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Unsolved problems in the study of coal formation processes and the manifestation of hazardous properties of coal seams

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Abstract

To establish problems, the solution of which for a specific stage of coal formation will allow a scientifically sound prediction of the occurrence and manifestation of hazardous properties of a specific coal seam during mining operations. The methodology is based on modern concepts of coal formation processes to confirm or establish the discrepancy between the processes occurring at different stages. Hazardous properties of coal seams during mining operations are formed not only at the stage of metamorphic transformations, and their occurrence is genetically associated, to a large extent, with previous processes of accumulation of the source material, which was subjected to successive transformation at the peat, brown coal, coal, or anthracite stages of coal formation. Based on the conducted research, scientifically substantiated proposals have been developed to clarify the general scheme of coal formation, which significantly changes the understanding of the formation of hazardous properties of mine seams and indicates the need to solve a number of urgent scientific problems associated with improving the regulatory framework for safe mining operations and clarifying geological processes at individual stages of coal formation.

Keywords: Coal formation, Hazardous properties, Problems, Processes, Regulatory frameworks, Safety.

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Contents

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Contribution of this paper to the literature
The possibilities of forming dangerous properties of coal seams are considered not only at the stage of metamorphic transformations but also in relation to their occurrence, which is genetically connected with previous processes of accumulation of the initial material, subjected to successive transformations at different stages of coal formation.

1. Introduction

The hazardous properties of coal seams (release of flammable and explosive gases, sudden outbursts of coal and gas, occurrence of endogenous fires, increased dust formation and its explosiveness, and some other features) during mining operations are considered in modern regulatory documents depending on the degree of metamorphic transformations of coals. As one of the main indicators of the degree of metamorphism and, in most cases, the only one, the yield of volatile substances (mass or volume) during thermal decomposition of coals without access to air is accepted [1-7].

In essence, the indicators V^{daf} and V_v^{daf}, according to the methods of their laboratory determinations based on analytical samples of coal, they are a mixture of gaseous fluids released from dry ash-free mass under artificially created conditions with elevated temperature. The mixture of gases released during the thermal destruction of coal has no direct relation to the occurrence of hazardous properties of coal seams. Fluids, including moisture, which determine hazardous properties during mining operations, have already been removed at the stage of preparing analytical samples by crushing to a size of 7 μm and drying the coal at a temperature above 100℃ [8]. For this reason, the influence on the occurrence and manifestation of hazardous properties of seams should be considered regardless of the indicators V^{daf} or V_v^{daf}, but on the elemental composition of coals and fluids that were formed and preserved (including possibly only partially) during geological processes before mining operations.

2. Materials and Methods

It follows from the state of the issue under consideration that problems related to the reliable prediction of hazardous properties of coal seams during mining operations have not been fundamentally resolved to date. This is confirmed by the ongoing and periodically recurring accidents at enterprises in coal-mining countries around the world caused by gas flares and explosions in mine workings, sudden emissions of coal and gas in production and development faces, the occurrence of endogenous fires, and some other manifestations of hazardous properties. Prevention of such accidents largely depends on the degree of validity of the requirements of regulatory documents governing the safe operation of coal deposits.

The effectiveness of measures taken to prevent accidents depends on the reliability of forecasts regarding the hazardous properties of mine seams. Therefore, research aimed at improving the regulatory framework for safe mining operations remains highly relevant for all coal-mining countries worldwide.

3. Results and Discussion

The formation of methane and carbon dioxide at the peat stage could not have significantly influenced the formation of the hazardous properties of coal seams, since their release occurred almost entirely into the atmosphere.

Water formation did not occur at the peat and brown coal stages, since geological transformations of the initial substance at the diagenesis stages consisted only of its removal. Similar processes of only moisture removal continued at the Carboniferous stage, which refers to metamorphic transformations. The absence of water formation processes at the Carboniferous stage is confirmed by experimental data on the graphs of the decrease in maximum moisture capacity and stable hydrogen content (≈ 4.5÷5.5%) depending on carbon in the range of its change from approximately 75 to 88% (Figure 2).

