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THE IMPACT OF THE DEGREE OF COALIFICATION ON THE SORPTION CAPACITY OF COALS FROM THE ZOFIÓWKA MONOCLINE

WPŁYW STOPNIA UWĘGLENIA NA POJEMNOŚĆ SORPCYJNĄ WĘGLI Z REJONU MONOKLINY ZOFÓWKI

The evaluation of threats connected with the presence of methane in coal seams is based on our knowledge of the total content of this gas in coal. The most important parameter determining the potential of coal seams to accumulate methane is the sorption capacity of coal a. It is heavily influenced by the degree of coalification of the coal substance, determined by the vitrinite reflectance R_0 or the content of volatile matter V^{daf}. The relationship between the degree of coalification and the sorption capacity in the area of the Upper Silesian Coal Basin (USCB) has not been thoroughly investigated, which is due to the zonation of methane accumulation in this area and the considerable changeability of methane content in various localities of the Basin. Understanding this relationship call for in-depth investigation, especially since it depends on the analyzed reflectance range. The present work attempts to explain the reasons for which the sorption capacity changes along with the degree of coalification in the area of Jastrzębie (the Zofiówka Monocline). The relationship between parameters R_0 and V^{daf} was investigated. The authors also analyzed changes of the maceral composition, real density and the micropore volume. Furthermore, coalification-dependent changes in the sorption capacity of the investigated coal seams were identified. The conducted analyses have indicated a significant role of petrographic factors in relation to the accumulation properties of the seams located in the investigated area of USCB.

Keywords: coal, methane, vitrinite reflectance, sorption capacity, Upper Silesian Coal Basin

Ocena zagrożeń związanych z obecnością metanu w pokładach węgla opiera się na wiedzy o całkowitej zawartości tego gazu w węglu. Pojemność sorpcyjna węgla a jest najistotniejszym parametrem określającym potencjał akumulacyjny pokładów. Istotny wpływ na pojemność sorpcyjną ma stopień uwęglenia substancji węglowej określany przez współczynnik refleksyjności witrynitu R_0 lub zawartość części lotnych V^{daf}. Relacja pomiędzy stopniem uwęglenia i pojemnością sorpcyjną na obszarze Górnośląskiego Zagłębia Węglowego nie jest gruntownie rozpoznana, ze względu na strefowość nagromadzeń metanu na tym obszarze oraz znaczną zmienność jego zawartości w różnych rejonach Zagłębia. Relacja pomiędzy rozpatrywanymi parametrami zmienia się w zależności od analizowanego zakresu refleksyjności. Celem niniejszej pracy jest próba wyjaśnienia przyczyn zmienności pojemności sorpcyjnej z uwęgleniem

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na przykładzie węgli z rejonu Jastrzębia (Monoklina Zofiówki). Prześledzono relację parametru R_0 w stosunku do V^{daf}. Przeanalizowano zmiany takich parametrów węgla jak skład macerałowy, gęstość rzeczywista oraz objętość mikroporów ze wzrostem stopnia uwęglenia pokładów. Określono zmiany zdolności sorpcyjnej badanych pokładów z uwęgleniem. W rezultacie przeprowadzonych analiz wskazano na znaczącą rolę czynników petrograficznych w odniesieniu do właściwości akumulacyjnych pokładów badanego rejonu GZW.

Słowa kluczowe: węgiel, metan, refleksyjność witrynitu, pojemność sorpcyjna, Górnośląskie Zagłębie Węglowe

1. Introduction

In the south-western part of the Upper Silesian Coal Basin, in an area known as the Zofiówka Monocline (Probierz 2012, Probierz et al., 2012a,b), coal seams characterized by particularly high methane content occur. The long-time mining activity in the area disturbs the strata equilibrium, which results in releases of methane from coal to the excavation area. Methane, a flammable and explosive gas, causes a number of serious threats, such as sudden methane outflows, fires, explosions, or methane and rock outbursts. Not only does the content of methane change across the whole USCB area, but it is also characterized by zonation. In the Jastrzębie area, where the Zofiówka Monocline is located, coal seams are characterized both by high methane content and a real threat of methane and rock outbursts.

Methane occurs in coal in two forms. Over 95% of its total content is deposited in pores of molecular dimensions (Gray, 1987). The gas, accumulated in an adsorbed form on the walls of pores, determines the storage capacity of coal seams (Moore, 2012). Additionally, it constitutes the greatest portion of methane that can be obtained from coal in a natural way, i.e. via releasing the gas during the desorption process. (Żyła, (edit.), 2000). The second form of methane occurrence is the free-form methane, an integral part of the system of macropores, cracks and fissures in coal. Both forms of methane occurrence are included in the balance of the total content of methane in a coal seam (methane-bearing capacity).

In order to evaluate the threats connected with the presence of methane in coal seams, one needs to know the total content of this gas contained in coal. Apart from the methane-bearing capacity, a parameter used in the mining industry worldwide, the sorption capacity and sorption isotherms are used in the process of assessing the storage potential of seams. These parameters, determined under laboratory conditions, provide us with significant information concerning the sorption properties of coal, and their analysis complements experts' opinions concerning the identification of the methane and outburst threat in mines (Dutka & Godyń, 2018).

The most significant coal parameter deciding about the total content of methane in a coal seam (methane-bearing capacity) is the sorption capacity of coal a. It determines the amount of gas sorbed by a unit mass of coal in the state of the sorption equilibrium in the coal-gas system, at a given temperature value. The mining practice has employed the coal sorption capacity value under the methane pressure of 1.0 bar and the temperature of 25°C. Comparing the sorption capacity of coal with the methane-bearing capacity of a coal seam allows us to draw conclusions as to the potential increase in the likelihood of the methane and outburst threats occurrence. A particular attention should be paid to these situations in which the high methane-bearing capacity of a coal seam is accompanied by a relatively low value of the sorption capacity. Such situations may point to an increase in the seam pressure of methane and heightened content of desorbable methane which can be released from coal into the excavation.

Undoubtedly, the degree of coalification has a crucial impact on the sorption capacity of coal. The increase of the degree of coalification is not only due to age of seam, but also the depth at which coal is deposited. This is particularly important in these areas where coal seams are arranged in a way not corresponding to their order or are tectonically distorted. A singular factor influencing a local increase in the degree of coalification may be the existence of zones in which a rise in the seam temperature occurred on a local scale, i.e. in the areas under the impact of the contact metamorphism. Such phenomena may occur under the influence of magma intrusions. The relationship between the degree of coalification and sorption capacity across the USCB area has become an object of some previous research (e.g. Kotas, 1994; Kędzior, 2009) however, the zonation-like character of methane accumulation in USCB induces further and more comprehensive analyses, also on a local scale.

