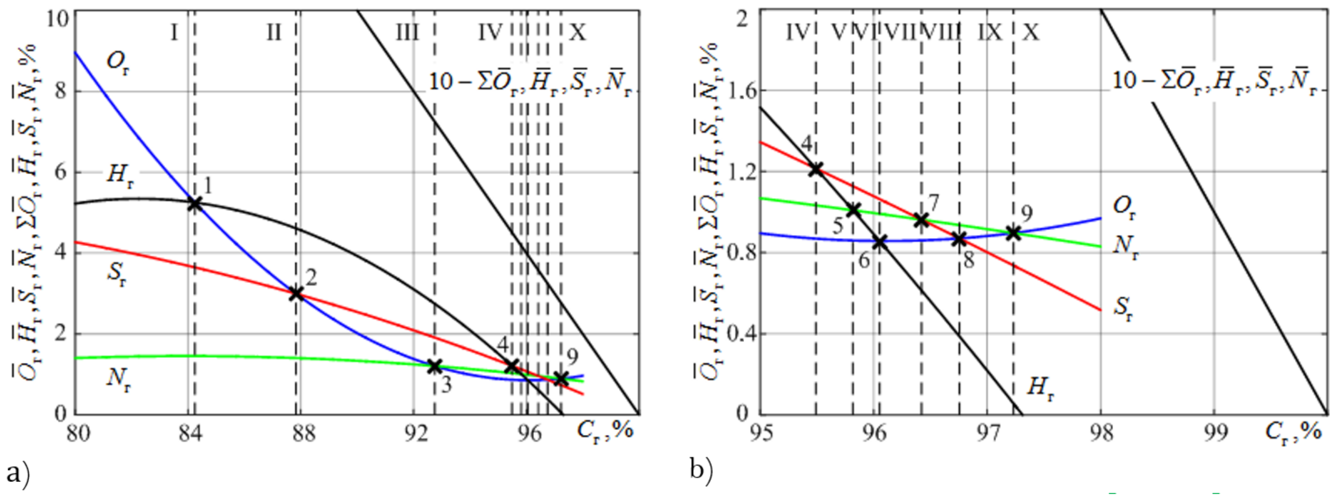

Figure 3. Graphs of changes in the average content of the main components in the combustible part of the fuel $(\bar{O}_г, \bar{H}_г, \bar{N}_г, \bar{S}_г)$ from the carbon content ($C_г$) based on statistical processing of experimental data on the coal seams of the Lviv-Volyn basins [3], a - full; b - enlarged.

Note: 1,2,3 – intersection points of averaging curves of changes in oxygen content ($\bar{O}_г$) with hydrogen ($\bar{H}_г$), sulfur ($\bar{S}_г$) and nitrogen ($\bar{N}_г$), respectively, at different stages of metamorphic transformations of mine seams; 4,5 – intersection points of averaging curves of hydrogen ($\bar{H}_г$), with sulfur ($\bar{S}_г$) and nitrogen ($\bar{N}_г$), respectively; 6,8,9 – intersection points of averaging curves of changes in oxygen content ($\bar{O}_г$), with hydrogen ($\bar{H}_г$), sulfur ($\bar{S}_г$) and nitrogen ($\bar{N}_г$), respectively, at late stages of metamorphic transformations of mine seams; 7 – intersection point of averaging curves of sulfur ($\bar{S}_г$) and nitrogen ($\bar{N}_г$); 10 – straight line dependence of the sum of the main components ($\sum \bar{O}_г, \bar{H}_г, \bar{S}_г, \bar{N}_г$) on the carbon content; I, II, III, IV, V, VI, VII, VIII, IX, X – stages of metamorphic transformations of coal seams established by the change in the ratio between the average values of the main components in the combustible part of the fuel.