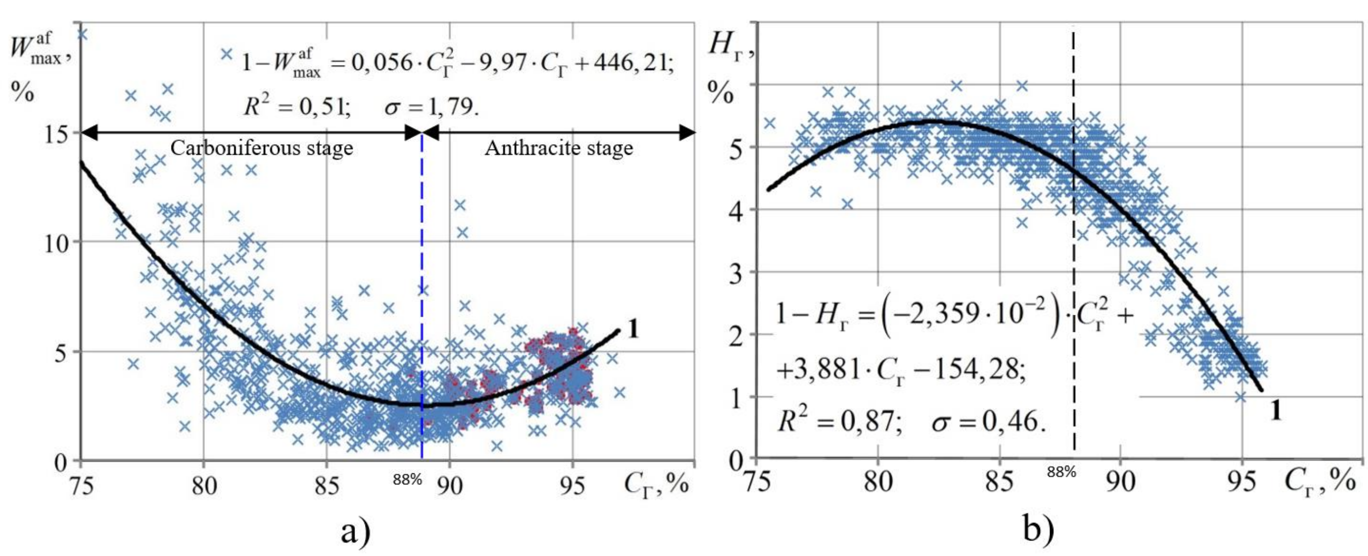

Figure 1. Dependence of the maximum moisture capacity of coal seams (a) and the hydrogen content in the combustible part of the fuel (b) on carbon for hard coals and anthracites.

Note: 1 – averaging curves; × – experimental data on maximum moisture capacity according to Handbook of the Quality [9]; ● – joint experimental data on coal moisture in coal seams according to catalogue [1] and carbon content in the combustible part according to data from Handbook of the Quality [9](a); ×- experimental data on hydrogen content according to reference book [10]; R^2, σ – coefficients of determination and standard deviation, respectively.

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The transition to the anthracite stage is characterized by a carbon content of more than 88% and an increase in moisture capacity after reaching minimum values (Figure 1a). The possibility of the formation and release of water at the anthracite stage is additionally indicated by a decrease in hydrogen content, also at a carbon content of more than 88% (Figure 1b). The ambiguous change in moisture content in coals allows the break in the curve, from decreasing to increasing, to be considered as the beginning of the anthracite stages of metamorphism [11].

Using only the indicators of the degree of metamorphic transformations, including V^{daf} and V_v^{daf}, it is impossible to reliably predict the gas content of coal seams in advance (Figure 2). Its quantitative values are random quantities, since the total amount of gases formed at individual stages of coal formation and their share subsequently released into the host rocks or onto the earth's surface are not taken into account.