2. The description of the research area

The research area is located in the south-western part of the Upper Silesian Coal Basin (cf. Fig. 1). The Basin, located mostly in Poland and partly in the Czech Republic, is one of the most important coal basins on the European continent. It emerged as a mountain sink in a shape of a triangular basin, filled with coal-bearing Upper Carboniferous creations (Osika, 1987; Gabzdyl, 1994; Probierz et al., 2012a,b). The formation was created during the orogenic movements of the Variscan Orogeny, mainly in its Asturian Stage (Gabzdyl, 1994). A no less significant impact on the Basin's structure was exerted by the Alpine Orogeny. A large number of faults created

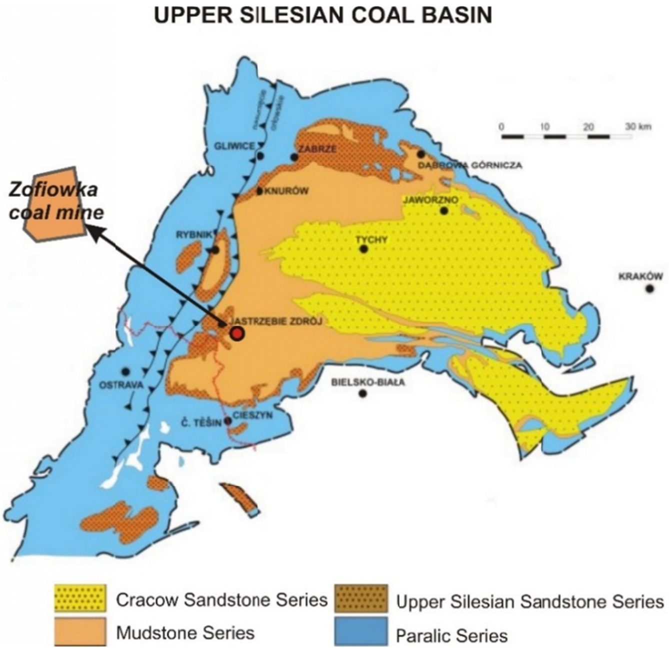


Fig. 1. Location of the research area in the south-western part of the Upper Silesian Coal Basin (www.pgi.gov.pl, 2014)

during the Variscan Orogeny was „rejuvenated" at that time; their amplitudes also changed. Thus, USCB is an orogeny-type basin, created in a mountain sink. Its coal-bearing creations are composed of four lithostratigraphic series (cf. Fig. 2), the oldest of which is known as a paralic series. On the paralic series, the Upper Silesian sandstone series is deposited, comprising Saddle layers and Ruda layers. Another series is known as a mudstone series. It is built of Załęże and Orzesze layers. The youngest Cracow sandstone series was shaped as Łaziska and Libiąż layers (Gabzdyl, 1994).

The area of USCB was divided into three zones differing with respect to their tectonic style (Gabzdyl, 1989; 1994). The first one, the fold zone, is located mainly along the western border of USCB. Its main tectonic elements are folds and thrusts. Another zone belongs to the fold-block area. It emerged mostly along the north-eastern borderline of USCB and is composed

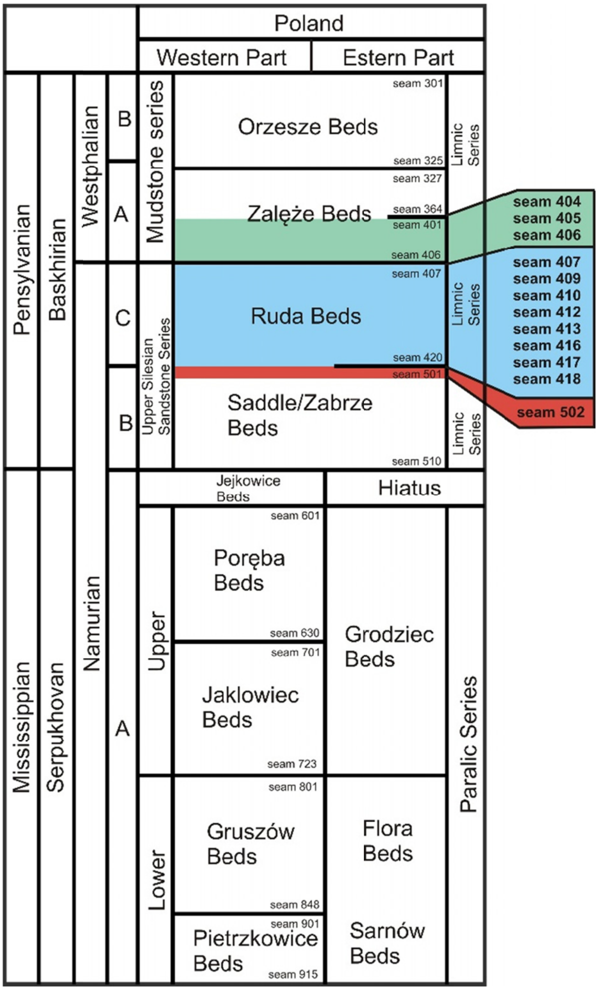


Fig. 2. A stratigraphic table of the carboniferous rocks of USCB, modified after Weniger et al. (2012). The color denotes the formations and coal seams from which research samples were collected

of minor folds and numerous transverse elevations and depressions. The third zone, encompassing the bigger share of USCB, is called the disjunctive tectonic (block) zone. It constitutes the central and eastern part of the Basin. The origins of that zone are connected with the block structure of the USCB substrate, where the main structural elements are faults of three dominant directions.

The research material was collected entirely from coal seams located in the Zofiówka mining facility in the Borynia-Zofiówka-Jastrzębie hard coal mine. Zofiówka mining facility is located in the so-called Zofiówka Monocline (Probierz et al., 2012a), which remains entirely under the impact of disjunctive tectonics, rich of structurally altered coal (Godyń, 2016).

3. Experiments

3.1. Coals

In the planned research, the authors used 24 coal samples collected from coal seams located in the area of the Zofiówka mining facility and representing the Załęże, Ruda and Saddle layers (Gabzdyl, 1994; Probierz et al., 2012a,b) (Fig. 2). The coal was comminuted, sieved and subjected to homogenization in four grain fractions, depending on the samples' intended usage. For the sake of density tests, lumps measuring from 10 to 20 mm were chosen. For the sake of petrographic analyses, coal material of the 0.5÷1.0 mm grain fraction was selected. For technical analyses, coal material of the grain fraction below 0.2 mm (1.0 g) was used, and for sorption analyses, the 0.125÷0.160 mm grain fraction (0.5 g) was prepared.

Information concerning the analyzed coal seams of the Zofiówka mining facility was provided in the first column of Table 1. The samples were marked in such a way as to include the name of the seam (the number identifying a hard coal seam according to the stratigraphic classification of the productive carbon in Polish coal basins) and the letter designation of the part from which a given coal sample was collected.

3.2. Methods

3.2.1. Technical, density and petrographic analyses

The basic properties of the investigated coals were obtained based on the results of a technical analysis, involving determining the content of volatile matter, total humidity and ash in coal. For each of the samples, the real density and the apparent density were determined, by means of the helium pycnometry method and the quasi-liquid pycnometry method, respectively. This was done in order to determine the volume of their micropores, in line with the following formula:

$$V_p = \frac{1}{\rho_p} - \frac{1}{\rho_r} \tag{1}$$

where:

V_p — is the volume of pores, cm^3/g,
ρ_r — is the real density of coal, g/cm^3,
ρ_p — is the apparent density of coal, g/cm^3.

