

HENRYK R. PARZENTNY*¹, LEOKADIA RÓG****GEOCHEMICAL CHARACTERISTICS OF THE BISMUTH AND ANTIMONY OCCURRENCE
IN SOME COAL SEAMS IN THE LUBLIN COAL BASIN (LCB)****GEOCHEMICZNA CHARAKTERYSTYKA WYSTĘPOWANIA BIZMUTU I ANTYMONU
W NIEKTÓRYCH POKŁADACH WĘGLA LUBELSKIEGO ZAGŁĘBIA WĘGLOWEGO (LZW)**

The study included 24 samples of coal with 7 cores, boreholes (7 coal seams), made by the Polish Geological Institute in Warsaw at the site of a Chelm field and 6 coal samples taken from 2 decks in the Lublin Coal mine „Bogdanka“ S.A. in LCB. Based on performed tests found generally low levels of Sb and Bi in coal. In the vertical profile of the LCB contents of Bi and Sb in coal generally increases from coal seams younger to older age. Content of Bi in coal from roof part coal seams is usually higher, and ash content in the coal content of Sb are generally lower than in the carbon of the middle part decks. The content of Bi in the lateral coal deposits is unlikely to vary, and the gap in the coal content of Bi between the sampling regions coal do not exceed 1.7 g / Mg. In contrast gap Sb content in coal on the extent LCB is from 1.7 g / Mg of 5.8 g / Mg. The biggest influence on the content of Bi and Sb in coal from the LCB is probably organic matter in which these elements are scattered and do not form their own minerals.

Keywords: Bismuth and antimony, bituminous coal, coal seams, Lublin Coal Basin

Celem artykułu było określenie zróżnicowania zawartości Bi i Sb w węglu LZW oraz określenie roli substancji organicznej w ich koncentrowaniu w węglu. Pierwiastki te należą do bardzo rzadko (Sb) lub w ogóle nie oznaczanych (Bi) w węglu kamiennym ze złóż Polski. Badaniami objęto 24 próbki węgla kamiennego z rdzeni 7 otworów wiertniczych (7 pokładów węgla), wykonanych przez Państwowy Instytut Geologiczny w Warszawie na obszarze złoża Chelm oraz 6 próbek węgla pobranych z 2 pokładów w kopalni Lubelski Węgiel „Bogdanka” S.A. w LZW (Rys. 1).

Na podstawie wykonanych badań stwierdzono, że skład petrograficzny węgla (Tab. 1) jest zbliżony do składu petrograficznego paleozoicznych węgli Europy w makroregionie North Atlantic ($V_t = 68\%$, $L = 12\%$, $I = 20\%$), określonego przez Lopo de Sousa e Vasconcelous (1999). Zawartość wityrnytu w badanym węglu jest porównywalna z zawartością tej grupy macerałów w węglu z serii paralicznej (63,8%) GZW. Ze względu na refleksyjność wityrnytu, badany węgiel zaliczono do średnio uwęglonego węgla (typ C) ortobitumicznego. Przeciętna zawartość Bi i Sb w badanym węglu LZW jest w przypadku Bi mała, a w przypadku Sb zbliżona do wartości klarków dla węgla bitumicznego i subbitumicznego (Tab. 2).

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Zawartość Bi i Sb w węglu ogólnie zwiększa się idąc od pokładów wiekowo młodszych do starszych (Rys. 2). Zawartość Bi w węglu z przystropowej części pokładów węgla jest zwykle większa, a zawartość popiołu w węglu i zawartość Sb w węglu są na ogół mniejsze, niż w węglu z części środkowej pokładów (Rys. 3). Ze względu na małą zawartość Bi i Sb w węglu, potencjalnie najkorzystniejszym do ewentualnego spalania jest węgiel z części przyspągowej pokładu 378. Zawartość Bi w węglu po rozciągłości pokładów jest mało zróżnicowana, a rozstęp zawartości Bi w węglu pomiędzy rejonami opróbowania pokładów węgla nie przekracza 1,7 g/Mg (Rys. 4). Natomiast rozstęp zawartości Sb w węglu po rozciągłości LZW wynosi od 1,7 g/Mg do 5,8 g/Mg.