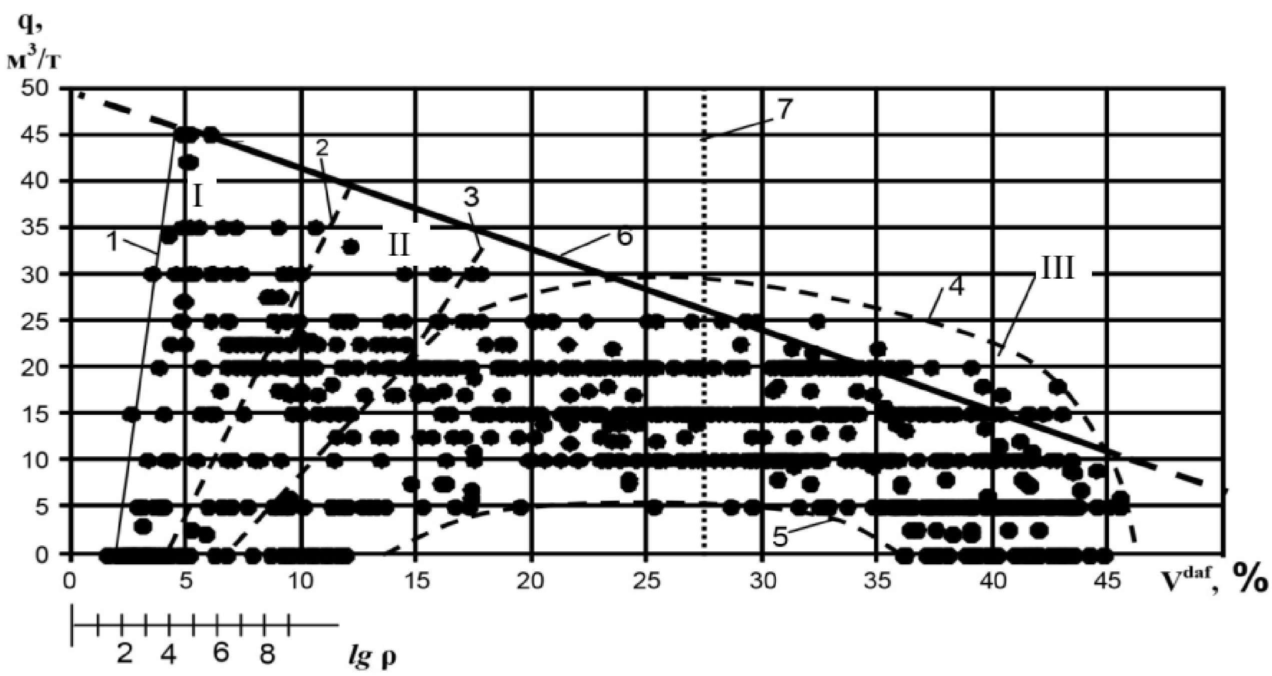

Figure 2. Dependence of gas content of Donbass mine seams (CH_4) on the yield of volatile substances (V^{daf}) according to statistical processing.

Note: 1,2 – respectively, the upper and lower boundaries of the change in the gas content of anthracites according to the joint data processing [12-14]; 3 – the expected lower boundary of the gas content during the transition from hard coals to anthracites; 4,5 – respectively, the upper and lower boundaries of hard coals; 6 – the averaging rectilinear boundary of the maximum values of gas content according to the results of data processing [13]; 7 – the conditional boundary of the division of hard coals by the degree of metamorphism into two parts, respectively, with an increase and decrease in gas content; I , II , III – gas content zones, respectively, for anthracites, transition and hard coals; ● – experimental data obtained during the joint data processing [1, 15].

Source: Antoshchenko and Shepelevich [16].

If conditions exist for the removal of all formed gases, then the methane content of coal seams is quantitatively equal to zero. If some part is retained, the maximum gas content of anthracite coal seams can reach 45 m³ per ton of dry ash-free combustible mass (m³/tdacm), and that of coal seams - 35 m³/tdacm. This difference indirectly confirms that at the anthracite stages, some additional methane is formed under the influence of elevated temperature.

The formation of carbon dioxide (CO_2) in significant quantities at the coal and anthracite stages is unlikely. This conclusion follows from the graph showing the decrease in the average oxygen content alongside a simultaneous increase in carbon in the combustible part of the fuel as the degree of metamorphic transformations increases (Figure 3).