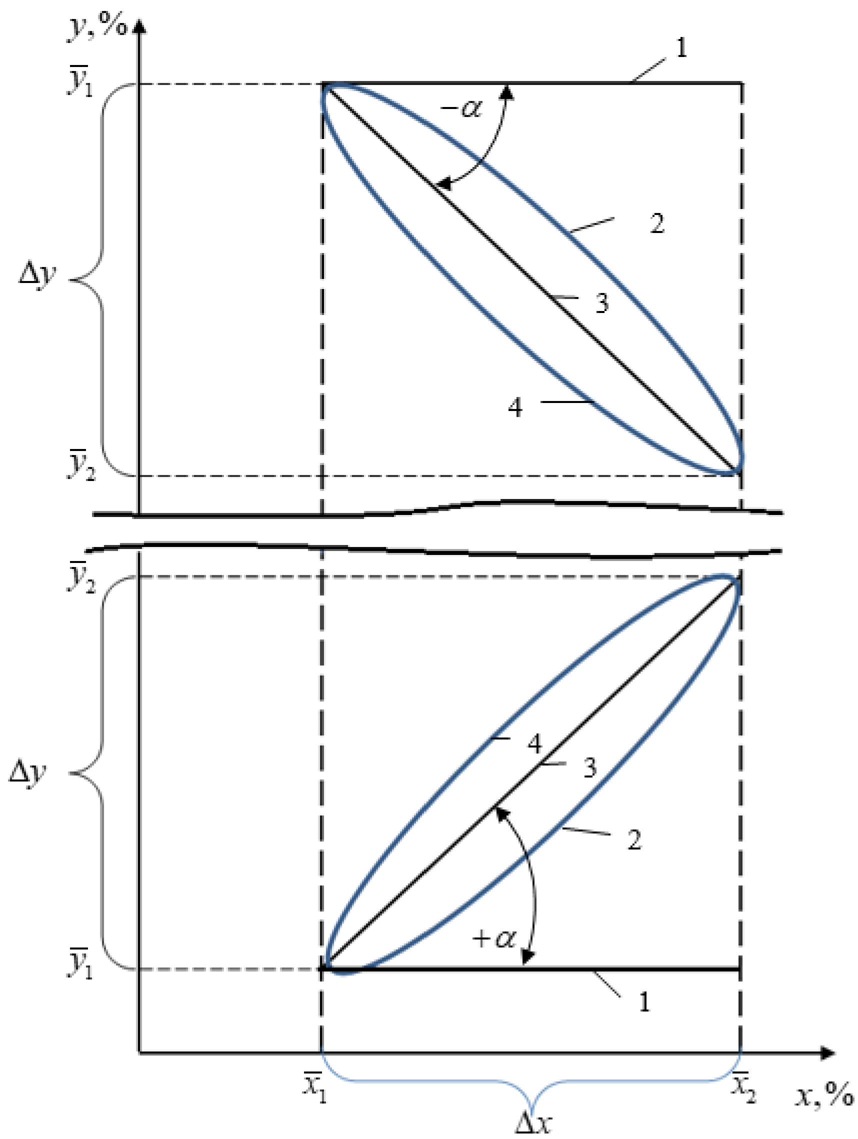

Figure 4. Possible options for changing the individual direction of the average content of a separate component in the combustible (organic) part of the fuel (y) within the boundaries of an isolated stage of the metamorphic processes of transformation of coal seams (carbon content x_1, x_2).

Note: 1,2,3,4 – possible variants of the type of segments of averaging lines characterizing the intensity and direction of change in the elemental content of the component under consideration; $\bar{x}_1$, $\bar{x}_2$– respectively, the lower and upper limits of the average carbon content at a separate stage of coal seam transformation with increasing influence of metamorphic processes (increase in carbon content x, %); Δx – range of carbon change, %; $\bar{y}_1$, $\bar{y}_2$– boundaries of change in the average content of the component under consideration with increasing influence of metamorphic processes, %; Δy– range of change in the average content of the component under consideration, %; α – angle determining the intensity of change in the content of the component under consideration at a separate stage of metamorphic transformations of coal seams according to the ratio $\frac{\Delta y}{\Delta x}$.

The inversely proportional individual decrease in the elemental content of a component can also be described by a straight line segment 3, located at an angle of $-\alpha$ to the abscissa axis (Figure 4, a).

Variants of possible growth or unchanged average content of the component within the limits (x_1 - x_2) are not excluded. An increase in carbon content at a specific stage of metamorphic transformations of the strata. For these cases, the individual change in the elemental content of a component can be described by segments 1, 2, 3, 4 (Figure 4, b).