Na podstawie korelacyjnej zależności, między zawartością Bi i Sb w popiele węgla i w węglu, a zawartością popiołu (Tab. 3) przypuszcza się, że największy wpływ na zawartość w węglu LZW omawianych pierwiastków ma substancja organiczna. To przypuszczenie potwierdziły wyniki rozwiązania funkcji RS, wyrażającej (opisaną wzorem nr 1) zależność zawartości pierwiastka w popiele węgla od zawartości popiołu. Stwierdzono, że 100% zawartości Bi i 97% zawartości Sb w węglu pochodzi z substancji organicznej węgla. Obserwacje powierzchni próbek węgla, za pomocą mikroskopu optycznego do światła odbitego i za pomocą skaningowego mikroskopu elektronowego (SEM-EDS), nie ujawniły obecności wtrąceń minerałów Bi i Sb w macerałach, ani domieszek tych pierwiastków w minerałach siarczkowych, siarczanowych, węglanowych i ilastych (Rys. 5). Nie stwierdzono także obecności minerałów własnych Bi i Sb. Przypuszcza się, że Bi i Sb mogą być rozproszone w niektórych macerałach i w minerałach, lecz z powodu przeważającej objętości i masy macerałów w węglu nad substancją mineralną, substancja organiczna ma największy lub wyłączny wpływ na zawartość tych pierwiastków w węglu. Związek pierwiastków z substancją organiczną oceniono jako ogólnie niekorzystny dla środowiska w przypadku wykorzystywania tego węgla do spalania. Omawiane pierwiastki Bi i Sb w zakresie temperatur 327°C-1127°C łatwo bowiem ulegają w paleniskach odparowaniu oraz szybko kondensują się i wzbogacają na cząstkach faz pierwiastków litofilnych (K, Ca, Na, Mg, Ti). Dlatego mogą one być łatwo uwalniane do atmosfery w formie lotnych związków, a skondensowane na najdrobniejszych cząstkach popiołu lotnego mogą w atmosferze wchodzić w skład pyłu zawieszonego.

Słowa kluczowe: bizmut i antymon, węgiel kamienny, pokłady węgla, LZW

1. Introduction

A large and steadily increasing share of coal from the LCB in total weight of burned coal in Poland, tends to insightful cognition of its geochemical properties. Geochemical tests of coal from LCB began in 1978 in Geological Institute in Warsaw (PIG). They consisted in defining the content (in coal and coal ash) of 25 minor and trace elements (As, Ba, Be, Br, Cd, Cl, Co, Cr, Cu, Ga, Ge, Li, Mn, Mo, Nb, Ni, P, Pb, Rb, Sr, Ti, V, Y, Zn, Zr). The coal came from 12 boreholes drilled by the PIG in Chelm deposit. The results of these studies have been mostly published (Bojakowska & Pasiczna, 2007; Cebulak, 1983; Cebulak & Rózkowska, 1983; Marczak, 1985; Marczak & Lewińska, 1982; Marczak & Parzenty, 1985, 1989; Parzenty, 2009). According to these studies it was found that the greatest variation in the carbon content of the LCB has Zn, and the smallest Mo. The increased content of V, Co, Be and Mo, and a small content of Cu, Zn, Cr, and Pb in the coal was found, in comparison to Clarke for sub-bituminous and bituminous coal. The content of most elements in coal decreases with the depth of the seams. The enrichment of Ga, Zn, Ti, Ba, Cr, As, Pb, Mn, Ge, Ni and Rb elements in roof and floor section of seams, or only in roof part of coal seam was observed in LCB. The Be, Cu and Zn are bonded with organic matter of coal. With a mineral substance, Li, Mo, Cd, Cr, V, Pb and Zn elements are bonded.

So far the results of geochemical coal from LCB are applicable for prediction of the quality of solid products from coal combustion. The content of elements in coal and their association with organic and mineral matter have in fact a greater impact on their content in fly ash and cinder, than the technical and technological conditions for coal combustion (Xu et al., 2003). The research on

finding LCB coal – with obtained ash after its burning that is characterized by a high content of elements critical in terms of their possible recovery – is also conducted (Cebulak, 1983; Strugała et al., 2014). The geochemical tests might have an important manner for predicting toxicity of the waste from thermal coal conversion processes and for the possible recovery of elements and of other so far poorly known elements in coal.