An analysis of the coal's petrographic features was conducted, together with a point quantitative analysis that encompassed both the composition of the main maceral groups (vitrinite Wt^{mmf}, inertinite I^{mmf} and liptinite L^{mmf}) and the mineral substance content M. In order to assess the degree of coalification, the mean vitrinite reflectance R_0 was measured. The analyses were carried out in line with the recommendations of the International Committee for Coal and Organic Petrology (*ICCP*) concerning the petrographic analysis of hard coal and anthracite (PN-ISO 7404-2:2005, PN-ISO 7404-3:2001and PN-ISO 7404-5:2002).

3.2.2. Sorption analyses

The sorption measurements involved determining the sorption capacity of methane under standard laboratory conditions, i.e. under the sorbate pressure of 1.0 bar and at the temperature value of 25°C. For the analyses, dry coal samples were prepared using a mechanical treatment, i.e. crushing and sieving of the 0.125-0.160 mm grain fraction and drying. The measurements were performed by means of the gravimetric method (Benham and Ross, 1989), using the intelligent sorption analyzer IGA-001 made by Hiden Isochema. Prior to the measurement, the samples were outgassed under the temperature of 80°C for 24 hours ($p = 10^{-6}$ mbar). The results were then converted into such as could be obtained under standard conditions (*STP*) and for the pristine coal substance (*daf*).

4. Results

4.1. The description of the coal samples

The results of the technical, densitometric and petrographic analyses of the investigated coals were presented in Table 1. The samples were arranged in such a way as to demonstrate the accordance with the stratigraphy, which corresponds to the increasing geological age. The investigated coal seams belonged to the stratigraphic layers of Westfal (the Załęże layers) and Namur (the Ruda layers and the Saddle layers) (Gabzdyl, 1994) (Fig. 2).

4.2. Petrographic analyses of coal

The results of the quantitative point analysis of the main maceral groups, the content of the mineral substance and the vitrinite reflectance for the studied coals were collected in Table 1. The content of particular maceral groups was turned – by means of calculation – into a state free of the mineral substance (*mmf*). The objects of the analysis were coal seams numbered from 404/2 to 502/1, for the coalification degree ranging (R_0) from 0.98% to 1.25% (cf. Table 1). The range of the measured reflectance value encompassed the metamorphism range characteristic of medium-rank coal, type C and B (UN-ECE, 1998). The changing content of particular macerals can be seen in Fig. 3.

The dominant components of the majority of the analyzed samples were macerals from the vitrinite group (cf. Pictures 1, 2), whose content changed within the range from 40% to over 87%. Reduction in the content of the macerals from the vitrinite group was compensated by the increased content of the macerals from the inertinite group (cf. Pictures 3, 4). The content of inertinite in half of the investigated samples was from 10% to 30%, whereas the remaining coals

TABLE 1

Technical, densitometric and petrographic analyses of the investigated coal samples

Coal sample	V^{daf} [%]	A^a [%]	W^a [%]	ρ_r [g/cm^3]	V_p [cm^3/g]	Wt^{mmf} [%]	I^{mmf} [%]	L^{mmf} [%]	M [%]	R_0 [%]
404/2 F	25,29	5,01	2,09	1,370	0,073	77,17	17,98	4,85	1,82	1,050
404/4 F	24,78	6,00	1,84	1,338	0,034	72,73	21,27	6,00	0,74	0,977
405/1 B	28,45	10,19	1,36	1,390	0,069	76,39	18,42	5,19	9,26	1,022
405/1 F	20,58	11,57	1,19	1,389	0,047	75,56	21,36	3,08	2,22	1,036
405/2 F	22,33	15,68	1,16	1,465	0,061	60,81	36,24	2,96	10,55	1,014
406/1 B	20,92	2,69	1,59	1,326	0,047	60,43	36,34	3,24	0,48	1,061
406/1 F	27,84	2,58	1,95	1,435	0,043	72,49	23,35	4,15	1,13	1,046
407/1 F	23,25	6,31	1,13	1,373	0,043	76,12	18,50	5,38	0,39	1,108
409/3 H	19,89	6,73	1,28	1,356	0,051	62,90	30,55	6,55	1,44	1,090
409/4 D	29,08	20,20	1,12	1,656	0,041	66,38	32,01	1,61	19,91	1,062
410 D	17,81	8,78	1,44	1,344	0,053	57,49	39,37	3,14	1,97	1,085
410 E	20,79	7,54	1,57	1,344	0,026	71,09	23,80	5,11	2,56	1,098
410 G	20,20	5,56	0,88	1,324	0,042	87,55	9,78	2,67	3,43	1,071
412 D	19,34	21,61	0,82	1,356	0,055	70,33	28,44	1,23	17,97	1,123
412 E	19,42	11,77	1,39	1,356	0,050	50,89	48,07	1,04	9,53	1,071
412 G	17,40	4,38	1,06	1,321	0,076	55,33	42,31	2,36	0,55	1,160
413/2 E	18,35	3,49	1,31	1,313	0,060	59,31	37,03	3,65	0,68	1,138
413/2 G	18,71	4,28	1,62	1,326	0,072	77,07	22,11	0,81	0,13	1,185
413/2 H	25,01	10,27	0,92	1,341	0,086	68,28	30,04	1,68	0,91	1,163
416/3 E	19,17	9,57	1,00	1,372	0,048	40,44	51,17	8,4	1,15	1,129
417/1 C	18,33	12,25	1,39	1,442	0,011	70,96	27,18	1,86	9,34	1,138
418/1-2 E	16,54	8,02	1,32	1,407	0,074	67,8	29,3	2,89	4,16	1,061
502/1 C	21,44	8,76	1,08	1,326	0,030	43,39	53,28	3,33	0,79	1,153
502/1 E	13,82	6,48	1,15	1,380	0,009	50,99	41,45	7,56	2,51	1,247

V^{daf} – volatile matter, A^a – ash content, W^a –moisture content, ρ_r – real density, Vp – volume of pores, Wt^{mmf} – vitrinite, I^{mmf} – inertinite, L^{mmf} – liptinite, M – mineral matter, R_0 – vitrinite reflectance

were characterized by an increased amount of this maceral (from 30% to over 53%). Coals from the seams 416/3 E and 502/1 C may be counted among coals with the dominant share of the macerals from the inertinite group. According to Probierz (1989), coals from the Jastrzębie region are characterized by the increased content of macerals from the inertinite group. Additionally, Probierz et al. (2012b) report on some anomalies in the petrographic composition of coals from the Zofiówka region, which result in an increase in the inertinite content at the cost of the vitrinite content. The research discussed in this work confirms the observations of the aforementioned authors. The analyzed coals indeed have higher inertinite content; moreover, the content of this substance increases together with the depth at which the seams are located, as well as their age – and, what follows, with the degree of coalification. This tendency was shown in Fig. 3. The content of macerals from the liptinite group (cf. Pictures 3, 5) in the investigated coals is small. In the case of the seam 413/2 G, it is a mere 0.81%, and in the seam 502/1 E – 7.65%. The average liptinite content in the analyzed coal seams was 3.7%.