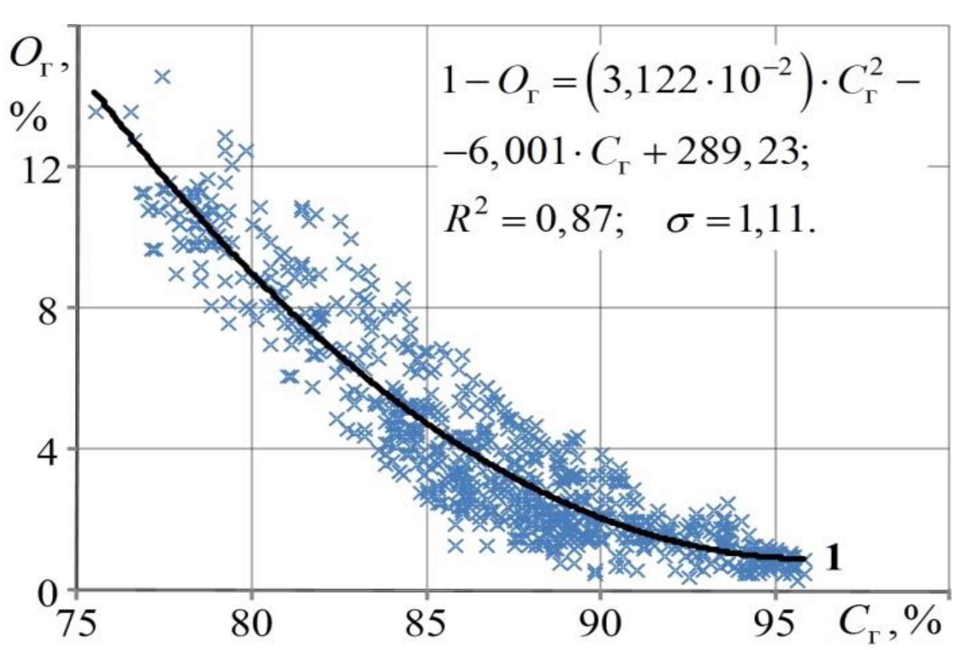

Figure 3. Dependence of oxygen content on carbon content for coal and anthracite seams.

Note: 1 - averaging curve; × - experimental data [10]; R^2 and σ - respectively, the coefficients of determination and the standard deviation.

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Table 1. Results of statistical processing of experimental data [10] on the quantitative increase in carbon content (C_{Γ}) and decrease in oxygen (O_{Γ}) at individual stages of metamorphic transformations.

Stages of metamorphic transformations of coals and anthracites	Average carbon content C_{Γ}, %			Average oxygen content O_{Γ}, %			The difference between the increase in the average carbon content and the decrease in oxygen, $\Delta C_{\Gamma}-\Delta O_{\Gamma}$, %	The main components of the combustible mass that took part in the formation of fluids	Vitrinite reflectance variation range R_a, %	Notes
	At the beginning of the stage	At the end of the stage	Increase in the content of ΔC_{Γ}	At the beginning of the stage	At the end of the stage	Reduction of content ΔO_{Γ}				
I	75.00	84.24	9.24	14.77	5.18	9.59	-0.35	O_{Γ}	0.40÷1.06	Transitional stage from brown coal to hard coal
II	84.24	87.83	3.59	5.18	2.89	2.29	1.30	O_{Γ}, H_{Γ}	1.06÷1.43	Carboniferous stage
III	87.83	92.75	4.92	2.89	1.12	1.77	3.15	O_{Γ}, H_{Γ}, S_{Γ}	1.43÷2.17	Transitional stage from hard coals to anthracites
IV-IX	92.75	97.23	4.48	1.12	0.39	0.39	3.75	O_{Γ}, H_{Γ}, S_{Γ}, N_{Γ}, C_{Γ}	2.17÷3.54	Anthracite stages
X	>97.23	-*	-*	<0.73	-*		>3.75	-*	>3.54	-

Note: * - No data.

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To establish a quantitative relationship between the increase in carbon content and the simultaneous decrease in oxygen in the combustible mass at each stage of metamorphic transformations, an empirical relationship was used [17]:

$$\bar{C}_{\Gamma} = 100 - 33{,}1 \cdot e^{-0{,}70 \cdot R_o}, \% \qquad (1)$$

where: $\bar{C}_{\Gamma}$and $R_o$are, respectively, the average carbon content in the combustible mass and the corresponding vitrinite reflectance index, %.

Dependence (1) is established on the basis of a generalization of experimental data obtained by different researchers. It practically functionally reflects the interdependence between the average carbon content and vitrinite reflection ($R^2 = 0.94$). This allows us to compare the growth of carbon content and the decrease in oxygen in the combustible mass at each characteristic stage of metamorphic transformations (Table 1). Such stages were preliminarily determined by the intersection points of the averaging pairs of curves of the dependences of the main components (oxygen, hydrogen, organic sulfur, nitrogen) as the metamorphic transformations intensify (growth C_{Γ}).

Each intersection point of two averaging curves indicates a change in the ratio between the content of the main components in the combustible (organic) mass, which undoubtedly affects the manifestation of hazardous properties of the mine layers. In this way, ten characteristic stages of metamorphic transformations of mine layers were established by the factor of changing the ratio between all the main components. Their boundaries are clearly defined by the average carbon content and the sum of the remaining components. Using the dependence of the average carbon content on the vitrinite reflectance (1), the ranges of change in the content of the main components at characteristic stages were also established by the vitrinite reflectance index (Figure 4).