The aim of presented work was to determine the diversity of bismuth (Bi) and stibnite (Sb) content in the LCB coal and to define the role of organic matter in their concentration in coal. In Polish coal deposits these elements are marked very rarely (Sb) or aren't marked at all (Bi). Bismuth (Bi) and stibnite (Sb) are called heavy metals. They have high mutual geochemical affinity, but with a different toxicity of their compounds for human body. Sb compounds cause chronic poisoning of the body, by contrast the compounds of Bi contrary, are used in medicine, among others, for the treatment of skin blemishes, they are a major component of surgical dressings, and drugs to combat gastroenterology diseases caused by bacteria *Helicobacter pylori* (Szczygieł, 2009). Some of the Bi compounds are widely used in industry. They are used among other as catalysts for olefin metathesis reaction that leads to higher octane gasoline. The Bi compounds are used also in the conversion of methane to higher hydrocarbons that can be used as a fuel for internal combustion engines. (Vogt et al., 2008).

2. The methodology of research

The study involved 30 samples taken from Chełm and Bogdanka deposits in LCB (Fig. 1). The samples from Chełm deposit come from 378, 382, 385, 387, 389, 391 and 394 seams and were identified in cores from 7 boreholes (Cyców IG-5, Cyców IG-6, Dorohuczka IG-4, Dorohuczka IG-6, Dorohuczka IG-8, Syczyn IG-2 and Syczyn IG-3). The Polish Geological Institute in Warsaw (PIG) have drilled the boreholes, collected and submitted the samples for tests. Six samples of seam's coal were obtained from Bogdanka deposit. Three samples came from 382 and 385/2 seams (according to the PN-G-04501:1998 standard). The average carbon samples for the petrographic tests were prepared from the raw coal samples (according to the PN-ISO 7404-2:2005 standard). The analytical average samples of coal (according to the PN-G-04502:2014-11 standard) and average fly ash samples for the chemical tests (according to the PN-77/G04528/00 standard) were also prepared. In coal samples the content of maceral groups – according to the PN-ISO 7404-3:2001 standard: vitrinite (*Vt*), liptinite (*L*) and inertinite (*I*) – was determined. The mineral content was also determined. These analyzes were performed by studying the white reflected light using a Zeiss Axio Imager D1m light microscope. By using the X-ray diffraction (XRD) and microscopic reflectance methods the content of mineral groups in the coal was determined (Table 1). After those tests the content of the ash (obtained after burning coal samples at temp. 525°C) according to the Swanson and Huffman (1976) assumptions was assessed. Using the method of inductively coupled plasma emission spectrometry (ICP – ES), the contents of Bi and Sb in the coal ash was determined. The content of elements in the coal ash was then calculated to their content in analytic state coal, and the results are given in Table 2 and in Fig. 2-4. Using the Scanning Electron Microscopy-Energy Dispersive Spectroscopy (SEM-EDS), the places of Bi and Sb occurrences were identified in some coal grains (Fig. 5).

