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METAMORPHISM OF COALS, STAGES OF TRANSFORMATION OF COAL SEAMS AND THEIR HAZARDOUS PROPERTIES

Rudniev Ye.S., Antoshchenko M.I., Filatieva E.N., Filatiev M.V, Tarasov V.Yu.
Volodymyr Dahl East Ukrainian National University

Abstract. The volatiles yield, taking into account the methods of determination, does not correspond to the classical (commonly accepted) parameter of metamorphism as a composition and properties change of coals in the process of geological transformations of coal seams. One indicator, even the most versatile one, unable reliably and comprehensively distinguish the change of the ratio of all components for the organic matter and its properties under the influence of metamorphic processes. The research methodology is based on the statistical processing of known experimental data obtained from regulatory documents for almost all coal seams of the Donbas and the Lviv-Volyn basin. As an alternative criterion of metamorphism in relation to the volatile release, the carbon content in the organic (combustible) mass of fuel is taken. Selection of an alternative indicator is based on the carbon content indicator has some advantages over the other indicators for prediction of the hazardous properties manifestation of coal seams. According to the correlation tightness between the indicators of the volatile release and the carbon content with the basic elements of the organic (combustible) mass, it was concluded that every of the indicators considered corresponds to a reliable description of a certain aspect of the metamorphic transformations of coal seams. The studies conducted have shown a significant multiple factor influence of metamorphic processes, both on the change of the element compound of the combustible (organic) mass of the initial coal matter, as well as on its physical and mechanical properties. It is not possible to describe such a range of changes for the compound and properties of coals during the metamorphism of coal seams, as it is presented in regulatory documents, by means of a single indicator, i.e. volatile release during coal thermal destruction. This indicates necessity to improve the regulatory framework for the safe conduct of mining operations. While making the necessary corrections, it is proposed to take into account the features of changes established within the element compound as well as properties of coals during metamorphic transformations of coal seams.

Keywords: coal, metamorphism, coal seams, indicators, carbon content, sulfur, combustible mass, mining, safety, regulatory framework, improvement.

Introduction. Nowadays, the problems associated with the manifestation of the hazardous properties of coal seams during mining operations have not been drastically resolved. This is proved by accidents at coal mines that take place over and over again, which are associated with outbreaks and explosions of gas in mine workings, sudden coal and gas emissions in stopes and development headings, outbreaks of coal spontaneous combustion and some other manifestations of hazardous properties. Due to the situation, studies aimed at improvement of the regulatory framework for the safe conduct of mining operations are relevant for the coal mining industry all over the word.

The formation and manifestation of hazardous properties of coal seams is largely, according to the requirements of regulatory documents [1-7], are determined by metamorphic stage of transformation of coal.

According to the officially accepted definition [8], metamorphism refers to the gradual conversion of brown coal (lignite) into bituminous coal (black coal) as well as anthracite as a result of the transformation of chemical composition as well as structure and physical properties of coal underground, mainly under high pressure and temperature.

In the general, such transformations are relevant not only to coal seams, but also to all metamorphic rocks. The essence of such transformations is changing the mineral composition or size and structure of grain aggregates, without significant changes of chemical composition (except for the content of H_2O and CO_2) under the influence of fluids, temperature and pressure [9].

Metamorphic rocks were formed due to the transformation of igneous or sedimentary rocks under the influence of high pressure, temperature and combustible gas-water solutions. Being physical bodies these rocks could be described by dense, elastic, strength, thermal, electrical, magnetic, radiation and other properties, which primarily depend on their mineral composition and macrostructure (structure and texture) [10].

According to the commonly accepted notions [8-10] the degree of metamorphic transformations depends on the changes of the element compound of coal and its physical and mechanical properties. Contrary to this definition of metamorphism in regulatory documents [1-7] the main criterion of metamorphic transformation of coal and forecast of hazardous properties of coal seams is the volatile release during coal pyrolysis. According to the methods of determining for the mass (V^{daf}) and volumetric (V_V^{daf}) volatile release during pyrolysis [11-13], these indicators, taking into account the methods of their determination, unable to directly and simultaneously describe changes of element compound as well as properties of coal in the process of metamorphic transformations of coal seams. In addition, metamorphism and thermal decomposition are different stages of their transformation. Quantitative and qualitative compound of volatile formed during thermal decomposition is not directly related to metamorphic processes that previously occurred in natural conditions, in which part of the volatiles have already been removed. Thermal destruction is a consequence of a new (next) artificial stage of transformation of the mined original organic matter [14].

Methods. Idea. The volatile release, taking into account the methods of determination, does not correspond to the classical (commonly accepted) feature of metamorphism as a change of the compound and properties of coal in the process of geological transformations of seams. Single indicator, even the most versatile, unable reliably and comprehensively describe the change of the ratio of all components of organic matter and its properties under the influence of metamorphic processes. If the change of the ratio for all basic elements of organic matter are considered comprehensively the particular stages of metamorphism might be distinguished. The stages of metamorphism able to describe the tendency of seams to manifest some dangerous properties during mining operations.

The aim is to prove the necessity of regulatory documents improvement for safe mining, accumulated over several decades as well as based on the experience of fuel quality, by means of using indicators that directly describe the change of element compound and properties of coal in the process of metamorphic transformations of coal seams at particular stages.

This allows predicting the tendency of coal seams to the manifestation of their hazardous properties, which take place due to change of element compound ratio between basic elements of organic matter within the established stages of metamorphic processes.

The research methodology is based on statistical processing of known experimental data obtained for almost all coal seams in the Donbas and Lviv-Volyn basin.

A piece of data about the compound of the basic elements in the organic matter is presented within the geological and coal chemical map [15] as well as within the regulatory documents and reference list, further, regulatory documents on the quality of coals [16, 17] contain the data that concerns basic elements of the coal combustible mass.

As an alternative criterion of metamorphism in relation to the volatile release, the indicator of carbon content $C^o(C_г)$ within the organic (combustible) mass of fuel is accepted. Selection of an alternative indicator is based on the advantages carbon content indicator has for predicting the manifestation of dangerous properties of coal seams [18]. The advantages are as follows:

– the carbon content directly represents the change of the organic (combustible) matter compound for in metamorphic processes;

– increasing influence of metamorphism provides increasing of carbon content unilaterally;

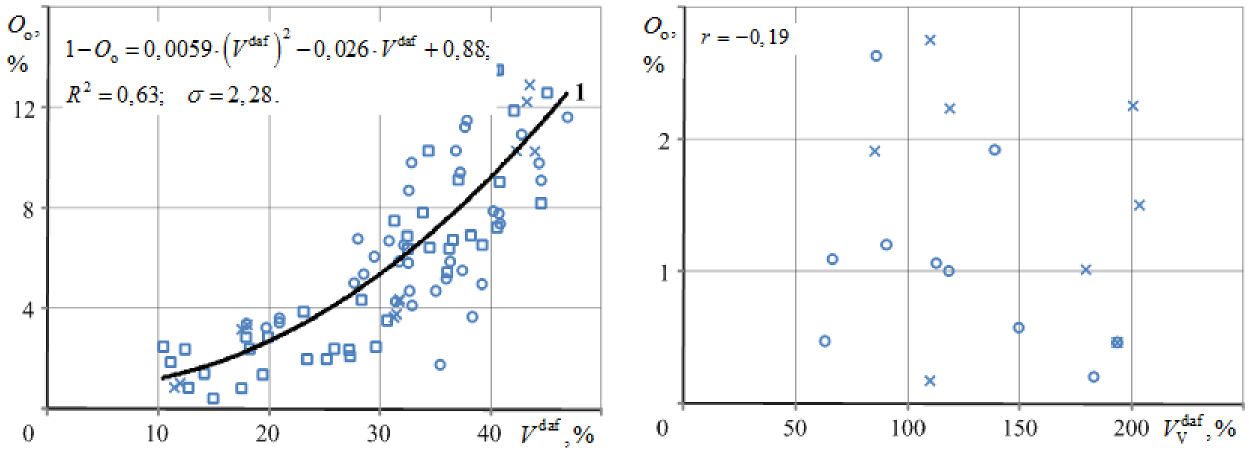
– the carbon content with high accuracy controls amount of the remaining basic elements of the organic (combustible) mass, i.e. hydrogen, nitrogen, oxygen as well as organic sulfur.

According to the correlation tightness between V^{daf} as well as $C^o(C_г)$ and the basic elements of organic (combustible) mass it is obvious that every analyzed indicator corresponds to a reliable description of a particular aspect of metamorphic transformations of coal seams associated with changes of the basic elements of organic (combustible) mass and some utility properties of fuel.