In all the cases considered (Figure 4), regardless of the type of segments (1, 2, 3, 4), the intensity of change in the average content of the component at the stage under consideration is determined by the ratio of the length of the segments Δ y *to* the segment Δx, i.e., the tangent of the slope angle α.

Using the method for determining the boundaries of individual stages of the processes of metamorphic transformations of coal seams (Figure 2, Equation 4, Figure 3) and possible options for changing the individual direction of the average content of each main component within the stage under consideration (Figure 4), as well as on the basis of experimental data on the content of the main components in the combustible part of coals [3] the value of Δy and Δx was calculated for each main component (Table 1).

The moisture content of fossil coals is one of the most important indicators of the degree of metamorphic transformations of coal seams and the quality of the fuel [1, 6, 10], but in the normative and reference documents [3, 4] it is not considered together with the main components in the composition of the combustible (organic) part of coal. Moisture largely determines the manifestation of the hazardous properties of coal seams during mining operations [11-14]. For this reason, Table 1 also presents the results of calculating the change in the average moisture content (W) at each stage of the metamorphic processes, established by the intersection points of the curves determining the change in the ratio between the main components of the combustible (organic) mass (Figure 5).

Table 1. Results of determining the average boundaries of the stages of metamorphic transformations of mine seams based on changes in the ratio of the main components of the combustible part of the fuel based on statistical processing of experimental data.

Stages of metamorphism	**Intersection points of averaging curves**		**Components of the combustible mass**															**Moisture content in samples, W**		
	Number of points	**Components of the combustible mass**	C_{Γ}		O_{Γ}			H_{Γ}			S_{Γ}			N_{Γ}			$\sum \Delta O_{\Gamma}, \Delta H_{\Gamma}, \Delta S_{\Gamma}, \Delta N_{\Gamma}$%	**Ranges, %**	ΔW %	$\frac{\Delta W}{\Delta C_{\Gamma}}$ **shares**
			Ranges, %	ΔC_{Γ}, %	**Ranges, %**	ΔO_{G}, %	$\frac{\Delta O_{\Gamma}}{\Delta C_{\Gamma}}$ **shares**	**Ranges, %**	ΔH_{G}, %	$\frac{\Delta H_{\Gamma}}{\Delta C_{\Gamma}}$ **shares**	**Ranges, %**	ΔS_{G}, %	$\frac{\Delta S_{\Gamma}}{\Delta C_{\Gamma}}$ **shares**	**Ranges, %**	ΔN_{G}, %	$\frac{\Delta N_{\Gamma}}{\Delta C_{\Gamma}}$ **shares**				
I	1	O_{Γ},H_{Γ}	75.00-84.24	9.24	13.58-5.18	-8.40*	-0.901*	4.20-5.18	+0.98*	+0.11	4.77-3.59	-1.18*	-0.128*	1.25-1.45	+0.20*	+0.022*	8.35	13.99 - 3.74	-10.25*	-1.109*
II	2	O_{Γ},S_{Γ}	84.24-87.83	3.09	5.18-2.89	-2.29	-0.741	5.18-4.61	-0.57	-0.184	3.59-2.89	-0.70	-0.227	1.45-1.40	-0.05	-0.016	3.61	3.74 - 2.53	-1.21	-0.392
III	3	O_{Γ},N_{Γ}	87.83-92.75	4.92	2.89-1.12	-1.77	-0.360	4.61-2.75	-1.86	-0.378	2.89-1.85	-1.04	-0.241	1.40-1.12	-0.28	-0.057	4.95	2.53 - 3.25	+0.72	+0.146
IV	4	N_{Γ},S_{Γ}	92.75-95.49	2.74	1.12-0.69	-0.43	-0.157	2.75-1.18	-1.57	-0.573	1.85-1.18	-0.67	-0.245	1.12-1.03	-0.09	-0.033	2.76	3.25 - 4.80	+1.55	+0.566
V	5	H_{Γ},N_{Γ}	95.49-95.85	0.32	0.69-0.68	-0.01	-0.031	1.18-1.01	-0.17	-0.531	1.18-1.06	-0.12	-0.375	1.03-1.01	-0.02	-0.063	0.32	4.80 - 4.98	+0.18	+0.563
VI	6	O_{Γ},H_{Γ}	95.81-96.05	0.24	0.68-0.77	+0.09	+0.375	1.01-0.77	-0.24	-1.000	1.06-0.99	-0.07	-0.292	1.01-0.99	-0.02	-0.083	0.24	4.98 - 5.22	+0.24	+1.000
VII	7	S_{Γ},N_{Γ}	96.05-96.66	0.61	0.77-0.68	-0.09	-0.148	0.77-0.45	-0.32	-0.525	0.99-0.88	-0.11	-0.180	0.99-0.88	-0.11	-0.180	0.63	5.22 - 5.73	+0.51	+0.836
VIII	8	O_{Γ},S_{Γ}	96.66-96.75	0.09	0.68-0.74	+0.06	+0.667	0.45-0.39	-0.06	-0.667	0.88-0.74	-0.14	-1.556	0.88-0.93	+0.05	+0.556	0.09	5.73 - 5.80	+0.07	+0.778
IX	9	O_{Γ},N_{Γ}	96.75-97.23	0.48	0.74-0.80	+0.06	+0.125	0.39-0.06	-0.33	-0.688	0.74-0.66	-0.08	-0.167	0.93-0.80	-0.13	-0.0271	0.48	5.80 - 6.24	+0.44	+0.917
X	10	-	>97.23	-**	-**	-**	-**	-**	-**	-**	-**	-**	-**	-**	-*		-**	-**	-**	-**