This allowed us to separately consider the quantitative dependencies of the increase in the average carbon content and the decrease in oxygen at each characteristic stage of metamorphism by the factor of change in the elemental composition (Table 1). The first stage is, to some extent, a transitional one from brown coals to hard coals (R_o= 0.40 ÷ 1.06%). This follows from the upper limit of the vitrinite reflectance index (R_o= 0.60%) for brown coals during their classification by genetic and technological parameters [18]. At the first stage, the increase in the average carbon content (ΔC_{Γ}) was due exclusively to the removal of oxygen (ΔO_{Γ}). The average possible quantitative increase in $\Delta C_{\Gamma}$was 9.24%, and the decrease in ΔO_{Γ} occurred by 9.59%. The difference between these indicators (0.35%) is within the permissible limits of accuracy for determining the desired values; therefore, other main components (hydrogen, organic sulfur, nitrogen) did not participate in the formation of fluids at this stage of metamorphism.

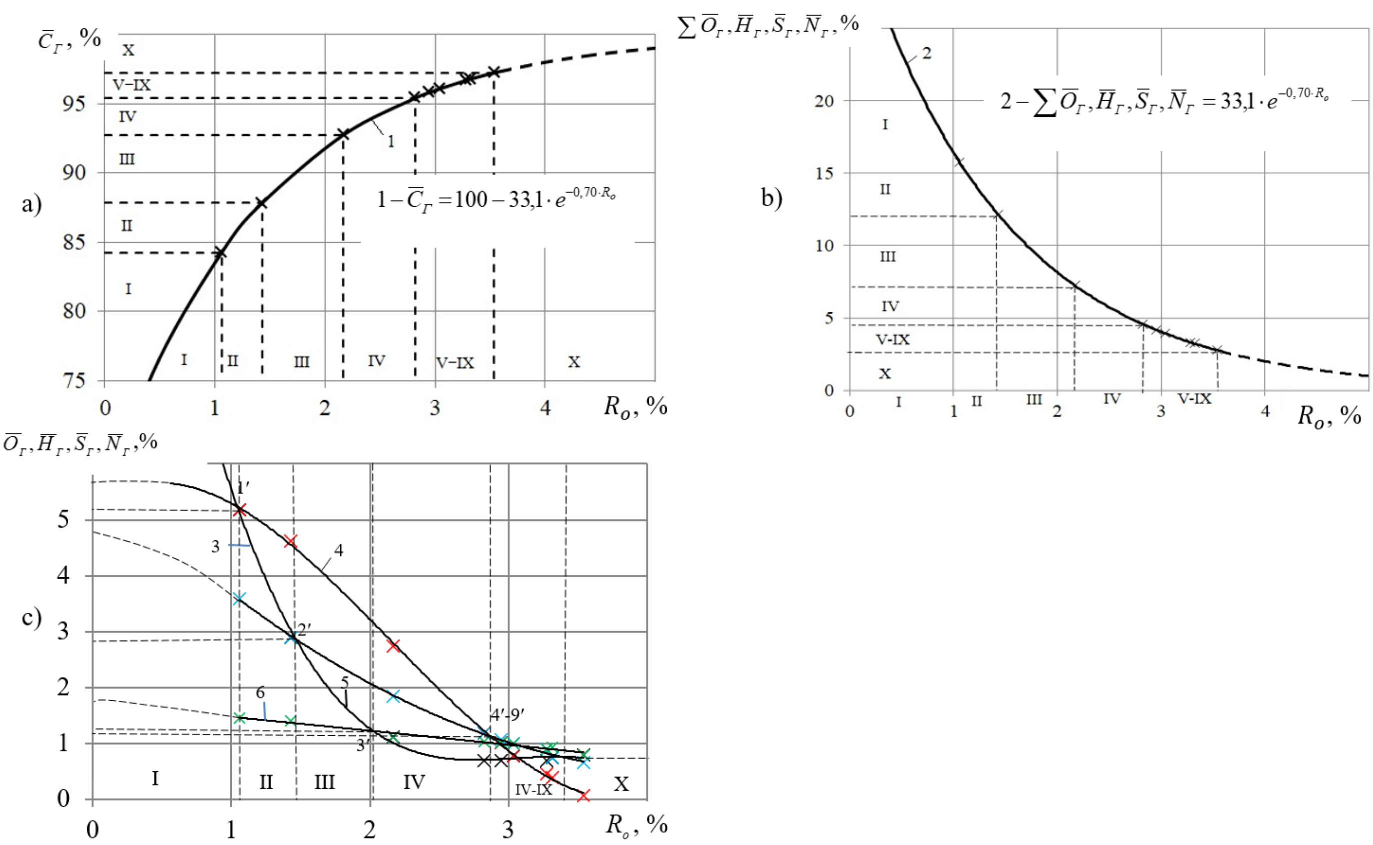

Figure 4. Dependences of the average carbon content (a), the sum of the remaining main components of the combustible part (b) and their individual change (c) at different stages of metamorphic transformations on the vitrinite reflectance.

Note: 1,2 – averaging curves, respectively, of the dependences of the carbon content ($\bar{C}_{\Gamma}$) and the sum of the remaining main components ($\sum \bar{O}_{\Gamma}, \bar{H}_{\Gamma}, \bar{N}_{\Gamma}, \bar{S}_{\Gamma}$); 3,4,5,6 – averaging curves, respectively, of the dependences of the individual content of oxygen ($\bar{O}_{\Gamma}$), hydrogen ($\bar{H}_{\Gamma}$), sulfur ($\bar{S}_{\Gamma}$) and nitrogen ($\bar{N}_{\Gamma}$); 1′,2′,3′,4′-9′ - intersection points of the averaging curves of the individual change in each main component of the combustible part; I, II, III, IV, V, VI, VII, VIII, IX, X - stages of metamorphic transformations, distinguished by the average content of the main components.

The second stage is characterized by the transformation of only coal seams (R_o= 1.06 ÷ 1.43%). At this stage, the growth of carbon content (ΔC_{Γ}= 3.59%) somewhat outpaces the reduction of oxygen (ΔO_{Γ}= 2.29%), which indicates the possible participation of other main components in the formation of fluids. Based on the graph of the experimental data (Figure 1b), the formation of fluids at this stage also occurred due to the reduction of the elemental content of hydrogen in the combustible mass. The upper limit of the carbon content at stage II (87.83%)