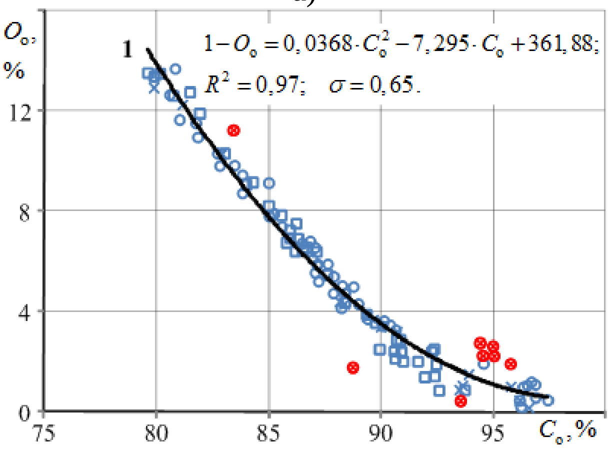
Results and discuccion. Given that V^{daf} and $V_V{}^{daf}$ do not directly represent the element compound for the basic components, similar dependences of these components on carbon content were considered as well.

Contrary to V^{daf} and $V_V{}^{daf}$, the carbon content more reliably determines the oxygen content at all stages of the conversion of both coal and anthracite (Fig. 1). This follows from the weak correlation between oxygen content and V^{daf} (Fig. 1, a) as well as its absence for $V_V{}^{daf}$ (Fig. 1, b). The dependence of the reduction of oxygen content with increasing of metamorphic transformations (C_o increasing) at all stages is described (Fig. 1, c) by almost functional connection (coefficient of determination R^2=0.97).

It should be noted that the correlation tightness was not affected even by the additional involvement of data on oxygen and carbon content for 41 coal seams (the whole considered set of 109 coal seams), that according to [15] do not comply with the Hilt’s law. According to this law, increasing of the stratigraphic depth for coal seams leads to decreasing of volatile release, i.e. the degree of metamorphic transformations is increased. Using the V^{daf} as a metamorphism index for a significant part of coal seams, the discrepancy of the V^{daf} values compared to those ones defined by means of the Hilt’s law was revealed. It was previously established that using C_o as an indicator of metamorphism provides elimination of inconsistency for the Hilt’s law.



a) b)



c)

1 – average of curves;

×, ○, □ - experimental data, for the same type coal and anthracite, different coal types and anthracite, for coal seams with different following to the Hilt's law respectively;

⊗ - experimental data with the maximum deviation from the average of curves;

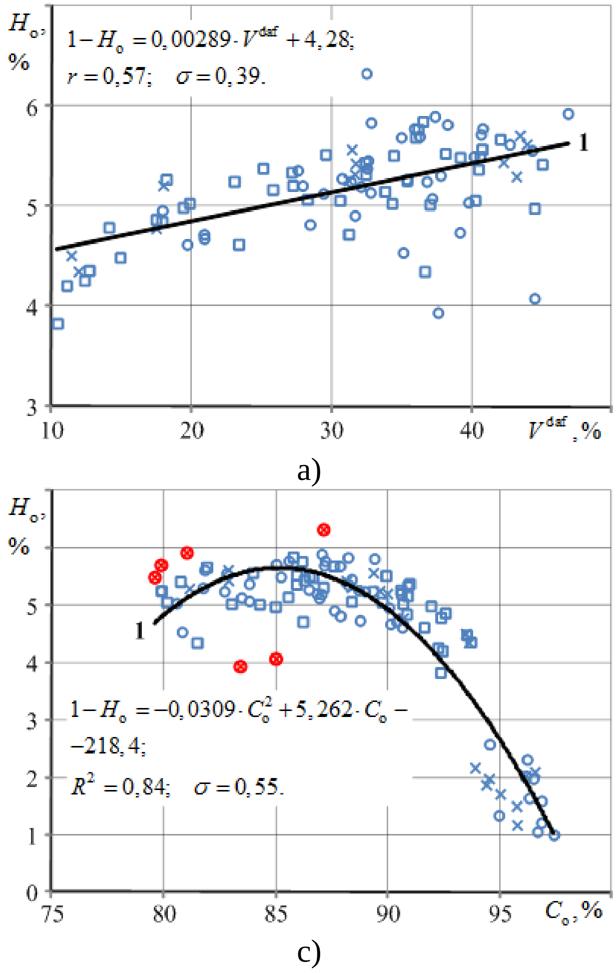
R^2 - coefficients of determination;

σ - standard deviation.

Figure 1 – Dependence of oxygen content in organic matter (O_o) from volatile release per mass unit (V^{daf}) (a), from volatile release per volume unit (V_V^{daf}) (b) from carbon (C_o) content (c) according to [15]

Indicators V^{daf} and V_V^{daf}, compared with C_o, describe the change of hydrogen content (H^o) in a variety of ways as the influence of metamorphic processes is increased. For indicators V^{daf} and V_V^{daf} a weak directly proportional dependence (Fig. 2, a and b) was established. The correlation coefficients (r) in both cases equal 0.57. The change of hydrogen content depending on carbon content occurs according to the nonlinear relationship (Fig. 2, c). The coefficient of determination (R^2=0.84) described the tight correlation. This indicates that V^{daf} and V_V^{daf} represents, compared to carbon content, various aspects of metamorphic transformations of bituminous coal and anthracite, which are necessary to establish their utility properties.

The change of nitrogen content in the organic mass barely depends on both the indicators of V^{daf} and V_V^{daf} as well as carbon content (Fig. 3). For all the cases quite low correlation coefficients (r=0.09 for V^{daf}), (r=0.54 for V_V^{daf}) and (r =-0.17 for C_o) are observed. The nitrogen content at all stages of metamorphism, as a rule, varies slightly within the range of 0.83÷2.01%. For some cases, a peak value was observed, when the nitrogen content (4.23%) in the coal of coal seam l_5 of mine #3 "Kochegarka" more than doubled the maximum values for other coal seams (Fig. 3, a and c). Further, considering the interdependence between the indicators of metamorphism V^{daf} and C_o, abnormal deviations of the indicators for coal seam l_5 from the average values of curve (1) were not found (Fig. 4, a). There is also no definite relationship between carbon content and volatile release per volume unit (Fig. 4, b).



1 – average of curves and lines;

×, ○, □ - experimental data, for the same type coal and anthracite, different coal types and anthracite, for coal seams with different following to the Hilt's law respectively;

⊗ - experimental data with the maximum deviation from the average of curves;

R^2, r - coefficients of determination and correlation respectively;

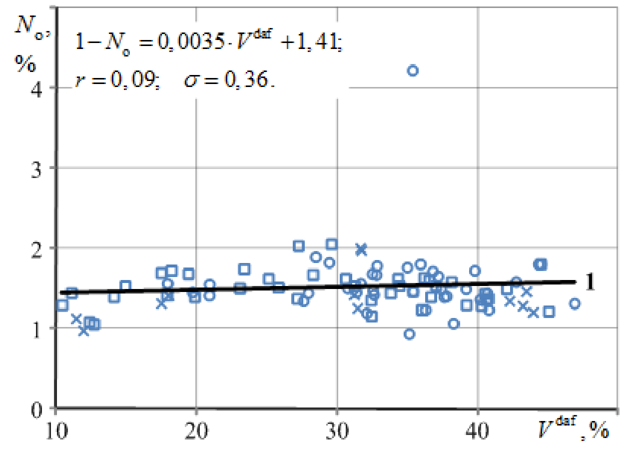
σ - standard deviation.

Figure 2 – Dependence of hydrogen content in organic matter (H_o) from volatile release per mass unit (Vdaf) (a), from volatile release per volume unit (V_V^{daf}) (b) and carbon (C_o) content (c) according to [15]

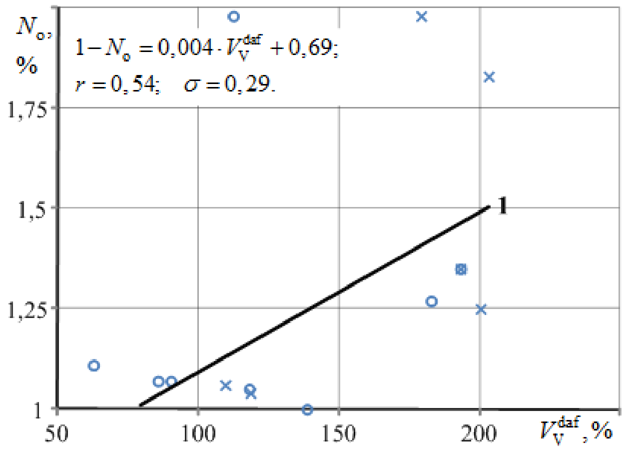
These results indicate that during determination of the bituminous coal types and anthracite by means of their reduction, factors except the oxygen content that determines the change of the ratio for the content of other basic elements are not taken into account. This is confirmed not only by abnormal deviations of nitrogen content from averaging lines (Fig. 3, a and c), but also by significant deviations of oxygen and hydrogen content from average of curves (Fig. 1, c and 2, c).