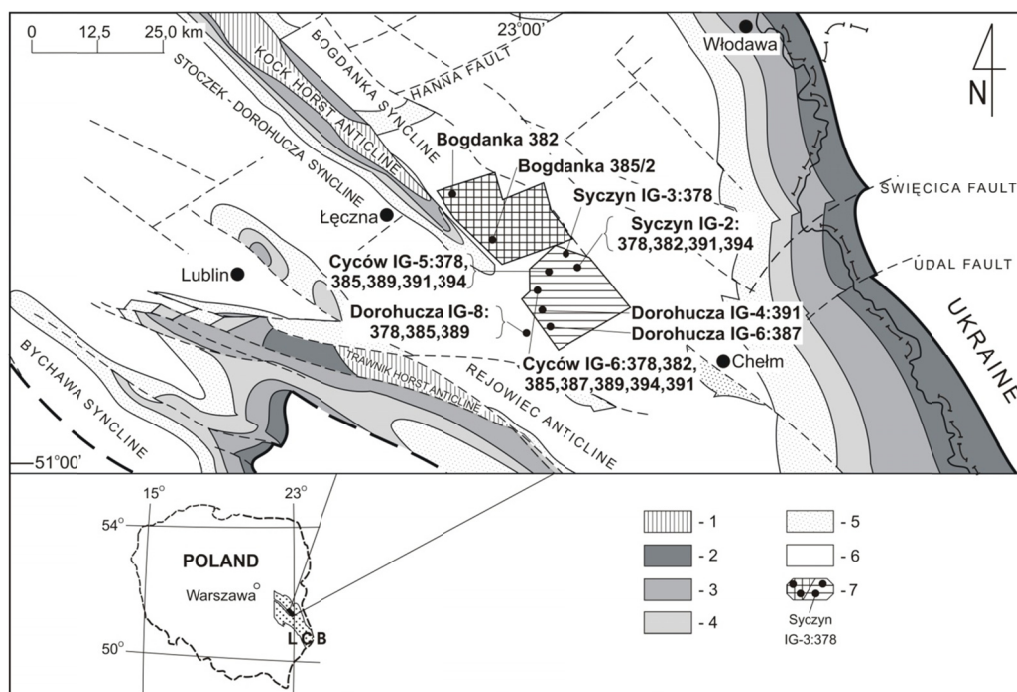


Fig. 1. The location of coal sampling points against geological LCB map (after Porzycki & Zdanowski, 1995)
 1 – Devonian, 2 – Huczwa Formation, 3 – Terebin Formation, 4 – Dęblin Formation, Bug Beds, 5 – Dęblin Formation, Kumów Beds, 6 – Lublin Formation and Magnuszew Formation, 7 – location of the places from where coal samples were taken, boreholes (Syczyn IG-1), marker coal seams (378)

Using the χ^2 Pearson and Kolmogorov-Smirnov ($p = 0.05$) tests it was found that the content of petrographic and geochemical components in coal are normally distributed and the value of the geometric mean estimates best the average value (Table 2). To find a relationship between the ash content and the content of Bi and Sb in coal and coal ash, and between the content of Bi and Sb in coal, and the content of 32 other designated elements in the tested coal (Ag, Al, As, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, K, La, Mg, Mn, Mo, Na, Nb, Ni, P, Pb, Sc, Sn, Sr, Th, Ti, U, V, W, Y, Zn, Zr), the value of Pearson correlation coefficient was calculated. After this step the linear regression coefficients of determination R^2 for the wanted relations were calculated. Verification of the linear regression model was carried out using F – Snedecor test with confidence level of $\alpha = 0.05$ and using the t Student test. The significance of the correlation coefficients for the level of significance < 0.05 was verified. The statistically significant Pearson correlation coefficients – with value of $> \pm 0.35$ – are given in Table. 3.

In interpretation of the data it was assumed that the correlation coefficients with values from $+0.35$ to $+1.00$ may indicate the relationship of Bi and Sb with the mineral substance, and the values of -0.35 to -1.00 may indicate the relationship between Bi and Sb with the organic substance of coal. In order to verify this assumption and to determine which part of the trace element concentration is associated with the organic and inorganic part of coal the RS function of distribution concentration was used. This function – developed by Marczak (1985) – expresses

the dependence of element content in the ash, from the ash content in coal. Its suitability for such a determination has previously been empirically confirmed (Lewińska-Preis et al., 2009; Parzenty, 1994). Equation (1) describes this dependence and has the following form:

$$C_A = C_o \frac{1-A}{1} + kA + C_m \quad (1)$$

where:

- C_A — content of element in coal ash [g/Mg],
- A — content of ash in coal, expressed as mass fraction, in the value of 0 to 1,
- C_o — element content [g/Mg] per unit of weight of organic matter,
- k — coefficient expressing the increase in the content of the element in coal ash [g/Mg], associated with the increase in the ash content by one unit,
- C_m — content of element in coal ash [g/Mg] with $A = 0$ – limit content,

The first two values (A and C_A) were determined experimentally, and other were obtained by solving equation (1). The calculation results are given in Table 3.

3. Tests results and interpretation

3.1. Petrographic content of coal

Semi-bright coal with epigenetic veins of pyrite and marcasite and syngenetic layers of kaolinite clay occurs most frequently in examined coal seams. The petrographic content of coal (Tab. 1) is close the petrographic composition of Paleozoic coals in the North Atlantic

TABLE 1

Petrographic composition (% vol.) and vitrinite reflectance (%) of coal from LCB

Component	Study area
Vitrinite reflectance	0.64
Coal ash content	13.91
Vitrinite (<i>Vt</i>)	57.0/63.3*
Liptinite (<i>L</i>)	23.2/25.7*
Inertinite (<i>I</i>)	9.9/11.0*
Mineral matter, summary	9.9
Clay minerals**	4.6
Quartz	2.6
Microcline	sg
Siderite	0.2
Dolomite+ankerite+calcite	0.8
Jarosite	sg
Pyrite+marcasite	1.7
Chalcopyrite+galene+sphalerite	sg