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corresponds to the minimum humidity (Figure 1a). This indicates that the next stage, Stage III, is a transition from hard coals to anthracites.

According to the classification of coal quality by genetic and technological characteristics [18] hard coals, by the upper value R_o, are classified if they are less than 1.6%. The lower limit for anthracites is the value R_o, equal to 1.4%. The absence of a specific boundary between hard coals and anthracites by the criterion R_o, confirms the presence of a transitional stage III between them. The range of change in the indicator R_o, by the factor of the ratio of average values of the main components for the transitional stage, was within 1.43 ÷ 2.17% (Table 1). According to the coal chemical map [19] by the brittleness criterion (output of dust class 1 ÷ 0 mm, cm^3), coals of the L and SA grades are classified as transitional from hard coals to anthracites. These grades correspond to values R_o, in the range of 1.50-2.20% [20]. In the case under consideration, close values of the ranges of change in the vitrinite reflectance index for the transition stage were obtained both by the change in the ratio of the elemental content of the main components and by the consumer and physical-mechanical (brittleness, dust yield) properties of coals. This indicates a fairly reliable determination of the boundaries of the transition stage from hard coals to anthracites using different criteria characterizing metamorphic transformations. An increase in the difference between the growth of carbon content at this stage (ΔC_Γ= 4.92%) and a decrease in oxygen (ΔO_Γ= 1.77%) to 3.15% (Table 1) indicates that carbon did not participate in the formation of fluids, including carbon dioxide. According to the graphs (Figure 4) and the results of statistical processing (Table 1), the main components of the combustible mass that participated in the formation of fluids were oxygen, hydrogen, and organic sulfur.

At anthracite stages from IV to IX, the oxygen content in the combustible mass decreased slightly from 1.12% to 0.68%–0.80%. Due to the small differences at these stages between the lower limits of the oxygen content (0.12%), its average value for all anthracite stages, which is 0.73%, was adopted for analysis (Table 1).

From a comparison of the significant increase in carbon content at the anthracite stages (ΔC_Γ= 4.48%) and the insignificant decrease in oxygen (ΔO_Γ= 0.39%) with its approximately constant and low average absolute value (0.73%), it follows that there is a practical absence of the possibility of the formation of common compounds containing carbon and oxygen.