The maximum deviations from the average of lines for the values of nitrogen, oxygen, hydrogen and the corresponding contents of other components are given in Table 1. Regardless of the individual relationships between the elements of organic matter (C_o, O_o, H_o, N_o), their sum for all the cases nearly equals 100%. For this reason, even for abnormal content of a single or several elements in the organic mass, the carbon content will nearly functionally control the sum of other basic elements:

$$C_o = 100 - \Sigma O_o, H_o, N_o, \%. \quad (1)$$



a)



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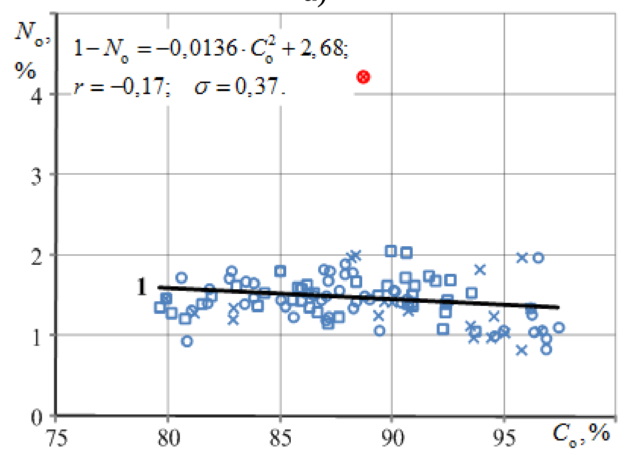


c)

1 – average of lines;

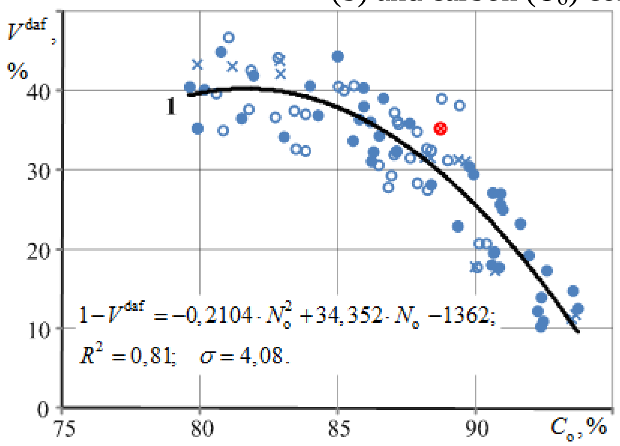
×, ○, □ - experimental data, for the same type coal and anthracite, different coal types and anthracite, for coal seams with different following to the Hilt's law respectively;

⊗ - the maximum deviation of the nitrogen content from the averaging line in the organic mass of the coal seam l_5 for the mine #3 "Kochegarka";

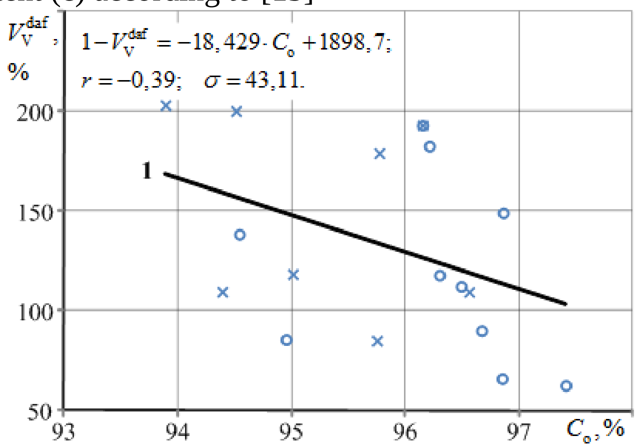
r - correlation coefficients;

σ - standard deviation.

Figure 3 – Dependence of nitrogen content in organic matter (N_o) from volatile release per mass unit (Vdaf) (a), from volatile release per volume unit (V_V^{daf}) (b) and carbon (C_o) content (c) according to [15]



a)



б)

1 – average of lines; ×, ○ - experimental data [15] for the same type and different type coal seams respectively; ● - experimental data V^{daf} [15], which were used to establish following of coal seams metamorphism to the Hilt's law;

⊗ - experimental data of indicators [15] for coal seam l_5 of mine #3 "Kochegarka";

R^2, r - coefficients of determination and correlation respectively; σ - standard deviation.

Figure 4 – Interdependence between the carbon content in the organic mass (C_o) and volatile release per mass unit (V^{daf}) during thermal decomposition of bituminous coal (a) as well as volatile release per volume unit (V_V^{daf}) during thermal decomposition of anthracites (b)

Table 1 – Information on the element compound of the main components of the organic mass for coal seams with maximum deviations of experimental data [9] from the average of lines

Elements of organic matter	Mine or sample number	Coal seam index	Maximum deviations from the average of lines		Element compound for referenced organic mass,%				The total compound of the elements,%	Note
			absolute, %	relative * fractions	C_o	O_o	H_o	N_o		
N_o	#3 "Kochegarka"	l_5	+2.76	+0.65	88.70	1.80	5.27	4.23	100.00	different types of coal
	n. after Rumiantseva	m_3	-0.87	-0.41	90.61	2.14	5.21	2.04	100.00	Hilt's law
	#10 n. after Artema	l_8	-1.06	-2.35	93.52	0.45	4.49	1.54	100.00	Hilt's law
	n. after Melnykova	l_6	+1.82	+0.16	83.39	11.27	3.94	1.40	100.00	different types of bituminous coal
	#4104	h_8	+1.59	+0.58	94.39	2.76	1.87	0.98	100.00	the same types of anthracite
O_o	#4054	k_2	+1.13	+0.50	94.51	2.26	1.98	1.25	100.00	the same types of anthracite
	#4387	k_5	+1.27	+0.57	95.01	2.24	1.71	1.04	100.00	the same types of anthracite
	#4306	h_8	+1.16	+0.60	95.75	1.92	1.50	0.83	100.00	the same types of anthracite
	№ 4236	l_2	+0.81	+0.42	94.54	1.93	2.58	1.00	100.05	different types of anthracite
	#4508	k_5	+1.70	+0.64	94.95	2.64	1.34	1.07	100.00	different types of anthracite
H_o	#2 "Trudovska"	l_3	+0.90	+0.16	79.60	13.54	5.50	1.36	100.00	Hilt's law
	#1/2	l_2	+1.03	+0.18	79.87	12.95	5.71	1.47	100.00	the same type of bituminous coal
	#1/2	k_8	+0.85	0.14	81.01	11.67	5.93	1.32	99.93	different types of bituminous coal
	n. after Melnykova	l_2	-1.52	-0.37	84.98	9.15	4.08	1.81	100.02	different types of bituminous coal
	n. after Melnykova	l_6	-1.63	-0.41	83.39	11.27	3.94	1.40	100.00	different types of bituminous coal
	#19/20	l_7^1	+0.85	+0.13	87.12	5.86	6.33	1.69	101.00	different types of bituminous coal

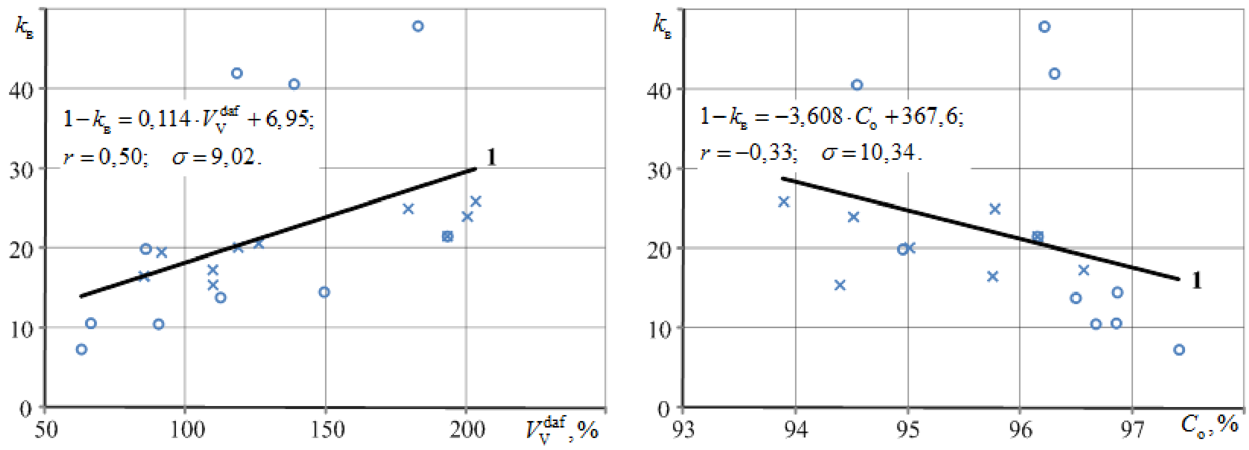
* - related to the element considered

Based on the obtained result the discrepancy between the oxygen content and the degree of metamorphic transformations of organic mass within the coal chemical map

[9] is determined, which involves a change of the element compound and volatile release. For this reason, ambiguous results have been obtained to determine the types of reduction for both bituminous coal and anthracite.

Changing the relationship between the element compound of organic matter is one of the aspect of metamorphic coal seams transformations. Along with these processes at different stages of metamorphism some changes of the properties for bituminous coal and anthracite associated with changes of their internal structure as well as physical and chemical parameters take place. At the stage of isometamorphic anthracites, the element's ratio of the organic mass components, including the oxygen content, barely determines the physical and chemical properties.