* Petrographic composition of coal without mineral matter, ** Mineral composition according to diminishing participation in coal: kaolinite >> illite, montmorillonite > halosite, sg – single grain

Europe macroregion ($Vt = 68\%$, $L = 12\%$, $I = 20\%$), determined by Lopo de Sousa e Vasconcelous (1999). The content of vitrinite in tested coal is comparable with the content of this macerals group in coal from paralic series (63.8%) of USCB, determined by Jurczak-Drabek (1996). In accordance to International Classification of Coal Seams (International Classification of Seam Coals, Final Version, 1995), the tested coal was classified as semi carbonated ortobitic coal (type C).

3.2. Volatility of Bi and Sb content in coal

The average contents of Bi and Sb in tested LCB coal for Bi is low and for Sb is close to the Clark value of coal (Table 2). The content of Bi and Sb in coal generally increases from the younger to the older seams (Fig. 2). Also significant differences in the content of Bi and Sb in coal (Fig. 3) in the profile of some singular coal seams were observed. The content of Bi in coal

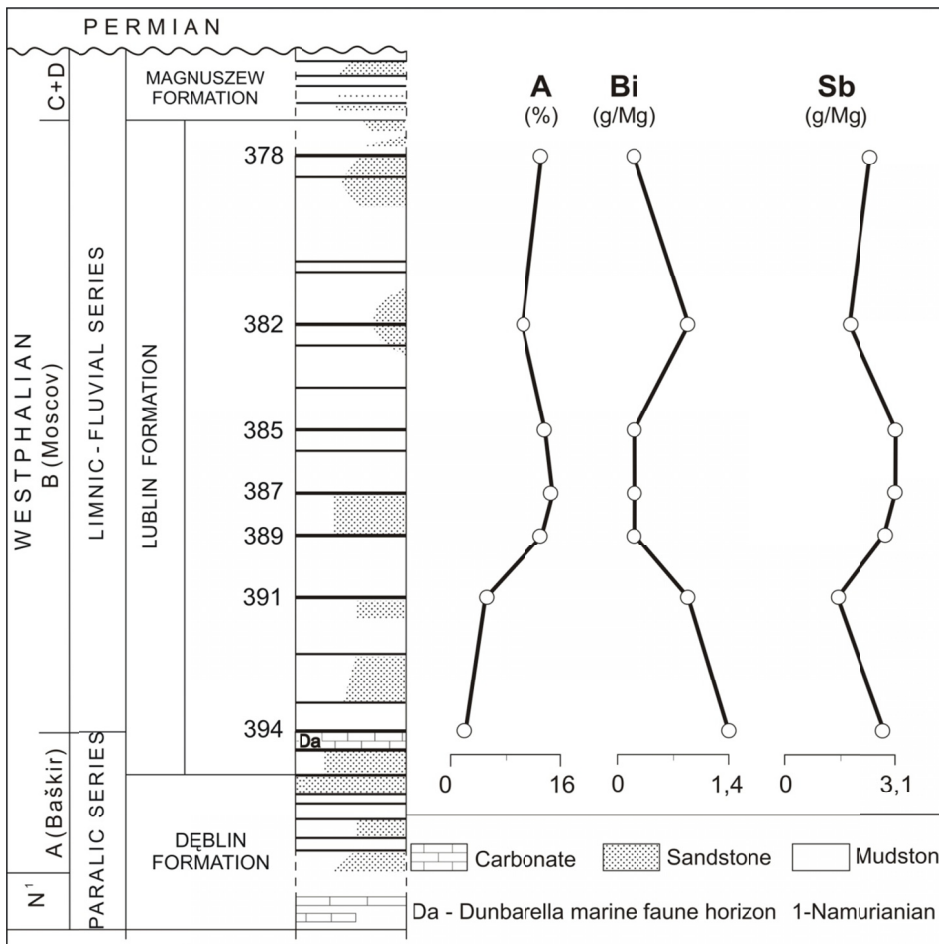


Fig. 2. Ash (A), bismuth and antimony content in coal seams in profile in LCB

for the roof part of seam (excluding 378 seam) is generally higher, the ash and Sb content in coal are roughly lower (excluding 378 seam due to the ash content and 391 due to the contents of Sb) than in coal from the middle part of seam. According to the low content of Bi i Sb, the potentially most preferred coal for combustion is coal from the floor part of 378 seam.