Due to some slowdown in the growth of carbon content and the constancy of a small amount of oxygen content at anthracite stages IV-IX (Figure 4), the formation of carbon compounds with other main components (hydrogen, sulfur, and nitrogen) is possible. The formation of such compounds is confirmed by studies [19].

Stage X is characterized by a high carbon content (more than 97.23%). The sum of all other main components accounts for less than three percent. This indicates a decrease in the role of the elemental content of the main components in the formation of fluids that determine the hazardous properties of anthracite coal seams. For this reason, hazardous properties are determined, to a greater extent, by the amount of gas formed at previous stages of coal formation and preserved until the anthracite stages.

The share of nitrogen and sulfur in the formation of fluids at all stages of metamorphic transformations can be assessed based on the graphs showing the dependence of their content on carbon in the combustible mass (Figure 5).

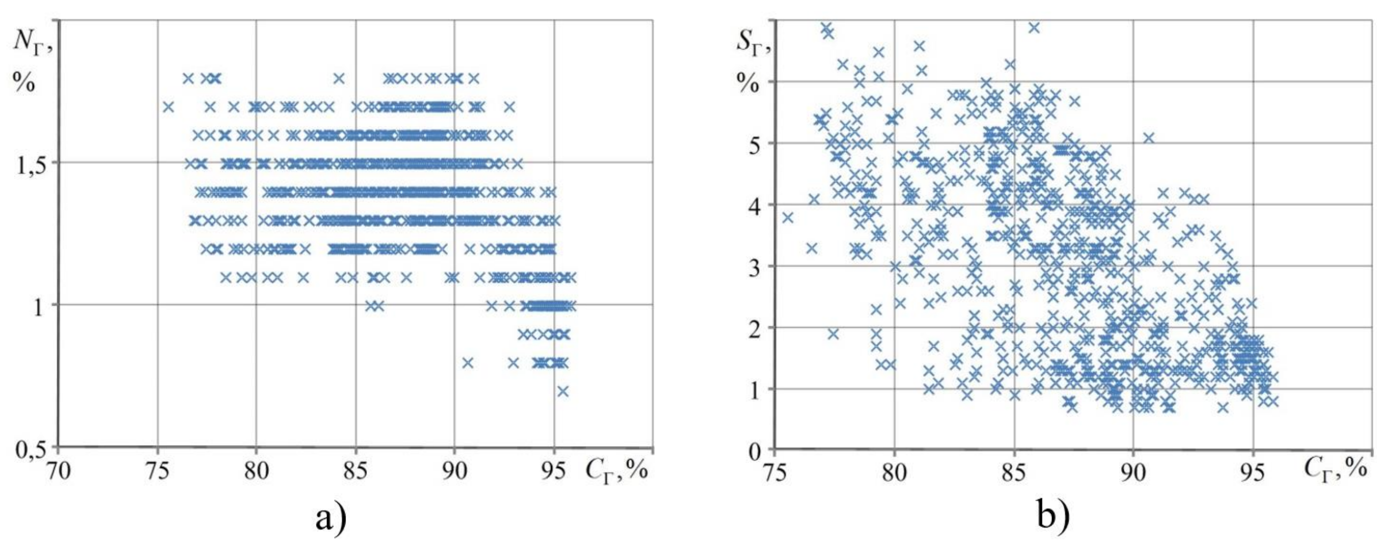

Figure 5. Dependence of nitrogen (a) and sulfur (b) content in the combustible part of the fuel on the carbon content for coal seams and anthracites.
Source: × - experimental data [10].

Changes in nitrogen and sulfur content are random for seams that have undergone the same metamorphic transformations according to the carbon content criterion.

The nitrogen content changed insignificantly and was within the limit of two percent, regardless of the carbon content. When the content reaches C_Γ more than 90% (at the anthracite stages), a certain tendency towards a decrease in nitrogen content is observed, and for some coal seams, its content is less than one percent (Figure 5a).

The nitrogen content in coals from different basins may differ significantly, but in most cases does not exceed three percent. There are anomalous cases of increased nitrogen content. For example, for the seam of ℓ_5 mine No. 3 "Kochegarka" in the Donetsk basin, the nitrogen content reached 4.2% [19] and for the seams of the Dolinskaya suite in the Karaganda basin, 8.6% [21]. Such fluctuations in the nitrogen content are obviously associated to a large extent with differences in the composition of the original material and the conditions of its accumulation. Regardless of the increase in the degree of metamorphic transformations (growth C_Γ), the sulfur content for the coal seams of the Donetsk basin fluctuates widely from 0.7 to 7.0% (Figure 5b).