An integral part of the coals from the Zofiówka mining area is a mineral substance (cf. Pictures 5, 6). Along with the maceral composition, it may have an impact on the content of the

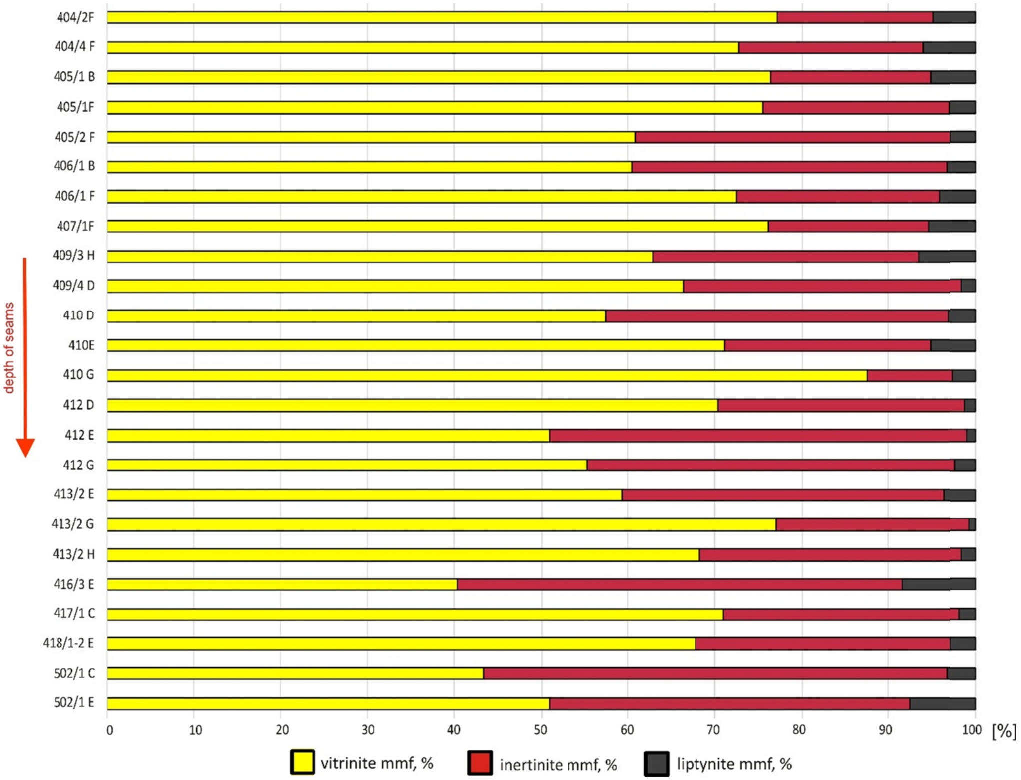
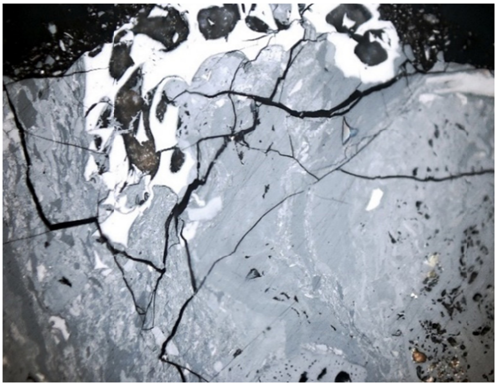


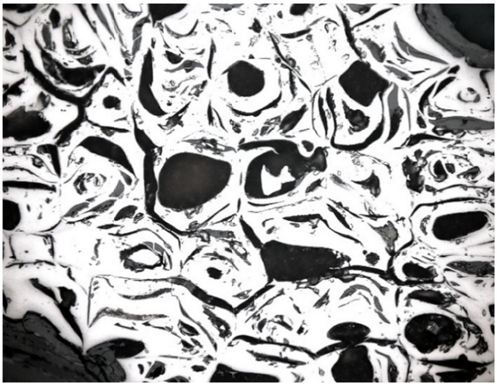
Fig. 3. The results of the point quantitative analysis of maceral groups

Picture 1. A cracked fragment of vitrinite-colotelinite, coal seam 406/1, magnification 500×, reflected light, immersion

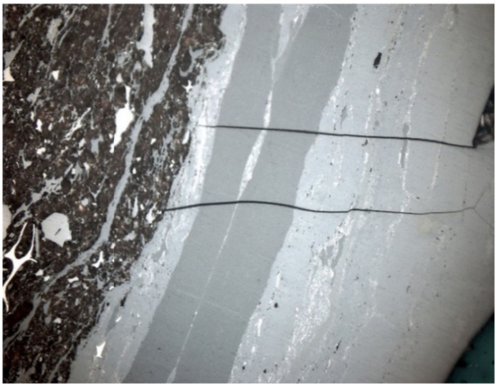
Picture 2. Telinite with a visible cellular structure. The cells are filled with micrinite and rezinite, coal seam 412 G, magnification 500×, reflected light, immersion



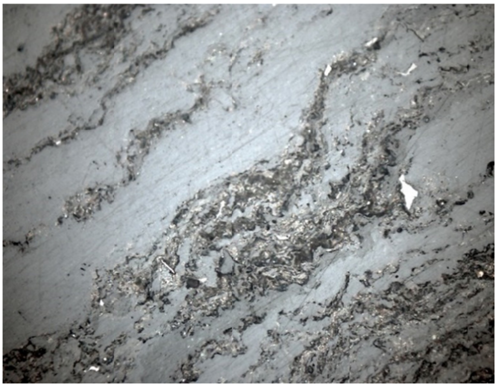
Picture 3. Trimacerite, coal seam 416/3 E,
magnification 500×, reflected light, immersion



Picture 4. Sclerotinite, coal seam 405/1 B,
magnification 500×, reflected light, immersion



Picture 5. Trimacerite with a large macrospore
in its center, the left side of the picture occupied
by a brownish mineral substance,
coal seam 405/1 B, reflected light, immersion



Picture 6. Vitrinite with strands of a brownish-grayish
mineral substance. Coal seam 417/1 C,
magnification 500×, reflected light, immersion

volatile matter in the coal substance. (Weniger et al., 2012). The conducted microscope analyses showed that, for a lot of samples, the presence of the mineral substance was minor and did not exceed 1% (for example, in the seam 413/2 G, the content of the mineral substance is a mere 0.13%); however, in some samples, the content of this substance approximates even 20% (coal seam 412 D; 17.97%).