Note: * - the sign "-" means a decrease in the content of the main components or moisture in the range of the stage under consideration, determined by the adjacent intersection points of the averaging lines, and "+" means an increase in the elemental content; ** - experimental data are not available.

Source: Donetsk Research Coal Institute [3].

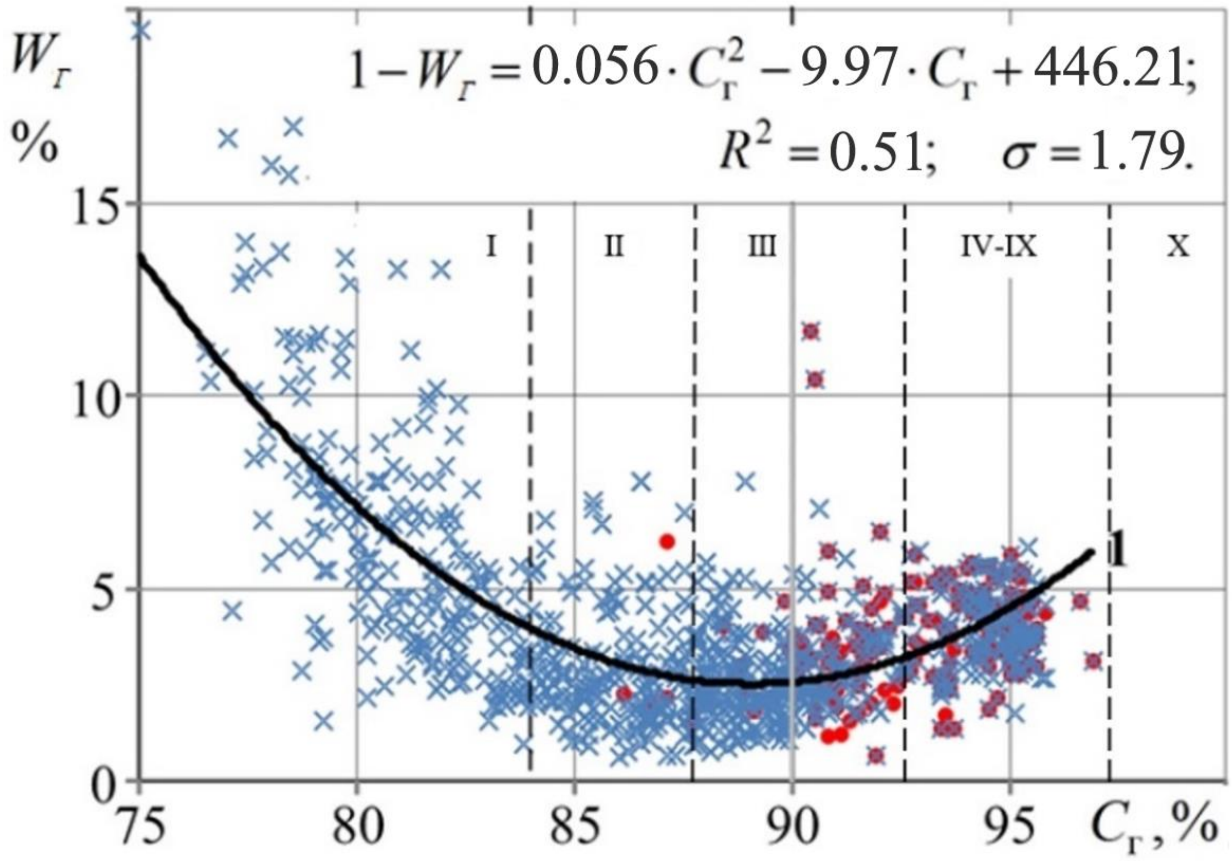

Figure 5. Dependence of moisture content (W_{Γ}) in coal samples on the carbon content (C_{Γ}) in the combustible part of the fuel.

Note: 1 – averaging curve; × – experimental data [4]; • – joint experimental data on the moisture content of anthracite and semi-anthracite coal seams according to the catalogue [14] and the carbon content in the combustible part in accordance with the data [4]; I, II, III, IV-IX, X – stages of metamorphic transformations of coal seams, established by the change in the ratio between the average values of the main components in the combustible part of the fuel (Figure 3); R^2, σ – determination coefficient and standard deviation.

The dependence of moisture on the carbon content for the seams of hard coals and anthracites of the Donetsk and Lviv-Volyn basins, according to Rudniev, et al. [15], with a fairly close correlation dependence, is described by the equation:

$$W = 0.056 \cdot C_{\Gamma}^2 - 9.36 \cdot C_{\Gamma} + 446.21 \quad (5)$$

Empirical dependence (5) was obtained on the basis of statistical processing of experimental data for hard coals, on moisture and carbon content, given in the quality handbook [4]. In this reference and normative document, information on the content of formation moisture is not given for coal seams containing anthracites and semi-anthracites. For a more detailed study of the change in moisture content during metamorphic transformations, information [14] on moisture content for anthracite and semi-anthracite coal seams were added to the available data [4]. This made it possible to consider in a wider range the change in carbon content for coal and anthracite coal seams and to establish dependence (5). The intensity and direction of moisture change at an isolated stage of metamorphic transformations ($\frac{\Delta W}{\Delta