This explains the absence of correlations between the reduction coefficients with the volatile release per volume unit (V_V^{daf}) and as well as carbon content (Fig. 5). The index $k_в$ describes the molecular ratio of hydrogen and oxygen content in the gases derived from the thermal decomposition of anthracites. Identification of these gases during determination of the reduction coefficient barely contributed to the accuracy of index $k_в$ determining. The index $k_в$ also barely depends on C_o (Fig. 5, b).



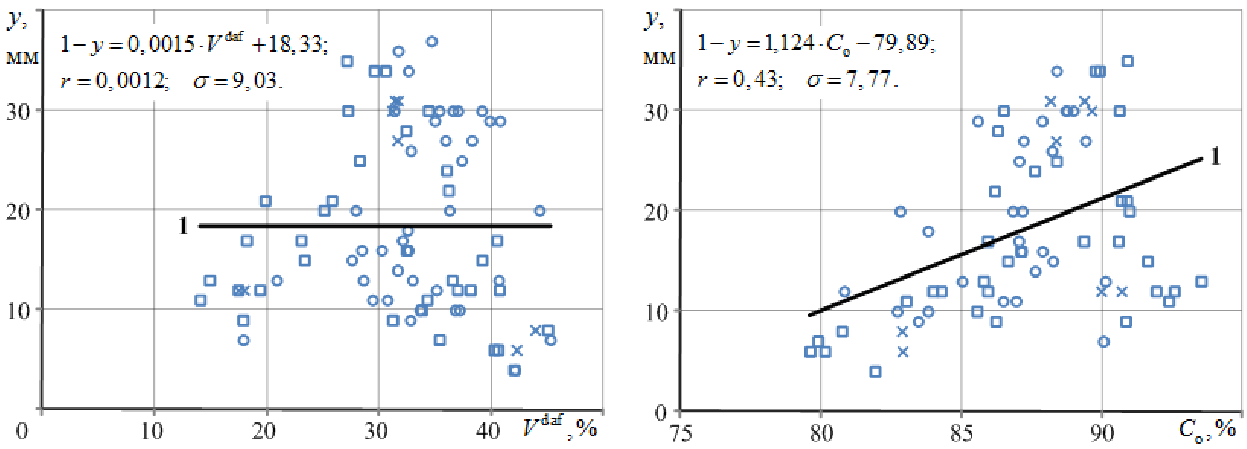
a) b)

1 – average of lines; ×, ○ - experimental data for
the same and different types of anthracites [15] respectively, which were used to determine $k_в$;
r - correlation coefficients; σ - standard deviation.

Figure 5 – Dependence of the reduction coefficient for determination of the types for isometamorphic anthracites ($k_в$) on volatile release per volume unit (V_V^{daf}) (a) and carbon content (C_o)(b)

Indicators V^{daf} and C_o does not properly describe a property of coal named thickness (y) of the plastic layer (Fig. 6).

The advantage of V_V^{daf} over C_o is the accuracy of the description for the physical and electromechanical properties of anthracites (Fig. 7 and 8). This indicates that for prediction of the hazardous properties for coal seams, in addition to the content of the basic elements as well as ratio of the basic elements for organic matter, it is necessary to consider the indicators that describe the physical and mechanical properties of bituminous coal and anthracite.



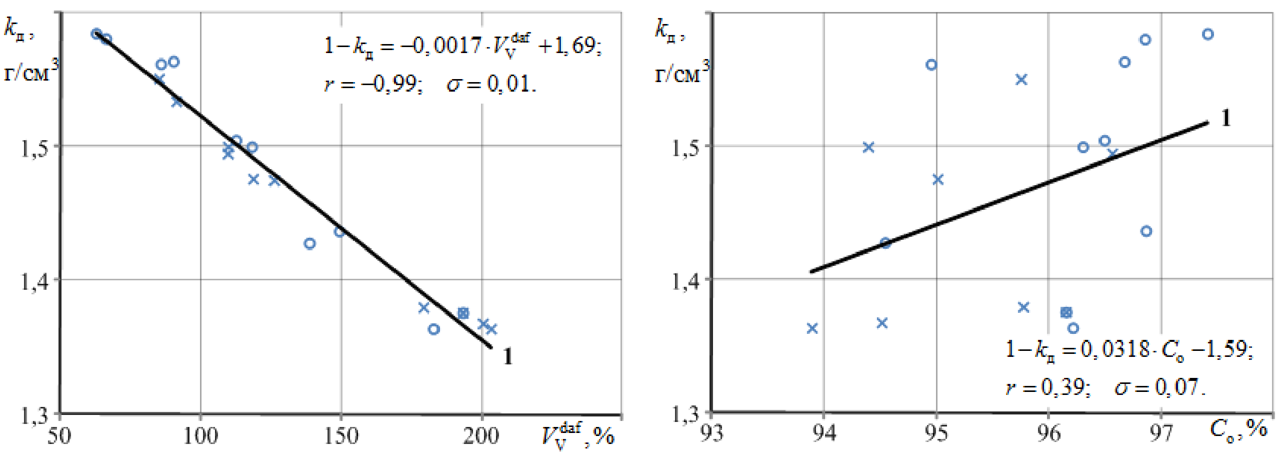
a) b)

1 – average of lines; ×, ○ - experimental data [15];
r - correlation coefficients; *σ* - standard deviation.

Figure 6 – Dependence of the thickness of the plastic layer (*y*) on volatile release per mass unit (V^{daf}) (a) and carbon content (C_o) (b)

Basically, some discrepancies in the determination of the hard coal and anthracite types by the difference of oxygen content were caused by the fact that the organic mass along with carbon, hydrogen, nitrogen and oxygen, the content of organic sulfur (S_o) was not considered. The content of organic sulfur is related to the organic mass [11] solely, thus during determination of the hazardous properties of coal seams, its content must be considered together with other basic elements C_o, O_o, H_o, N_o. The total content of the basic elements in the organic mass (Σ C_o, H_o, N_o, O_o, S_o) is about 100%. In this case, the ratio of the basic elements could not be calculated by means of equation (1), but based on the dependence as follows:

$$C_o = 100 - \Sigma O_o, H_o, N_o, S_o\%. \tag{2}$$

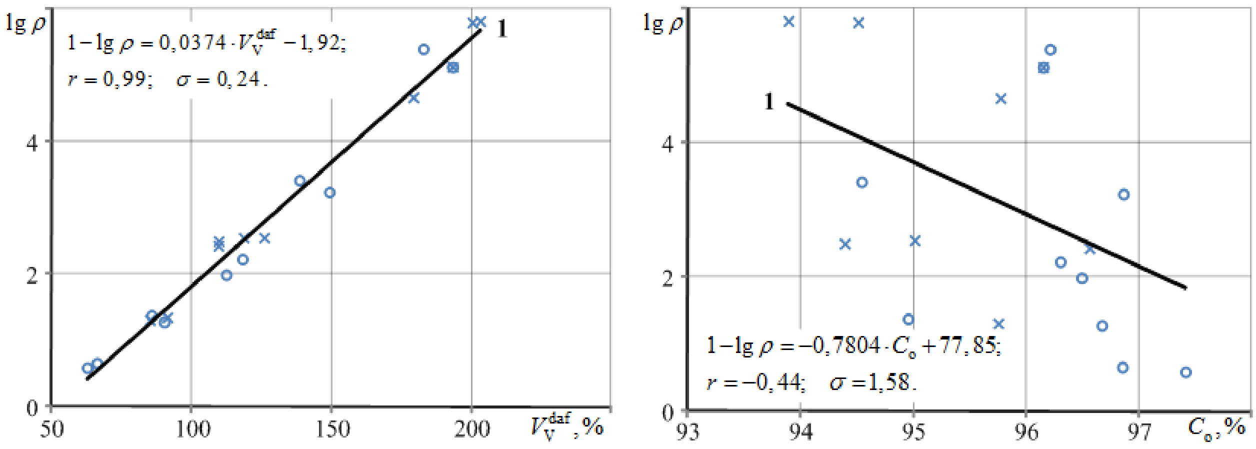


a) b)

1 – average of lines; ×, ○ - experimental data [15];
r - correlation coefficients; *σ* - standard deviation.