4.3. The results of the gravimetric sorption tests

Table 2 presents the measured sorption capacity values of coals under the equilibrium methane pressure of 1 bar and the laboratory temperature (25°C). Depending on the coal seam, the sorption capacity of the investigated coals ranged from 2.36 cm^3CH_4/g^{daf} to 3.66 cm^3CH_4/g^{daf}.

TABLE 2

The sorption capacity of the investigated coal samples

Coal sample	a [cm^{3}CH$_4$/g^{daf}]
404/2 F	3.25
404/4 F	3.66
405/1 B	2.94
405/1 F	2.95
405/2 F	3.01
406/1 B	2.99
406/1 F	2.78
407/1 F	2.43
409/3 H	2.90
409/4 D	1.97
410 D	2.97
410 E	3.06
410 G	2.51
412 D	2.64
412 E	2.88
412 G	2.97
413/2 E	2.84
413/2 G	2.40
413/2 H	2.82
416/3 E	2.54
417/1 C	2.71
418/1-2 E	2.36
502/1 C	2.63
502/1 E	2.81

5. Discussion

5.1. The relationship between the coalification degree and the content of volatile matter as illustrated by the example of medium-rank coals from the Zofiówka region

The value of the vitrinite reflectance coefficient makes it possible to evaluate the degree of coalification of organic matter (Zielińska, 2012). The parameter's increase is proportional to the content of carbon. Changeability of this parameter for coal samples obtained at various seams of the Zofiówka mining facility was presented in Fig. 4. According to the tendency showed in the figure, the reflectance coefficient increases systematically in line with the rising numbers of the coal seams from which the investigated samples were collected. The observed relationship, in turn, is in line with Hilt's rule, which says that the coal rank increases with depth (Kędzior, 2015). Coal 418/1-2 E is the only one that fails to fit into the rule.

Another indicator which is commonly used to estimate the advancement of natural coalification process is the parameter V^{daf} determining the content of volatile matter in coal (Krevelen and Schuyer, 1959). A certain drawback of this parameter is its strong dependence on the changing petrographic composition (Probierz, 1989; 2012). According to Probierz, the dominant share of macerals from the inertinite group in coal reduces the value of the parameter V^{daf}; on the other hand, an increase in the content of macerals from the vitrinite group improves the coking properties of coal and boosts the content of volatile matter. Although the highest content of volatile matter is typical of macerals from the liptinite group, the share of these macerals in coals usually does not exceed several percent. Therefore, their impact on the content of volatile matter is insignificant.

Fig. 5 shows the relationship between the reflectance coefficient R_0 of the investigated coals and the content of volatile matter in these coals. As far as the analyzed reflectance values are concerned, this relationship can be explained by means of a linear decreasing trend, which confirms an intuitive conclusion that an increase in the carbonization degree reduces the content of volatile matter in coal.

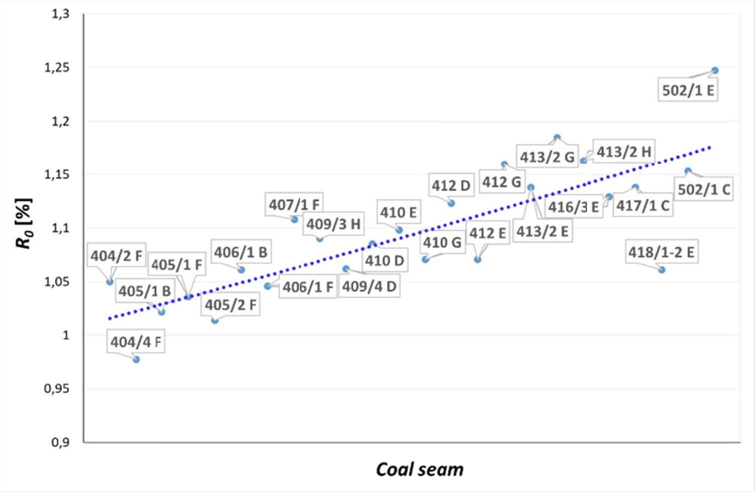


Fig. 4. Vitrinite reflectance for coals obtained at various seams of the Zofiówka mining facility

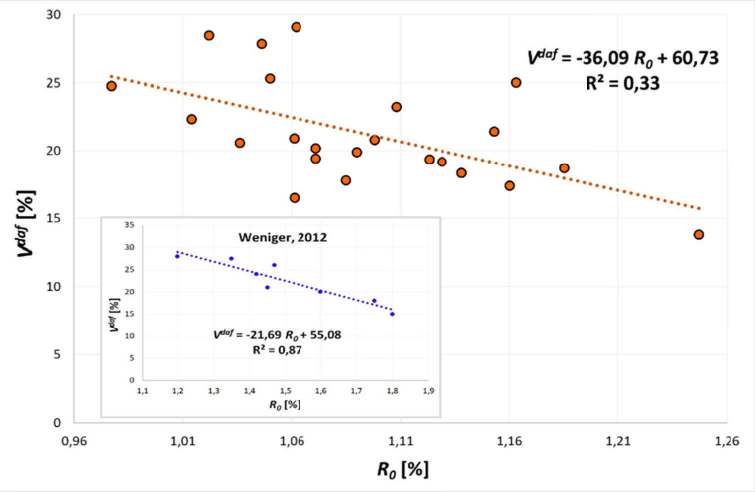


Fig. 5. Correlation between the maturity parameters R_0 and V^{daf} for medium-rank coal samples (as compared with the result obtained by Weniger et al., 2012)

The relationship between the vitrinite reflectance and the volatile matter content for coals of high- to low-volatile bituminous rank from the Czech part of the Upper Silesian Coal Basin was presented in a work by Weniger et al. (2012). It was demonstrated that the best correlation between the content of volatile matter and the value of reflectance coefficient occurs for coals whose coalification degree ranges from 1.0% to 1.8% R_0 (cf. Fig. 5). It was assumed that the relationship obtained for the investigated area allows predicting the values of the R_0 coefficient on the basis of the archive data concerning the content of volatile matter. The studies conducted by Weniger et al. (2012) confirmed the previous results obtained for the USCB coals by Kotarba et al. (2002) and Martinec et al. (2005), among others, and for the coals from Upper and Lower Silesia by Olajossy (2014).

According to Kędzior (2015), individual parameters of coal (R_0 and V^{daf}) are strongly correlated with each other, and the relationship obtained for the ~0.5%-2.4% R_0 range can be approximated with a decreasing linear trend. Fig. 6 proves that the relationship presented by Kędzior could also be approximated by two straight lines of slightly differing angles – steeper for higher reflectance values and less steep for lower reflectance values.