C_{\Gamma}}$) were determined using a method similar to the method (Figure 4) for calculating the direction and intensity of change in the content of each main component of the combustible part of the fuel ($\frac{\Delta y}{\Delta x}$) at individual stages. The results of calculations of changes in formation moisture, along with data on changes in the content of the main components, are also summarized in Table 1. This made it possible to jointly analyze changes in the content of the main components of the combustible part of the fuel and moisture.

According to generally accepted concepts of metamorphic processes in rocks, including coals, they occur without significant changes in chemical composition. The exception is the content of moisture and carbon dioxide (CO_2) [8]. Along with this, according to the hypothesis [16], during the metamorphism of the original substance and the transformation of coals from long-flame to anthracite, a significant amount of moisture and methane should have been formed and released. The formation of methane, at least in significant quantities, refers to the later stages of metamorphic processes [1]. In the cases under consideration 16, the formation and removal of a large amount of fluids during metamorphism should facilitate and lead to a change in the chemical composition of the original substance. From the classical definition of metamorphic processes follows the practical constancy of chemical composition [8]. Some discrepancy between the classical definition of metamorphic processes and the hypotheses [1, 16] is caused by the fact that the complete cycle of coal formation was not taken into account during their development.

Metamorphism does not refer to the complete transformation cycle of the original organic matter, but is a continuation of the processes of sedimentation, peat formation, burial and diagenesis [17]. Natural decomposition processes occurred during the stages of sedimentation and peat formation [18, 19]. The degree of coal metamorphism characterizes the changes in the composition and properties achieved during coal formation and determines its position in the genetic series: brown coal – hard coal – anthracite [7].