Figure 7 – Dependence of the specific weight of the anthracites organic mass ($k_д$) on volatile release

per volume unit (V_V^{daf}) of (a) as well as carbon content (C_o) (b)

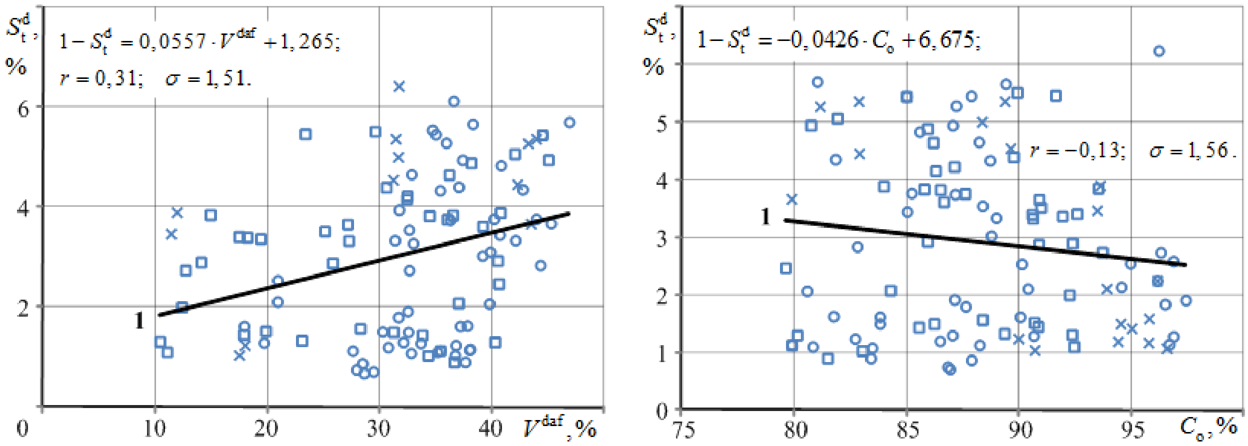


a) b)

1 – average of lines; ×, ○ - experimental data [15];
r - correlation coefficients; *σ* - standard deviation.

Figure 8 – Dependence of the logarithm of electrical resistance (lg *ρ*) on the volatile release per volume unit (V_V^{daf}) (a) as well as carbon content (C_o) of (b)

The possible redistribution of the basic elements ratio for the organic mass during their simultaneous consideration with organic sulfur could be estimated based on the total content of sulfur (Fig. 9). Total sulfur does not belong to the indicators of metamorphic transformations of coal seams, which is confirmed by the absence of correlations between both V^{daf} and C_o. Its content at all the stages of metamorphic transformations of the analyzed set of coal seams was predominantly within the range of 1.0÷6.0%.



a) b)

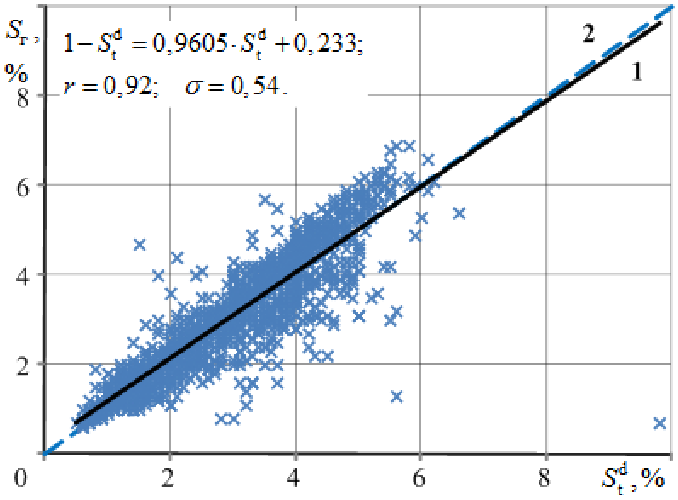
1 – average of lines; ×, ○ - experimental data [15] for the same types of coal and anthracite, different types of coal and anthracites, coal seams with different following with the Hilt's law respectively; *r* - correlation coefficients; *σ* - standard deviation.

Figure 9 – Dependence of total content of sulfur (S_t^d) on volatile release (V^{daf}) (a) as well as carbon content (C_o) (b).

The total content of sulfur in the coal seams of other basins ranges from 0.2 to 10% [11]. Sulfur is a harmful part of fuel. During coal burning, it is released as SO_2

compound, polluting and poisoning the environment and causing corrosion of metal surfaces. It is also reduces the heat of combustion of fuels, and during coking process gets into coke, deteriorating its properties as well as metal quality. The selection of coal application depends partially on the total content of sulfur as well as its forms. For this reason, the total content of sulfur is the most important indicator of coal quality and it was used for these purposes to make up a coal chemical map [15].

Total content of sulfur (S_t^d) largely determines the sulfur content in the combustible part of the fuel (Fig. 10). In turn, based on the preliminary enrichment of analytical samples as well as ash release less than 10%, approximate equality of S_r and S_o could be stated.



1 – average of line; 2 - bisector of the grid; × - experimental data [16, 17];
r - correlation coefficients; *σ* - standard deviation.

Figure 10 – Interdependence of sulfur content in combustible part of fuel (S_r) and total content of sulfur (S_t^d).

Organic sulfur as a species has not been studied enough, because there are no methods for organic compounds extracting from coal [11]. Organic sulfur is determined by the following expression:

$$S_o = S_t^d - \left(S_p + S_{SO_4}\right), \%, \quad (3)$$

where S_p - fraction of the total sulfur content for coal, which is part of the inorganic mass of coal in the form of metal disulfides (pyrite and marcasite),%; S_{SO4} - sulfate sulfur fraction, which is part of the inorganic mass of coal,%.

The fraction of sulfate sulfur (S_{SO4}) is pretty small and usually equals 0.1÷0.2% [11]. For this reason, the content of organic sulfur is determined mainly by the difference between total sulfur content and sulfur S_p , which is a fraction of the inorganic part of the fuel. Given that the main fraction of inorganic mass of coal is its non-combustible part, and the ash content of analytical samples does not exceed 10%, the values of organic sulfur content will be as close as possible to the sulfur content of the fuel’s combustible part. In these circumstances, changes of the content for the basic elements of the fuel’s combustible part, including the sulfur content S_r, under the influence (i.e. C_r increasing) of metamorphic processes were considered (Fig. 11).

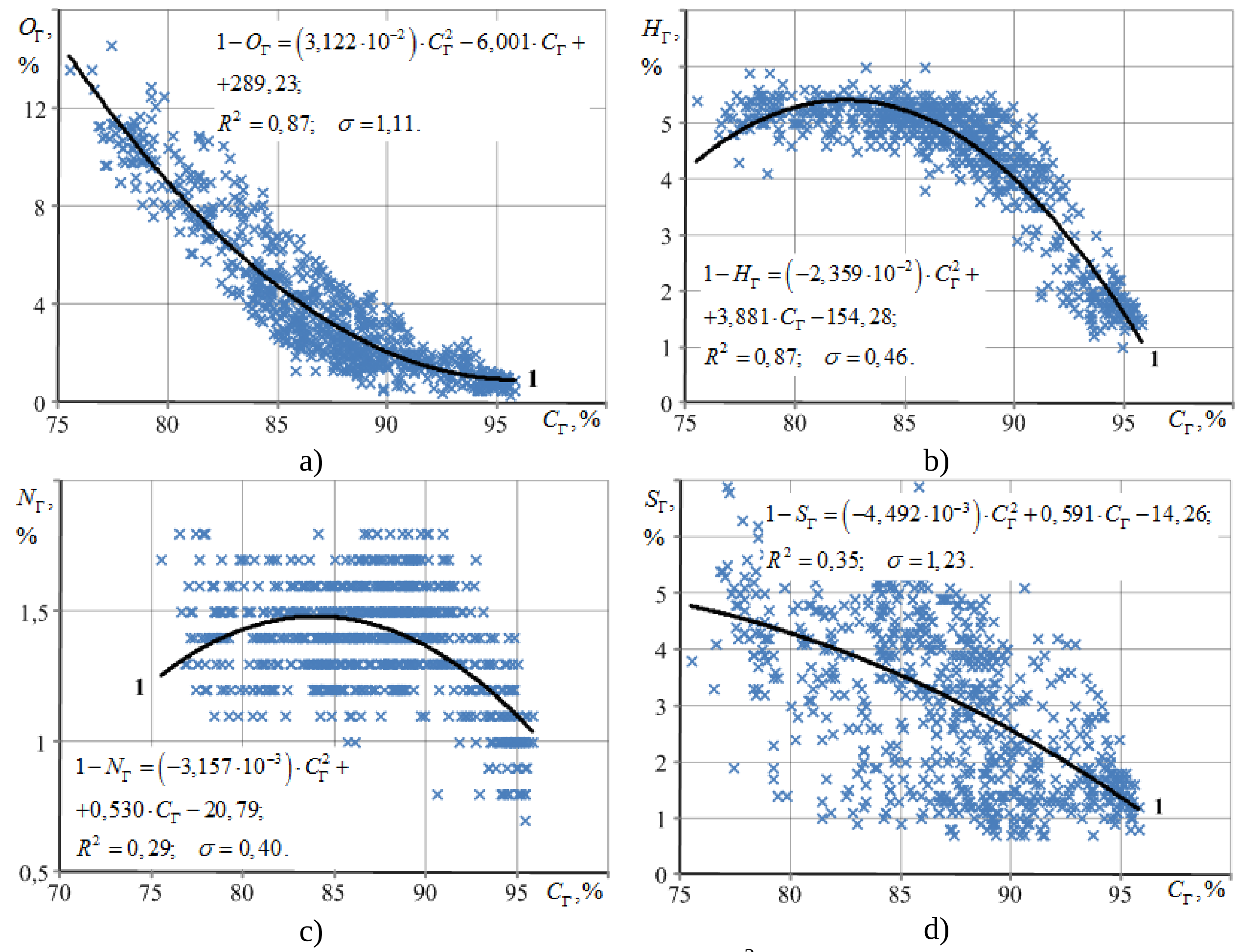
This made it possible to assess the effect of sulfur content in the combustible (organic) part of the fuel on the ratio redistribution o for the other basic components of the coal.