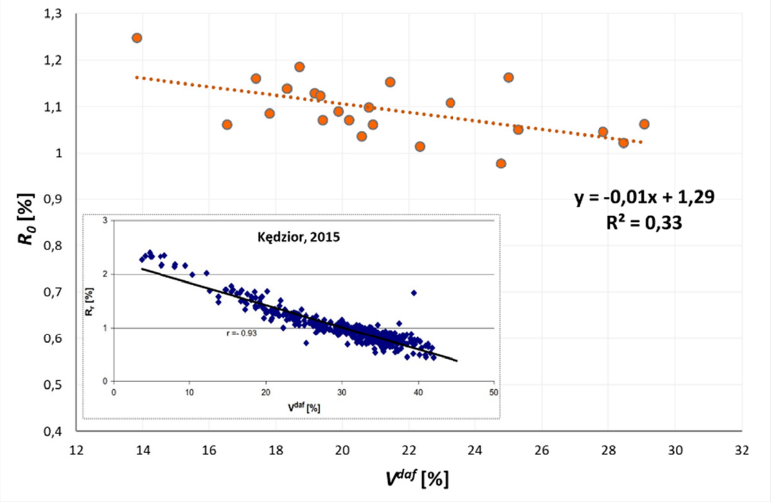


Fig. 6. Correlation between maturity parameters V^{daf} and R_0 for middle-rank coal samples (as compared with the result obtained for coals from the Upper Silesian Coal Basin by Kędzior et al., 2015)

Based on the results obtained in the course of this research and the studies conducted by other authors, we can state beyond doubt that there exists a correlation between the coal's maturity parameters, even though they differ considerably. The present work shall apply the value of the vitrinite reflectance coefficient to the task of finding a correlation between the coalification degree and the coal's ability to accumulate methane, as illustrated by coals threatened with the heightened risk of occurrence of gasogeodynamic phenomena.

5.2. Development of the microporous structure of coal in relation to the degree of coalification

Figures 7 and 8 show the impact of the degree of coalification on the real density and volume of micropores of the investigated coals. The density of the coal skeleton of the studied samples decreases as the degree of coalification rises, with the highest values of ρ_r measured for coals of the reflectance $R_0 < 1.06\%$. The remaining coals' skeletal density ranges from 1.31 g/cm^3 to 1.38 g/cm^3, which is typical of the Upper Silesian coals. At the same time, the volume of micropores in the investigated coals changes visibly across the analyzed reflectance range. Coals of $R_0 < 1.06\%$ and $R_0 > 1.16\%$ have the most porous structure, and the chart illustrating the changeability of the micropore volume in relation to the degree of coalification resembles a parabola whose minimum is in the R_0 range of 1.06%÷1.16%. The coal samples of the smallest micropore volume have the least developed pore structure ($V_p < 0.04$ cm^3/g).

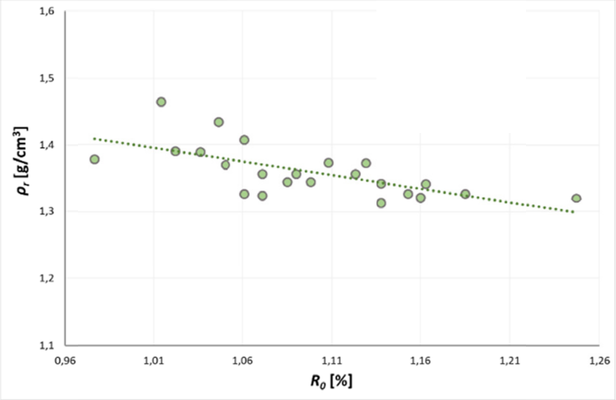


Fig. 7. Changes in the real density of the investigated coals occurring as a result of the rising degree of coalification

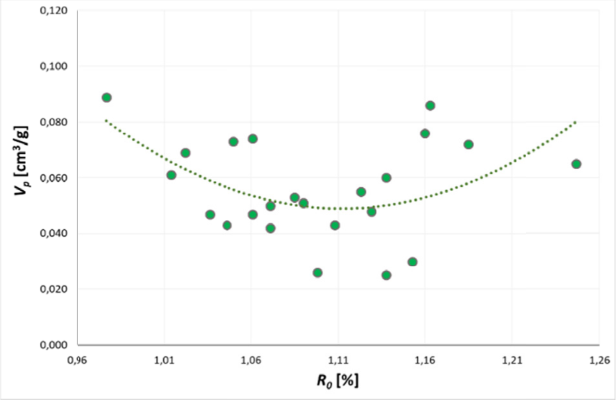


Fig. 8. Impact of the degree of coalification on the micropore volume

According to Kawęcka (1988) and Laxminarayana and Crosdale (1999), the coalification process contributes to greater ordering of the coal structure in coal seams characterized by stronger metamorphism, while also influencing the properties of their pore structure. As the degree of coalification increases, the porosity of coal is gradually reduced from ca. 20% (for the carbon content of 82%, R_0 approximates 0.73%) to the minimum of 3% (for the carbon content of 89%, R_0 approximates 1.3%), and then it starts increasing again as the coalification process progresses (Crosdale et al., 1998). According to Levy et al. (1997), the smallest micropore volume occurs when reflectance R_0 approximates 1.09%, and it corresponds to the carbon content of ca. 87%. This property corresponds with the lowest sorption capacity of coal.

5.3. Changes in the content of the main maceral groups related to the coalification degree

According to Kędzior (2015), the spatial changeability of the maceral composition of the USCB coals results from the lithostratigraphic development of individual parts of the Basin. A significant property of coals from that region is the gradual decrease of the content of vitrinite and increase of the content of inertinite, progressing with the depth at which coal seams are located (Probierz et al., 2012a). This is a specific feature of coals of this region and occurs locally in the area of the Zofiówka Monocline. Fig. 9 demonstrates the impact of the coalification degree on changes in the content of macerals from the vitrinite group, and Fig. 10 shows an increase in the content of inertinite along with an increase in R_0. The content of liptinite does not reveal dependence on the degree of coalification, due to the minor share of this particular maceral in the composition of coals (cf. Table 1).

The charts in Figures 9 and 10 prove that the content of macerals from the vitrinite group in the analyzed range of R_0 decreases as the degree of coalification rises, and that this is accompanied by a simultaneous increase in the content of macerals from the inertinite group. Such a phenomenon occurring in the Jastrzębie region was reported by Probierz (1989) and Probierz et al. (2012b). The Jastrzębie region is characterized by the presence of inertinites that did not emerge in the Carboniferous peat-bogs but were created as a result of funisation of the inertinite macerals, a fact which, to a certain degree, has an impact on an increase in the percentage share of inertinite macerals in these coals. The research presented in this work confirms this observation.