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Unpredictable relationships between the carbon content and nitrogen and sulfur do not affect the accuracy of controlling the carbon (about one percent) of the sum of all other components of the combustible (organic) mass. For this reason, the equality:

$$C_{\Gamma} = 100 - \sum O_{\Gamma}, H_{\Gamma}, N_{\Gamma}, S_{\Gamma}, \% \qquad (2)$$

Remains true for any mine seam, regardless of the degree of its metamorphic transformations and the different ratios between the main components.

Different ratios between the main components can significantly affect the manifestation of hazardous properties of coal seams. The high probability of such an effect is indicated by a significant change in the consumer qualities of coals with different oxygen contents. With an equal degree of metamorphic transformations of coal seams, coals with a lower oxygen content are distinguished by higher sintering, solubility in hydrocarbons, and heat of combustion. Taking the yield of volatile substances as the only criterion for the degree of metamorphic transformations, it is impossible to establish different oxygen contents with the same reference values V^{daf}. For this reason, the terms "reduction" and "oxidation" of fossil coals were introduced to characterize the consumer qualities [19]. Differences in consumer properties were explained by genetic differences in coal types. It was proposed to distinguish four types of coal: type "a" - slightly reduced coals; type "b" - intermediate; type "c" - reduced; type "bb" - highly reduced. It was believed that the mineral part of coal is also genetically related to the composition and properties of the organic matter.

According to the developed methodology [19], the type of hard coals by reduction is determined based on a comparison of the yield of volatile substances (V^{daf}) and the oxygen content in organic matter (O^{o}) for two coal seams. Additional indicators of reduction are considered to be the indicators of total sulfur (S_t^d) and the main oxides in the ash. The coal seams, which with approximately equal values of V^{daf}, the oxygen content and the indicators of the content of total sulfur and main oxides in the ash (Fe_2O_3, CaO, MgO) also differed slightly from each other were considered to be of the same type.

The different types mainly included coal seams with coals, the yield of volatile substances of which was approximately the same, but there were some differences in the oxygen content.

To date, the permissible differences in the values of the index have not been determined V^{daf}, At which two compared mine layers must be classified as belonging to the same degree of metamorphic transformations. The permissible differences in oxygen content, at which mine layers must be classified as of the same or different types, have also not been established.

4. Conclusions

Preliminary analysis indicated that the reasons for the existing uncertainties in establishing the genetic types of coals may be due to provisions adopted during the development of the methodology that are insufficiently scientifically substantiated [19]. The main ones are:

- The yield of volatile substances, taking into account the methods of determination, does not correspond to the classical (generally accepted) characterization of metamorphism as a change in the composition and properties of coals in the process of geological transformations of coal seams [22-24].
- Metamorphism and thermal decomposition of coals are different stages of their transformation. The quantitative and qualitative composition of the volatile substances formed during thermal decomposition has no direct relation to the previously occurring metamorphic processes in natural conditions, in which part of the gaseous products had already been removed. Thermal destruction is the result of a new (next) artificial stage of transformation of the original organic matter raised to the earth's surface [16].
- Using only the yield of volatile substances as an indicator of metamorphism, it was not possible to confirm the fulfillment of Hilt's rule for many mine seams [19].
- The organic mass consists of five main components. When establishing the reduction of coals and the uniformity of coal seams, only the oxygen content is taken into account [19], which is genetically related to the total individual content of carbon, hydrogen, nitrogen, and sulfur. In this case, organic sulfur is not considered a genetic component of the organic mass.
- Mineral impurities, roughly determined by the ash yield, are not indicators of metamorphic transformations of coals due to their preliminary enrichment during the preparation of analytical samples. To partially eliminate the influence of mineral impurities on the consumer qualities of fuel, coals are enriched until the ash yield is, as a rule, less than 10% [8].

It follows from the above analysis that the yield of volatile substances and the oxygen content in the organic mass do not unambiguously characterize either the types of coal seams by the supposed reduction of coals or the manifestation of hazardous properties during mining operations. In order to establish the true causes of changes in the properties of coal seams and their tendency to manifest hazardous phenomena, taking into account the genetic interdependence between the main components, it is necessary to consider individual relationships between them for each coal seam, and not limit ourselves to determining the content of only oxygen and total sulfur.

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