Simultaneous consideration of sulfur in the combustible part of fuels containing carbon, oxygen, hydrogen and nitrogen has significantly affected the tightness of the correlations between these elements. The joint consideration of sulfur in the combustible part had a particularly significant effect on the change of oxygen content (Fig. 11, a). Compared with the change in oxygen ($O_о$) in the organic mass (Fig. 4, c), first of all, a decrease for the coefficient of determination (R^2) from 0.97 to 0.87 and an increase of standard deviations from the average of curves (σ) from 0.65 to 1.11% is observed.

Additional consideration of sulfur in the combustible part of the fuel in combination with other basic elements, taking into consideration by the closely spaced values of R^2 and σ, had no significant effect on the nonlinear nature of changes for hydrogen content and its quantitative indicators. Such circumstances are outlined by the closely spaced values of the coefficients of determination as well as standard deviations for the dependence of the hydrogen content in the organic mass, i.e., R^2=0.84 and σ=0.55 respectively (Fig. 2, c) along with similar values for the hydrogen content in the fuel's combustible part (Fig. 11, b). These features indicate that the average values of hydrogen and sulfur ratios in the process of metamorphic transformations of coal seams remain nearly constant for both the organic mass of fuel and its combustible part. Nearly constant values of hydrogen content within the range of 5-6% in both cases for most mines are observed when $C_о$>85% and $C_г$>85%. Significant decrease of hydrogen content to its minimum values (≈1.0%) is observed while carbon content is about 95%.

The nitrogen content in the organic mass, except a single anomalous case ($N_о$=4,23%), remained nearly constant within 1-2% (Fig. 3, c). In the combustible part of the fuel for many coal seams, when the value of $C_г$ exceeds 92%, the nitrogen content is decreased by less than 1% (Fig. 11, c). The low correlation of nitrogen both in the organic mass of fuel ($N_о$) and in its combustible part ($N_г$) with carbon content is indicated by small values for the coefficients of determination (0.14 and 0.29, respectively). The standard deviations were approximately the same (0.39 and 0.40% respectively), which confirms the presence of nitrogen only in the organic mass of coal [11].

The change for the sulfur content in the combustible part of the fuel is random (Fig. 11, d). This is proved by a small value for coefficient of determination (R^2=0.35) as well as high value of the standard deviation compared to the average of curve (σ=1,23%). The standard deviation of sulfur exceeds this index for other basic components for the combustible part of coal. This indicates the significant influence of sulfur on the distribution of the content fraction between the basic elements in both the organic mass and the combustible part of the fuel. This effect is confirmed by the significant values for the standard deviations of the total sulfur content (S_t^d) from the average of curves both for its dependence on the carbon content in the organic mass (σ=1.58%) as well as carbon content in the combustible part of the fuel (σ=1.10%).



1 – average of curves; × - experimental data [16]; R^2 - coefficients of determination;
σ - standard deviation for the average of curves.

Figure 11 – Dependence of oxygen (a), hydrogen (b), nitrogen (c) and sulfur (d) in the combustible part of the fuel on the carbon content for bituminous coal seams as well as anthracite coal seams.

Consideration of the sulfur content in the combustible part of the fuel ($S_г$) along with other basic elements ($C_г$, $O_г$, $H_г$, $N_г$) significantly affected the ratio redistribution for these elements (Fig. 11).

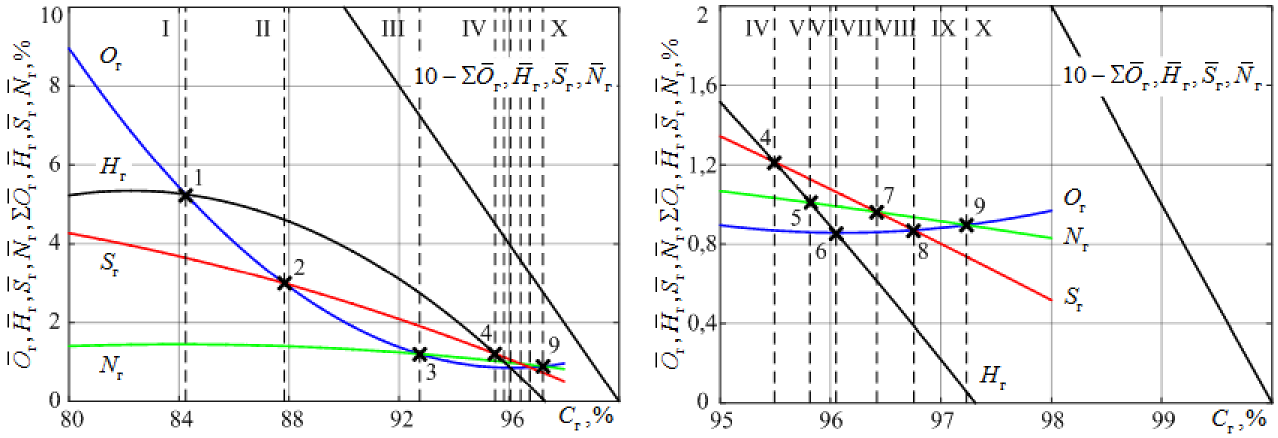
Despite the correlation tightness only for the dependence of oxygen and hydrogen content on carbon (Fig. 11, a and b), it is impossible to state the control of carbon content over the content of these elements.

Thus, at the late stages of metamorphic transformations of coal seams ($C_г$>90%) the standard deviation (σ=1.14%) could significantly exceed the absolute oxygen content, which experimental values are less than 1% for some coal seams [16]. High non-uniformity at all the stages of metamorphism is also observed for the hydrogen content. The content of nitrogen and sulfur at all the stages of coal conversion for the analyzed set of coal seams is obviously random (Fig. 11, c and d).

Despite the ambiguous relationships between the basic elements of the combustible part of the fuel and carbon, the latter reliably controls their total amount. This reliability is guaranteed by the proximity for the sum of all elements of the combustible part of the fuel to 100% (with an error of about 1.0%) as well as the following equation for this case:

$$C_{\text{г}} = 100 - \Sigma O_{\text{г}}, H_{\text{г}}, N_{\text{г}}, S_{\text{г}}, \%. \tag{4}$$

Taking into consideration abovementioned equation and empirical relationships (Fig. 11), possible changes of the ratios between the average values of the basic elements for the combustible part of the fuel in the process of increased metamorphic transformations (increasing $C_{\text{г}}$) of coal seams (Fig. 12) were observed. Each point of intersection of the two curves, that describe the change for the average values of the content of these elements in the fuel's combustible part, indicates a change for the element compound ratio of the basic elements.



a) b)

Figure 12 – Dependence of the average content for the basic elements in the combustible part of the fuel $\left(\bar{O}_{\Gamma}, \bar{H}_{\Gamma}, \bar{N}_{\Gamma}, \bar{S}_{\Gamma}\right)$ on the carbon content ($C_{\text{г}}$) based on statistical processing of experimental data for coal seams in the Lviv-Volyn basin [16], a – full scale; b – enlarged scale.

1 ($C_{\text{г}}$=84.24), 2 ($C_{\text{г}}$=87.83), 3 ($C_{\text{г}}$=92.75) - points of intersection for the average of curves for oxygen content change $\left(\bar{O}_{\Gamma}\right)$ with hydrogen $\left(\bar{H}_{\Gamma}\right)$, sulfur $\left(\bar{S}_{\Gamma}\right)$and nitrogen $\left(\bar{N}_{\Gamma}\right)$ content respectively, at the early, middle and late stages of metamorphic transformations for coal seams;

6 ($C_{\text{г}}$=96.06), 8 ($C_{\text{г}}$=96.75), 9 ($C_{\text{г}}$=97.23) - points of intersection for the average of curves for oxygen content change $\left(\bar{O}_{\Gamma}\right)$with hydrogen $\left(\bar{H}_{\Gamma}\right)$, sulfur $\left(\bar{S}_{\Gamma}\right)$and nitrogen $\left(\bar{N}_{\Gamma}\right)$ content respectively, at the late stages of metamorphic transformations for anthracite coal seams;

4 ($C_{\text{г}}$=95.49), 5 ($C_{\text{г}}$=95.81) - points of intersection for the average of curves for hydrogen content $\left(\bar{H}_{\Gamma}\right)$ with sulfur $\left(\bar{S}_{\Gamma}\right)$ and nitrogen $\left(\bar{N}_{\Gamma}\right)$ content respectively;

7 ($C_{\text{г}}$=96.66) - points of intersection for average of curves for sulfur content change $\left(\bar{S}_{\Gamma}\right)$ and nitrogen content $\left(\bar{N}_{\Gamma}\right)$;

10 - dependence of the sum of the basic elements $\left(\Sigma\bar{O}_{\Gamma}, \bar{H}_{\Gamma}, \bar{N}_{\Gamma}, \bar{S}_{\Gamma}\right)$on the carbon content;

I, II, III, IV, V, VI, VII, VIII, IX, X - stages of metamorphic transformations of coal seams identified by the ratio change between the average values of the basic elements in the combustible part of the fuel (points of intersection for curves that describe the dependence of major components on carbon content)

Such a changes of the element compound should affect the manifestation of the dangerous properties of coal seams. Coal seams for which the indicators of the content for the main components ($O_г, H_г, N_г, S_г$) are located between two adjacent intersection points of the curves could be considered, at a first approximation, as sufficiently close ones for their manifestation of hazardous properties due to metamorphic transformations of the coal seams.