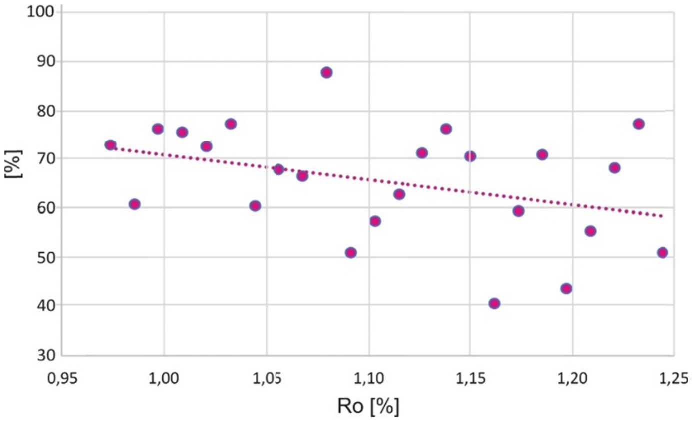


Fig. 9. Changeability of the content of macerals from the vitrinite group in relation to the degree of coalification as a specific feature of Zofiówka Monocline coal seams

5.4. The impact of the degree of coalification, maceral composition and coal's textural properties on accumulation properties

The properties of coal as a methane sorbent are strictly connected with the degree of coalification (Gürdal & Yalçın, 2001), which translates into changes in the sorption capacity of coal seams located at various depths. The present state of knowledge allows us to identify these coal param-

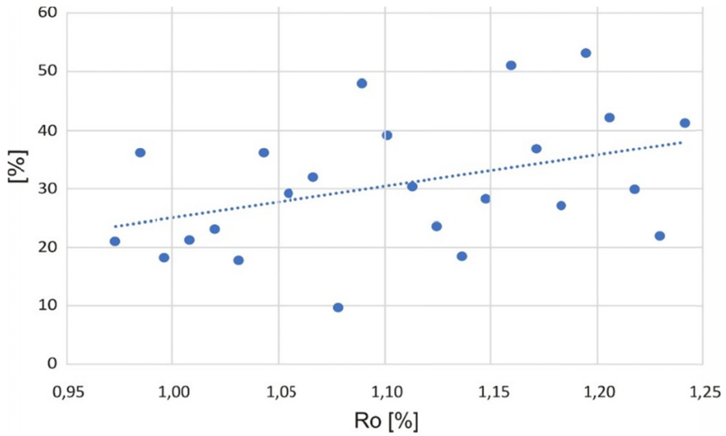


Fig. 10. Changeability of the content of macerals from the inertinite group in relation to the degree of coalification as a specific feature of Zofiówka Monocline coal seams

eters that have a crucial impact on the sorption capacity of coal and are strictly connected with the degree of coalification of the organic substance making up the coal skeleton. Among these parameters, the maceral composition of coal and its textural properties are worth particular attention.

Fig. 11 shows changes in the methane sorption capacity of coal resulting from an increase in the degree of coalification. Across the reflectance range of 0.98%÷1.25%, the relationship between the vitrinite reflectance and the sorption capacity is a decreasing one, which points to the fact that the ability of coal to absorb methane declines as the degree of coalification rises and as the depth at which coal seams are located increases. Additionally, one can estimate that the reduction of the sorption capacity stemming from the rising degree of coalification – from 0.98% to 1.25% – measures ca. 20%.

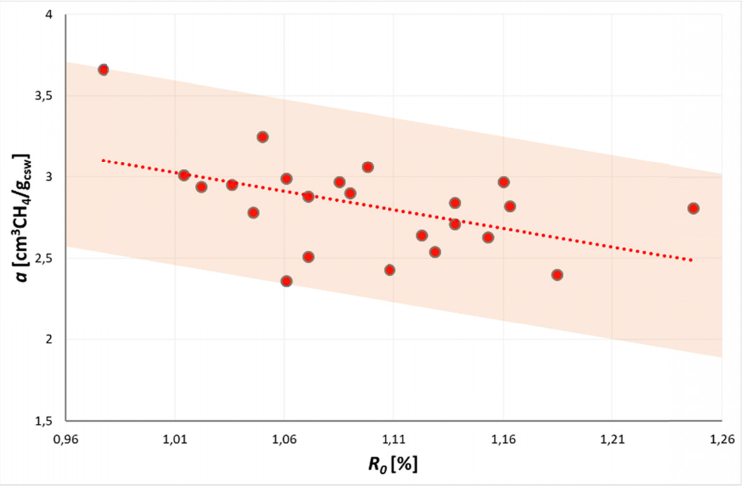


Fig. 11. Methane sorption capacity vs. coal rank

The observations made above are in line with the results included in the work by Laxminarayana and Crosdale (1999), in which the phenomenon of methane accumulation capacity of coal decreasing together with the increasing coal rank is described. Similarly, to the investigated coals, the reduction in the sorption capacity progressing with the increasing coal rank concerns medium-rank coals. That effect was attributed to the blocking of micropores by carbohydrates of a low boiling temperature (Levine, 1993; Levy et al., 1997). It was stated that an increase in the availability of sorption sites occurs only when the coal rank increases as a result of bursting of the carbohydrate components, which leads to an increase in the sorption capacity of coal.

The relationship between coal's methane sorption capacity and the coal rank was analyzed by a lot of researchers, including Jasienko 1995, Ceglarska-Stefanska & Brzóska, 1998; Laxminarayana & Crosdale, 1999; Olajossy, 2014; Nie et al., 2016. However, conclusions provided by these authors differ slightly. The majority of researchers managed to demonstrate that, during a long-term coalification process, coal loses humidity, while the content of micropores in the coal structure increases, which is conducive to a greater methane sorption capacity (Hildenbrand et al., 2006). Studies by Levy et al. (1997), Laxminarayana and Crosdale (1999), Prinz et al. (2004) and Dutta et al. (2011) showed that the relationship between the methane sorption capacity and the coal rank can be illustrated with an U-shaped curve, described by means of a parabola. According to the parabolic trend, high sorption capacities occur in the area of both low and high degree of coalification, with the minimum at the reflectance R_0 value of ca. 1.0% (86% of carbon). Similarly, Levy et al. (1997) determined the minimum sorption capacity, corresponding to the content of carbon of ca. 87% ($R_0 \sim 1.09\%$). The reduced capacity of coal to accumulate methane, which can be referred to the results of the research by Prinz et al. (2004) and Prinz and Littke (2005), corresponded to the lowest micropore volumes.

The 2014 research by Olajossy, who studied the impact of the degree of coalification on the sorption capacity of Polish coals from the Upper- and Lower Silesian Coal Basin across the wide coalification range, confirmed the presence of the parabolic relationship. The aforementioned range encompassed R_0 values from ca. 0.6% to 2.2%, which corresponded to the range of carbon content of 78%÷93%. The results of this research were shown in Fig. 12. Coal rank-dependent changes in the sorption capacity, described by means of a parabolic tendency in the previous research, are characteristic of dry coals (Gensterblum et al., 2013).