During sedimentation and peat formation, during the anaerobic decomposition of organic matter, as a result of the activity of methanogenic bacteria, methane (CH_4) and carbon dioxide (CO_2) are formed, which are released into the atmosphere [20]. As the depth of immersion increases, conditions are created that hinder the release of gases to the surface and the biochemical processes attenuate. In parallel with the attenuation of biochemical processes, the role of the chemical transformation of coals increases. During the process of coalification under conditions of high pressures and temperatures, methane and carbon dioxide were formed, associated with the transformation of coal seams. According to Patteisky, the methane content in $1m^3$ of coal reached $350m^3$, and carbon dioxide - up to $200m^3$

[19]. This amount of gases was released during the entire complete cycle of transformation of organic matter in the genetic series brown coal - hard coal - anthracite. Gaseous and liquid coal products, due to their physical state, were removed during peat formation and the development of brown coal seams, which are characterized by diagenesis. Unlike fluids, solid residual products were completely preserved in their original location and were subjected to metamorphic processes during the formation of hard coal and anthracite seams. The amount of gases retained until the metamorphic transformation of the seams was significantly less than the amount formed during the preceding stages of peat formation and diagenesis. Part of the methane migrated into the host rock or to the surface through fractures in geological faults, while carbon dioxide dissolved in water. Experimental data on the moisture and oxygen content during the transition from peat to brown coal and then to hard coal confirm the existence of such fluid migration processes. In peat, the moisture content is approximately 75%, in brown coals 60% or less, and the oxygen content is 31-40% and 18-26%, respectively [21]. This indicates that during the transition from brown coals to hard coals, the degree of their transformation is associated, to a large extent, with the preliminary amount of moisture and oxygen removed from the brown coals under the influence of external conditions characteristic of metamorphic processes (pressure, temperature, period of time). Under the influence of these factors, brown coal loses up to 90% of its moisture and begins to acquire the qualities inherent in hard coals (caking, coking, resin yield, etc.). The maximum moisture content in hard coals from the Donbas and Lviv-Volyn basin mine seams reaches 20% with a carbon content of 75-80% (Figure 5). Further removal of accumulated moisture during the formation of brown coals continued with the increasing influence of geological processes and their transition from diagenesis to metamorphism. The formation of a significant amount of additional moisture during the metamorphic stages of hard coals did not occur, as evidenced by the intensive decrease in moisture to minimum values during the formation of coal seams. According to the averaging curve (1), the minimum moisture content is approximately 2.5% with a standard deviation of 1.79% and an approximate carbon content of 88-89% (Figure 5). Moisture is an important classification indicator of the metamorphic transformations of coal seams. According to GOST 25543–2013 [6], with the same vitrinite reflectance value for brown and hard coals, it serves as a criterion for dividing brown coals into types. In addition, the moisture content of hard coals allows us to outline the boundary of the anthracite stage of metamorphism. After reaching minimum values at the stages of hard coal, the moisture content increases during the transition to anthracite. The bend in the curve can be considered as the beginning of the anthracite stages [1]. This nature of the change in moisture content is confirmed by the results (Table 1, Figure 5) of statistical processing of experimental data [3, 4]. They indicate the preferential removal of moisture accumulated during diagenesis at coal stages I and II. At subsequent anthracite stages III-IX, an additional increase in the total moisture content occurs due to the release of crystallization water from mineral impurities and pyrogenetic water [2]. Gaseous products of metamorphic processes have a significant impact both on the material balance of these processes and on the manifestation of hazardous properties of mine seams during mining operations. The most hazardous gases in coal mines include the release of methane (CH_4) and carbon dioxide (CO_2) [22]. In many cases, mines working coal and anthracite seams are not hazardous in terms of the emission of these gases, since their release into mine workings is close to zero. At the same time, there are mines working such seams that are hazardous in terms of the emission of methane and carbon dioxide. The maximum gas content of methane in coal seams reaches 30m^3/t, and in anthracite seams - 45m^3/t [23]. If, in general, we compare the possible values of gas content (0 ÷ 45m^3/t) with the amount of methane formed in the overall coalification process (about 350m^3/m^3), this fact fully confirms the migration of its overwhelming majority to the earth's surface or into the host rocks [19].