The point's coordinates obtained by the intersection of the two curves were calculated analytically, equating the right-hand members of respective equations (Fig. 11). As a result, for all the cases, after transformations being performed, quadratic trinomials (second-order polynomials) that equal zero were obtained. Their solution first of all made it possible to determine the coordinates of the points for the horizontal axis ($\bar{C}_г$), further their solution allows to calculate, according to the relevant empirical relationships, the ordinates that describe the content of every element (Table 2). As a control of the calculations accuracy according to equation (4) for every point of intersection of the curves the sum of all basic elements for the combustible part of the fuel was determined. The maximum deviation from 100% for a single case was 1.12%, for the rest of the cases deviation did not exceed 0,5%, which is acceptable for engineering calculations.

The result of data statistical processing for the analyzed set of coal seams [16] identified ten distinguished stages of their metamorphic transformations by the factor of ratio changing between the average values of the basic elements of the fuel's combustible part (Table 2). Every stage of metamorphic transformations corresponds to the range of the average content of the elements. The ranges for every stage are significantly different, which indicates a different tendency of coal seams to the manifestation of certain dangerous properties. For example, oxygen which is the most prone to chemical activity, in a sequential transition from stage I to stage IV, has been moved from the first place within the range to the last one.

Such rearrangements also affected the content of the other elements. After the loss of oxygen priority within the range at the first stage, the first place in the following stages consistently were taken by the indicators such as the average hydrogen content (three stages), sulfur (three stages) and nitrogen (two stages). At the final X stage, the priority has returned to the average oxygen content. Such a changes in the relationships between the basic elements indicate the important role of every single element at the stage of metamorphic transformations. For this reason, during prediction of the specific hazardous properties for the coal seam, it is necessary to consider not only changes for carbon content in metamorphic transformations according to equation (4), but also every basic element, regardless of its chemical activity. The average content of the other elements could be calculated with sufficient accuracy to predict the hazardous properties of coal seams (about 1.0%) by means of the following equations:

Table 2 – The results of the stages of metamorphic transformations establishing for coal seams by the ratio change for the basic elements of the combustible part of the fuel on the basis of statistical processing of experimental data [16]

Stages of transformation	Number of intersection points for average of curves as well as elements with the same average content values		Ranges of change for carbon content as well as the total amount of other components, %		The average content of the basic elements in the combustible part of the fuel and their possible ranges of value changing for the points of intersection of the average of curves, %										Range of elements according to their average values within the ranges between the points of intersection of the average of curves	The total amount of the basic elements of the fuel's combustible part, $\Sigma\bar{C}_г, \bar{O}_г, \bar{H}_г, \bar{S}_г, \bar{N}_г$, %
	№№ point	component crossed				oxygen		hydrogen		sulfur		nitrogen			
			$C_г$	$\Sigma O_г.H_г.S_г.N_г$	$\bar{C}_г$	$\bar{O}_г$	$\bar{O}_г \pm 1,21*$	$\bar{H}_г$	$\bar{H}_г \pm 0,46*$	$\bar{S}_г$	$\bar{S}_г \pm 1,23*$	$\bar{N}_г$	$\bar{N}_г \pm 0,40*$		
I	1	$O_г$. $H_г$	<84.24	>15.76	84.24	5.18	3.97÷ 6.39	5.18	4.72÷ 5.64	3.59	2.36÷ 4.82	1.45	1.05÷ 1.85	$\bar{O}_г, \bar{H}_г, \bar{S}_г, \bar{N}_г$	99.64
II	2	$O_г$. $S_г$	84.24÷ 87.83	15.76÷ 12.17	87.83	2.89	1.68÷ 4.10	4.61	4.15÷ 5.07	2.89	1.66÷ 4.12	1.40	1.00÷ 1.80	$\bar{H}_г, \bar{O}_г, \bar{S}_г, \bar{N}_г$	99.62
III	3	$O_г$. $N_г$	87.83÷ 92.75	12.17÷ 7.25	92.75	1.12	0.00÷ 2.33	2.75	2.29÷ 3.21	1.85	0.62÷ 3.08	1.12	0.72÷ 1.52	$\bar{H}_г, \bar{S}_г, \bar{O}_г, \bar{N}_г$	99.59
IV	4	$H_г$. $S_г$	92.75÷ 95.49	7.25÷ 4.51	95.49	0.69	0.00÷ 1.90	1.18	0.72÷ 1.64	1.18	0.00÷ 2.41	1.03	0.63÷ 1.43	$\bar{H}_г, \bar{S}_г, \bar{N}_г, \bar{O}_г$	98.88
V	5	$H_г$. $N_г$	95.49÷ 95.81	4.51÷ 4.19	95.81	0.68	0.00÷ 1.89	1.01	0.55÷ 1.47	1.06	0.00÷ 2.29	1.01	0.61÷ 1.41	$\bar{S}_г, \bar{H}_г, \bar{N}_г, \bar{O}_г$	99.57
VI	6	$O_г$. $H_г$	95.81÷ 96.05	4.19÷ 3.95	96.05	0.77	0.00÷ 1.98	0.77	0.31÷ 1.23	0.99	0.00÷ 2.22	0.99	0.59÷ 1.39	$\bar{S}_г, \bar{N}_г, \bar{H}_г, \bar{O}_г$	99.57
VII	7	$S_г$. $N_г$	96.05÷ 96.66	3.95÷ 3.34	96.66	0.68	0.00÷ 1.89	0.45	0.00÷ 0.91	0.88	0.00÷ 2.11	0.88	0.48÷ 1.28	$\bar{S}_г, \bar{N}_г, \bar{O}_г, \bar{H}_г$	99.55
VIII	8	$O_г$. $S_г$	96.66÷ 96.75	3.34÷ 3.25	96.75	0.74	0.00÷ 1.95	0.39	0.00÷ 0.85	0.74	0.00÷ 1.97	0.93	0.53÷ 1.33	$\bar{N}_г, \bar{S}_г, \bar{O}_г, \bar{H}_г$	99.55
IX	9	$O_г$. $N_г$	96.75÷ 97.23	3.25÷ 2.77	97.23	0.80	0.00÷ 2.01	0.06	0.00÷ 0.52	0.66	0.00÷ 1.89	0.80	0.40÷ 1.20	$\bar{N}_г, \bar{O}_г, \bar{S}_г, \bar{H}_г$	99.55
X	–	–	>97.23	<2.77	–	–	–	–	–	–	–	–	–	$\bar{O}_г, \bar{N}_г, \bar{S}_г, \bar{H}_г$	–

* - standard deviations from the average of curves according to the values of oxygen, hydrogen, sulfur and nitrogen for the analyzed set of coal seams

$$\bar{O}_{\text{г}} = 100 - \Sigma \bar{C}_{\text{г}}, \bar{H}_{\text{г}}, \bar{N}_{\text{г}}, \bar{S}_{\text{г}}, \tag{5}$$

$$\bar{H}_{\text{г}} = 100 - \Sigma \bar{C}_{\text{г}}, \bar{O}_{\text{г}}, \bar{N}_{\text{г}}, \bar{S}_{\text{г}}, \tag{6}$$

$$\bar{N}_{\text{г}} = 100 - \Sigma \bar{C}_{\text{г}}, \bar{H}_{\text{г}}, \bar{O}_{\text{г}}, \bar{S}_{\text{г}}, \tag{7}$$

$$\bar{S}_{\text{г}} = 100 - \Sigma \bar{C}_{\text{г}}, \bar{H}_{\text{г}}, \bar{O}_{\text{г}}, \bar{N}_{\text{г}}. \tag{8}$$

But at the same time, determination of the average values for the elements does not guarantee their reliable ranging at every stage of metamorphic transformations of coal seams. Changes of the range within every stage of metamorphic transformations could be caused by individual deviations of the indicators from the average of lines. They are described, to some extent, by standard deviations from the average of curves. The standard deviations (σ) from the average of curves were determined for every basic element (Fig. 11), which allows to establish the approximate ranges of change for every basic element for every point of intersection for the average of curves (Table 2). Individual fluctuations of the content for the basic elements, in separate ranges of their metamorphism, are able to change the hierarchical ranges for the average of curves. For example, oxygen change for the point 1 (Fig. 12, Table 2) is within the range of 3.97-6.39%; hydrogen change range is within 4.72÷5.64%; sulfur change range is within 2.36÷4.82%. Any of these elements for a single coal seam may take the first place within the actual range, which may differ significantly from the series of range based on the average of curves. At this stage of metamorphic transformations, only nitrogen within the range of its possible content (1.05÷1.85%) is unable to take a prior place within the content range for the basic elements. This disposition of possible relationships between the content of the basic elements is changed drastically at the later stages of metamorphism (VIII, IX), when nitrogen, even for the average of curves, is in first place in the ranking. This is due to the relatively high consistency of nitrogen content at all the stages of metamorphic transformations of coal seams as well as the tendency to the significant reduction of the elements' content for the other elements with increasing of the metamorphism process.