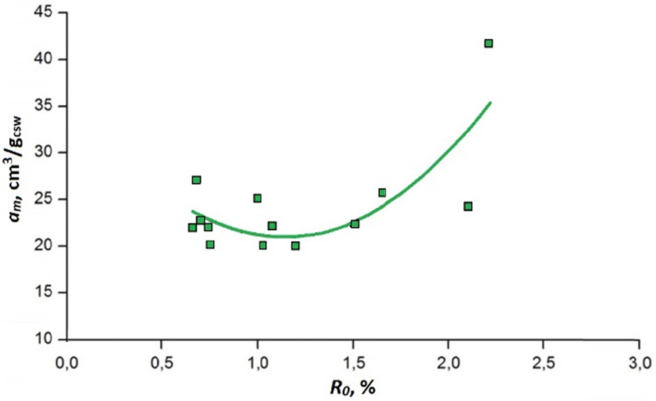


Fig. 12. Impact of the coal rank on the maximum methane sorption capacity of coal, Poland (Olajossy, 2014)

It is worth noticing that the lowest sorption capacity of coals presented in Fig. 11 corresponds with the minimum micropore volume as illustrated by the example of medium-rank coal samples from the Jastrzębie region (cf. Fig. 8). Although the values of micropore volume are the lowest for R_0 of ca.1.11%, the methane sorption capacity is consistently reduced in the studied reflectance range of 0.98%÷1.25% (cf. Fig. 11).

In the work by Weniger et al. (2012), it was demonstrated that, in the reflectance range of 1.0%÷1.8% (medium-rank coals to high-rank coals), the methane sorption capacity increased in a linear manner along with the coalification degree. The scope of the analyzed relationship corresponded with the rising course of the parabolic curve. In the case of the investigated samples, the range of the coalification degree constitutes 25% of the approximated by Weniger et al. (2012) coalification range. Additionally, the analysis of charts provided by these authors shows that an attempt to approximate the obtained experimental points solely in the reflectance range from ~1.0% to 1.25% would make it possible to describe the obtained relationship by means of a decreasing function: a first or second-degree polynomial. In contrast, the reduction in the maximum methane sorption capacity occurring along with the rise in coal rank (low-rank to medium-rank coals) presented by Faiz et al. (2007) is in line with the falling course of the parabolic curve, and it corresponds to the tendency confirmed by the present paper for medium-rank coal samples. In scientific sources, the results of research in which the methane sorption capacity did not correlate with the coal rank can also be found (Li et al., 2010).

The provided data from relevant scientific sources proves that the relationship between the coal rank and coal's sorption capacity depends on the studied area, the range of the analyzed reflectance values, and the coal composition. The coal rank cannot be the only factor deciding about the sorption capacity of coal – due to the fact that the petrographic components of coal will distort the unequivocal nature of the described relations, if not for anything else (Pini et al., 2010; Feng et al., 2014). Undoubtedly, the observed reduction in the sorption capacity, occurring along with the increasing degree of coalification, is an unfavorable phenomenon. An increase in the methane-bearing capacity of coal seams, the presence of the geothermal gradient and the reduced capacity of coal to accumulate methane will be conducive to a heightened risk of occurrence of gasogeodynamic phenomena.

Studies into the influence of the maceral composition on the sorption properties of coal, described in various scientific sources, differ with respect to their results. A lot of authors confirmed that, for the same coal rank, bright coals (i.e. rich in vitrinite) have a bigger sorption capacity than dull coals (with the dominant share of inertinite) (Crosdale et al., 1998; Chalmers and Bustin, 2007; Dutta et al., 2011; Pan et al., 2012; Weishauptová et al., 2015). Although vitrinite is less porous than inertinite, the former is characterized by a greater share of micropores (Walker et al., 2001). Some studies show, however, that also inertinite coals may display an increased sorption capacity (Hemza et al., 2009). Undoubtedly, the impact of the maceral composition on the sorption capacity of coal depends on the coal rank (Laxminarayana & Crosdale, 1999). Chalmers and Bustin (2007) suggested that the maceral composition has a bigger impact on sorption processes in the case of higher-rank coals, due to their considerable microporosity. Laxminarayana and Crosdale (1999), in turn, proved that the maceral composition of the Australian coals from the Bowen Coal Basin has a bigger impact on the sorption capacity of coals displaying a smaller degree of metamorphism. According to Jureczka et al. (2005), in the south-western part of USCB, coals displaying a higher degree of metamorphism prevail, which is why the impact of the maceral composition on the sorption capacity cannot be clearly defined. Similarly, in the works by (Laxminarayana and Crosdale, 1999, 2002; Mastalerz et al., 2004; Faiz et al., 2007, Weniger et al., 2012), no relationship between the sorption capacity and the maceral composition was identified. The share of macerals may have no visible impact on the sorption capacity due to differing sorption capacities of macerals from the inertinite group (semifusinite is characterized by much greater microporosity than fusinite).

Interesting conclusions can be found in the work by Hemza et al. (2009), in which the authors investigated coals from the Czech part of USCB. They proved that the methane sorption capacity decreases along with an increase in the content of macerals from the inertinite group in the reflectance coefficient range of 0÷50%. For a greater share of inertinite, gradual increasing of the methane sorption capacity of coal was observed.

Results presented in Figures 9 and 10 show that the reduction of the content of macerals from the vitrinite group and simultaneous increase in the content of macerals from the inertinite group might lead to lowering of the methane sorption capacity of coal.

6. Conclusions

Based on the conducted research, the following essential conclusions can be drawn:

1. The values of the vitrinite reflectance coefficient increase in line with the rising numbers of coal seams from which the samples were collected. This regularity, in turn, is in line with the Hilt's rule. The coal rank of the investigated coal samples, characterized by the value of R_0, correlates with another maturity parameter – namely, the content of volatile matter. In the case of medium-rank coals, the relationship between the parameters used to assess the maturity of coal corresponds with the trends identified for the USCB coals by other researchers.

2. The analysis of changes in the textural properties of the investigated coals proved that the measured micropore volumes alter considerably, and the least developed porous structure ($V_p < 0{,}04$ cm^3/g) is typical for coals taken from the middle of the studied reflectance range.

3. Petrographic analyses demonstrated that – although the dominant component of the most coal samples is vitrinite – macerals from the inertinite group constitute a very important element. They occur in substantial quantities, and, in two cases, they even dominate in the petrographic composition. Moreover, it was proved that the share of macerals from the inertinite group increases as the coal rank rises, which simultaneously leads to the reduction of the share of macerals from the vitrinite group.

4. The analysis of the values of sorption capacity as used in the mining practice a (25°C, 1.0 bar) points to the lowering of the ability of coal to accumulate methane along with an increase of the coal rank and the depth at which coal seams occur.

5. The lowest sorption capacity of coal does not correspond with the minimum micropore volume occurring for the reflectance of 1.06÷1.16%.

6. Macerals from the vitrinite group, due to their microporous structure, are, to a large extent, responsible for the sorption processes occurring in coal. The inertinite macerals usually have a macroporous structure, which is why they display a lower sorption capacity. In the analyzed coal seams, an increase in the content of macerals from inertinite group, occurring along with a rise in the degree of coalification, might contribute to the reduction of the sorption capacity. The obtained results show close conformity with the observations included in the work by Hemza et al. (2009).

7. Under the conditions of rising methane-bearing capacity of coal seams and the presence of the geothermal gradient, the lowered sorption capacity of coals in the Jastrzębie region undoubtedly contributes to a heightened risk of occurrence of gasogeodynamic phenomena.

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