For this reason, the initial natural gas content of coal seams at the stages of metamorphic transformations has an element of chance, which is directly related to the previous processes of diagenesis. To establish the reasons for the complete migration of methane or its partial preservation, it is necessary to consider the geological history of the formation of the coal deposit. It, at a minimum, should include the influence of geological disturbances and their types, the properties of the host rocks, the depth of occurrence, etc [19, 24]. The considered mechanism of methane formation and release during diagenesis and metamorphism explains the reasons for the different gas content of coals with the same degree of transformation of the original organic matter and the different hazard of coal seams in terms of the gas factor. During the transition to the anthracite stages of transformation of coal seams, their gas content increases due to the intensification of chemical and structural transformations of coals. The main factor in the formation of gaseous products, including methane, at the anthracite stages was high (over 500°C) temperature [25]. This is confirmed by the nature of the change in the ratio of the main components of the combustible (organic) mass (Table 1, Figure 3). Stage III of metamorphic transformations is a transitional stage from hard coals to anthracites, which is confirmed by both the achievement of minimum moisture values (at carbon content values of 88-89% (Figure 5)), and the beginning of the intensity of its growth ($\frac{\Delta W}{\Delta C_{\Gamma}}$= + 0.146, Table 1). In addition, stage III is characterized by an intensive decrease in hydrogen ($\frac{\Delta H_{\Gamma}}{\Delta C_{\Gamma}}$= - 0.378) at a carbon content of more than 87.83% (Table 1). The presented results of statistical processing of the experimental data indicate that the nature of the transformation of mine seams, starting from stage III and at subsequent stages (from III to IX inclusive), is directly related to the change in the ratio between the content of the main components of the combustible (organic) mass. This indicates the possibility of fluid formation at all anthracite stages of metamorphic transformations of the strata (from III to IX) as a result of chemical reactions. The possible fluids formed during chemical reactions can be judged from the individual ratios of the main components with carbon content ($\frac{\Delta y}{\Delta x}$) and the values of the tangents of the angles α, which characterize the intensity and direction of changes in the content of the main components of the combustible (organic) mass of coal and moisture (Figure 4, Table 1).

The quantitative rates of change in the contents of the main components at different stages of metamorphic processes can be judged by the absolute values of the angles α (Table 2). The most intense decrease in oxygen content occurs in the early (I, II) stages of metamorphic processes. Moisture is also rapidly removed during these stages. A gradual decrease in oxygen content continues at stages III, IV, and V, while at stages VI-IX, the oxygen content remains virtually unchanged, averaging less than one percent. A noticeable increase in the rate of increase in moisture content is observed at stages III-IX.

The hydrogen content at stages I, II remains practically constant, and at subsequent stages (III-IX) a consistent decrease is observed (α = -20.7°÷-33.8°).

The rate of sulfur content reduction at all stages occurs with approximately the same intensity (α = -7.3° ÷ -20.6°). Nitrogen content changes virtually unchanged at stages I-V (α = -1.3° ÷ -3.6°). The rate of its reduction increases somewhat at stages VI-IX (α = -8.4°), indicating the possible formation of fluids involving nitrogen.

The obtained quantitative values for the individual changes in each major component of the combustible (organic) mass and moisture allow us to predict the potential formation and removal of fluids at each stage of the metamorphic transformation of coal seams. The different origins of gases and moisture, and the depth of chemical and structural transformation of coal, determine the distinctive properties of coal seams. This, to a large extent, explains the different susceptibility of seams to exhibiting hazardous properties during mining operations.

Table 2. Results of determining the angles α characterizing the average intensity of change in the content of each main component of the combustible part of the fuel at a separate stage of metamorphic transformations of the mine seams according to the results of statistical processing of experimental data.