The examples considered have shown that the ratios of the basic elements for the combustible (organic) mass of some coal seams are genetically random. For this reason, considering the average indicators of the element's content change in metamorphic processes, it is impossible to reliably predict the manifestation of hazardous properties of a particular coal seam during mining operations.

Considering one of the possible aspects of metamorphic transformations associated with changes of the elements' content for the basic elements in the combustible (organic) mass, it is necessary to individually take into account the actual relationship between all the components of every single coal seam. This makes it possible to assess the chemical activity of coal seams and the associated manifestation of some hazardous properties during mining operations.

The second possible aspect of metamorphism, which affects the manifestation of hazardous properties of coal seams, includes changes of the structure as well as physical and mechanical properties of coal. Their description requires a completely different additional set of indicators associated with a particular type of manifestation of a hazardous property.

Conclusions. Study has shown a large multiple factor influence of metamorphic processes both on the change of the element compound of the combustible (organic) coal mass as well as its physical and mechanical properties. This range of changes of the compound and properties of coal during the metamorphism of coal seams could not be described, as presented in regulatory documents [1-7], by means of a single indicator i.e., the volatile release during the thermal destruction of fuel. This indicates the necessity to improve the regulatory documents for the safe conduct of mining operations. While making the necessary amendments, it is proposed to take into account the established features of changes within the element compound and properties of coal during metamorphic transformations of coal seams. These features are as follows:

- a single indicator of metamorphism, even the most versatile one is unable reliably and comprehensively describe the ratio change for all the elements of combustible (organic) mass as well as its properties;
- the carbon content more reliably, compared to the volatile release per volume unit as well as mass unit, determines the oxygen content at all the stages of metamorphic transformations of both bituminous coal and anthracite;
- the use of carbon content in the organic mass as an indicator of metamorphic transformations of coal seams instead of volatiles eliminates the inconsistency of Hilt's law over the influence of stratigraphic depth of coal seams;
- coal chemical map compiling has allowed to determine a discrepancy between the oxygen content and the stage of metamorphic transformations for coal seams, as predicted by the authors, as well as the value of the volatile release. The volatile release mainly refers to indicators of fuel quality, rather than to the indicators for metamorphic transformations of coal seams assessing;
- at the stage of isometamorphic anthracites the element compound of the basic elements of organic matter barely determine their physical and mechanical properties;
- the advantage of the volatile release per volume unit over the carbon content is the higher accuracy of the description for the physical and electromechanical properties of anthracites. This indicates that during forecasting the hazardous properties of coal seams in addition to the content as well as ratio of the basic elements of organic matter, it is necessary to additionally consider indicators that describe the physical and mechanical properties of bituminous coal and anthracite;
- every point of intersection of the two average of curves, that describe the change of the average values of the content for the basic elements in the combustible part of the fuel, indicates a ratio change of the element compound between the elements considered. Indicators of the ratio for the content between the two intersection points for the average of curves indicate the possible proximity of coal seams to the manifestation of their hazardous properties;

- according to the results of statistical processing of experimental data for the analyzed set of coal seams ten particular stages of their metamorphic transformations has been identified at a first approximation by the factor of ratio change between the average values of the basic elements for the combustible part of the fuel;
- changes in the relationship between the content of the basic elements play an important role at every stage of metamorphic transformations. Predicting the specific hazardous properties of the coal seam makes it necessary to consider not only the change of carbon content in the process of metamorphic transformations, but also every basic element, regardless of its chemical activity;
- knowledge of the average values of the basic elements does not guarantee their reliable range at every stage of metamorphic transformations of coal seams. Rearrangement of the range within one stage could be caused by individual deviations from the average of lines;
- the relationship between the content of the basic elements is genetically random for the array of coal seams but it has completely individual nature for the coal seam, which is considered apart from the other coal seams;
- considering one of the possible aspects of metamorphic transformations associated with changes of the element compound for the basic components, it is necessary to thoroughly take into account the actual relationship between all the basic elements for every single coal seam;
- the second possible aspect of the metamorphism manifestation, which affects the manifestation of the hazardous properties of coal seams, is change of the structure as well as physical and mechanical properties of coal. For their description a completely different additional set of indicators associated with a particular type of manifestation of a hazardous property required.

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About authors

Rudniev Yevhen, Candidate of Technical Sciences (Ph.D.), Associate Professor, Head in Department of Electrical Engineering, Volodymyr Dahl East Ukrainian National University (VDEUNU), Severodonetsk, Ukraine, rudnev_es@snu.edu.ua

Antoshchenko Mykola, Doctor of Technical Sciences (D.Sc), Professor, Professor in Department of pharmacy, production and technology, Volodymyr Dahl East Ukrainian National University (VDEUNU), Severodonetsk, Ukraine, antoschenko@snu.edu.ua

Filatieva Elvira, Senior lecturer in Department of chemical engineering and ecology, Volodymyr Dahl East Ukrainian National University (VDEUNU), Severodonetsk, Ukraine, elafilatyeva@gmail.com

Filatiev Mykhailo, Doctor of Technical Sciences (D.Sc), Associate Professor, Professor in Department of construction, urban planning and spatial planning, Volodymyr Dahl East Ukrainian National University (VDEUNU), Severodonetsk, Ukraine, filatev@snu.edu.ua

Tarasov Vadym, Doctor of Technical Sciences (D.Sc), Associate Professor, Head in Department of pharmacy, production and technology, Volodymyr Dahl East Ukrainian National University (VDEUNU), Severodonetsk, Ukraine, tarasov@snu.edu.ua

МЕТАМОРФІЗМ ВУГІЛЛЯ, СТАДІЇ ПЕРЕТВОРЕННЯ ШАХТОПЛАСТІВ ТА ЇХ НЕБЕЗПЕЧНІ ВЛАСТИВОСТІ

Руднєв Є.С., Антощенко М.І., Філатьєва Е.М., Філатьєв М.В., Тарасов В.Ю.

Анотація. Вихід летких речовин, враховуючи методи визначення, не відповідає класичній (загальноприйнятій) характеристиці метаморфізму як зміна складу та властивостей вугілля у процесі геологічних перетворень шахтопластів. Один показник, навіть самий універсальний, не може достовірно та всебічно характеризувати зміну співвідношення всіх компонентів органічної маси та її властивостей під впливом метаморфічних процесів.

Методика проведення досліджень базується на статистичній обробці відомих експериментальних даних, одержаних практично для всіх шахтопластів Донбасу та Львівсько-Волинського басейну.

В якості альтернативного критерію метаморфізму по відношенню до виходу летких речовин прийнятий показник вмісту вуглецю в органічній (горючій) масі палива. Такий вибір альтернативного показника ґрунтується на тому, що при прогнозуванні прояву небезпечних властивостей шахтопластів показник вмісту вуглецю має певну перевагу перед іншими.

За тіснотою кореляційних зв'язків показників виходу летких речовин та вмісту вуглецю з основними компонентами органічної (горючої) маси робився висновок про відповідність кожного з аналізованих показників для достовірної характеристики однієї зі сторін метаморфічних перетворень шахтопластів, пов'язаних зі зміною вмісту основних компонентів органічної (горючої) маси та деяких споживчих якостей палива.

Проведені дослідження показали великий багатофакторний вплив метаморфічних процесів, як на зміну елементного складу горючої (органічної) маси вихідної речовини, тай і його фізико-механічні властивості. Такий спектр змін у складі та властивостях викопного вугілля при метаморфізмі шахтопластів неможливо характеризувати, як це представлено в нормативних документах, практично одним показником - виходом летких речовин при термічній деструкції палива. Це вказує на необхідність удосконалення нормативної бази щодо безпечного ведення гірничих робіт. При внесенні необхідних коригувань пропонується враховувати встановлені особливості зміни в елементному складі та властивості вугілля при метаморфічних перетвореннях шахтопластів.

Ключові слова: вугілля, метаморфізм, шахтопласти, показники, вміст вуглецю, сірка, горюча частина, гірничі роботи, безпека, нормативна база, удосконалення.

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