Stage of metamorphism	Information on the carbon content in the combustible part of the fuel, %			Angle α characterizing the intensity of change in the average content of the components under consideration (Figure 4), deg.				Angle α characterizing the intensity of change in moisture content, degrees	Notes
	Limits of change	Range of change, ΔC_r	Average carbon value, $\bar{C}_r$	$\bar{O}_r$	$\bar{H}_r$	$\bar{S}_r$	$\bar{N}_r$		
I	75.00-84.24	9.24	79.62	-42.1	+6.3*	-7.3	+1.3*	-48.0	Intensive removal of oxygen and moisture, slight increase in hydrogen and nitrogen content
II	84.24-87.83	3.09	86.04	-36.5	-2.3	-12.8	-0.9	-16.8	Removal of oxygen and moisture, beginning of reduction of hydrogen content
III	87.83-92.75	4.92	90.29	-19.8	-20.7	-13.5	-3.3	+8.3	A decrease in the content of oxygen and hydrogen, the beginning of an increase in the moisture content
IV	92.75-95.49	2.74	94.12	-8.9	-29.8	-13.8	-1.9	+29.5	A decrease in the rate of oxygen reduction and an intensive decrease in hydrogen content, an increase in the rate of moisture growth
V	95.49-95.85	0.32	95.65	-1.8	-28.0	-20.6	-3.6	+29.4	A slight decrease in oxygen, the rate of decrease in hydrogen and moisture content remained virtually unchanged compared to the previous stage
VI-IX	95.81 - 97.23	1.42	96.52	0	-33.8	-15.7	-8.4	+41.6	The oxygen content is minimal (less than 1%) remains virtually unchanged, there is an intense reduction in hydrogen, sulfur and nitrogen, and the rate of increase in moisture increases
X	>97.23								

Note: * - when changing C_r in the range of 75.00-84.24%, the average values $\bar{H}_r$and, $\bar{S}_r$according to experimental data [3], remain practically constant. Their insignificant increase (the "+" sign) is due to the nature of the second-order polynomial curve.

Source: Donetsk Research Coal Institute [3].

4. Conclusions

The following conclusions, which are important for both geological science and the coal industry in terms of predicting the manifestation of hazardous properties of mine seams.

- The division of the processes of metamorphic transformations into equal ranges according to the coke yield does not correspond to the sizes of the intervals that determine the changes in the ratio between the content of the main components of the combustible (organic) mass of fossil coals, which indicates the presence of distinctive features of the processes occurring at different stages of metamorphism.
- The establishment of stages of processes of metamorphic transformations of seams at the intersection points of averaging curves of changes in the main components of the combustible (organic) part corresponds to the generally accepted definition of the concept of metamorphism, which includes changes in the chemical composition, structure and physical and mechanical properties of the original organic matter.
- The volatile matter yield during thermal destruction does not, in essence, fully correspond to the definition of metamorphism, but rather serves as a criterion for the consumer properties of coals and anthracites. It cannot independently and comprehensively characterize the interdependent changes in both the elemental composition of the original organic matter of mine seams and their physicochemical and mechanical properties, which determine the manifestation of hazardous properties during mining operations.
- For a complete and comprehensive characterization of the metamorphic processes of transformation of coal seams and the establishment of their hazardous properties, it is necessary to use several dozen indicators that

directly determine the composition and ratio of the main components, and the physicochemical properties of coals.

- The material balance of solid residual products of metamorphic processes is fully and unambiguously (up to 100%) determined for each seam by the sum of the five main components of the elemental composition: carbon, hydrogen, nitrogen, sulfur, and oxygen. An increase in carbon content practically functionally controls the sum of the remaining main components.
- Establishing the boundaries of the stages of metamorphic transformations of coal seams at the intersection points of the averaging curves of the dependences of the main components on the carbon content corresponds to the definition of metamorphic processes in terms of changing the elemental ratio of the content of the main components in the combustible (organic) part of coals.
- The developed method for determining the boundaries of individual stages of metamorphism allows for the quantitative calculation of the intensity and direction of individual changes in the content of the main components of combustible (organic) matter and moisture, which makes it possible to assess the type and quantity of fluids formed within each stage.
- During the metamorphic transformation stages of coal, moisture accumulated during diagenesis is predominantly removed. During subsequent anthracite stages, additional crystallization and pyrogenetic water are released.
- The random nature of the gas content of coal seams with the same degree of metamorphism is a consequence of the removal of gases in different quantities to the earth's surface or into the host rocks, depending on the geological history of the formation of the coal seam.
- During the transition to the anthracite stages of transformation of coal seams, their gas content can increase due to the intensification of chemical and structural transformations of coals.
- The transition from the coal stages to the anthracite stages occurs at a carbon content of 88-89% and is characterized by the additional formation of fluids (moisture and gases) as a result of chemical reactions.

The different origins of gases and moisture, and the depth of chemical and structural transformation of coals, determine the distinctive hazardous properties of coal seams during mining operations. A reliable prediction of hazardous properties is possible using the material balance method for metamorphic processes in coal seams.

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