TABLE 2

Bi and Sb content in coal from LCB against the hard coal Clarke values by Ketris and Yudovich (2009)

Element	Range	Arithmetic mean	Geometric mean	Standard deviation	Hard coal Clarke values
Bi	0.1-2.4	0.6	0.4	0.6	1.1 ±0.1
Sb	0.1-6.1	2.2	1.5	1.3	1.00 ±0.09

The enrichment process in rare and trace elements of roof and floor part of seam or only from its roof or floor part in relation to the central part of the seam, occurs according to Yudovich and Ketris (2002) in two steps. This process lasts from the coverage of organic matter by the gangue and up to the end of the coal diagenesis. In a first step, the elements are concentrated on the border of the gangue with roof and floor layer of peat or lignite, having a thickness of about 10 cm. In the second step, the elements slowly diffuse into seam. Although Yudovich and Ketris did not specify how large is the group of elements covered by the

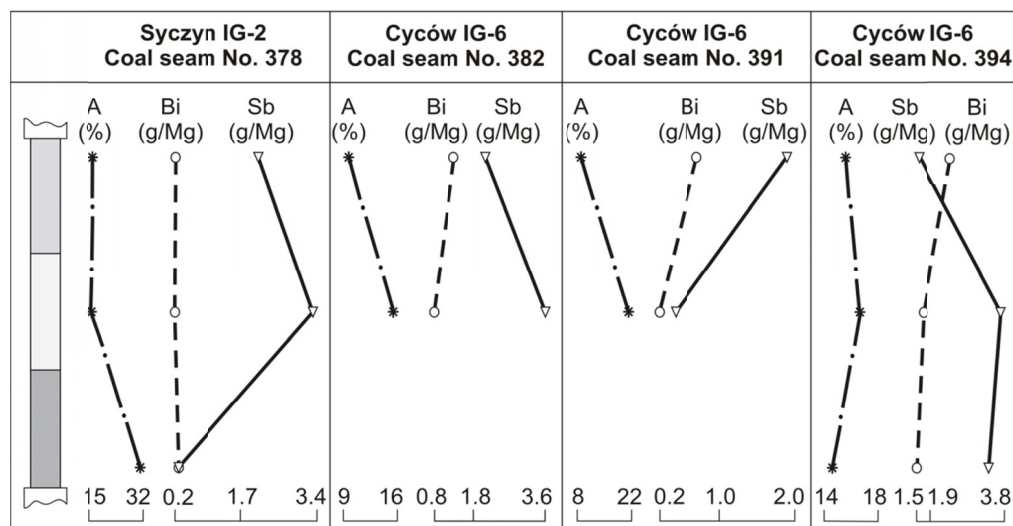


Fig. 3. Bi, Sb and ash (A) content in coal in vertical profile of the same coal seams in LCB

described processes, although in the present article we assumed that these processes could be the cause of Bi and Sb enrichment in roof and / or floor part of tested coal seams in LCB. Enrichment of specific parts of coal seams in certain trace elements have been already recorded in LCB (Cebulak, 1983) and in some parts of USCB (Cebulak, 1983; Hanak & Kokowska-Pawłowska, 2004, 2006; Idzikowski, 1959; Parzenty, 1989). In contrast, the described in this article higher

ash and Bi and Sb content in a coal form the middle part to the lower and upper part of the coal seams is a tendency rarely found. It is caused – according to Yudovich and Ketris (2002) – by:

- desorption of the elements in the outer part of peat and / or lignite due to increase in the pH of aqueous solutions and / or by diffusion of fulminan trace elements outside the seam and / or;
- disorders of elements sorption and diffusion in the surrounding of gangue interlayers in seams and / or the diffusion of elements from inserts carbonaceous rocks outside the coal seam.

Throughout the LCB a generally larger variety of Sb than of Bi content in coal was observed (Fig. 4). The content of Bi in coal through 378, 382, 385, 387 and 389 seams has low diversity. The values of Bi content between regions with taken samples from coal seams does not exceed 0,5 g/Mg. Only the differences between the Bi content in coal from 391 and 394 seam reach value of 1,7 g/Mg. On the other hand the dispersion of Sb content in coal seams on the extent of LCB is large and increase in the order of seams: 391 (1.7 g/Mg), 378 (1.6 g/Mg), 382 (2.6 g/Mg), 385 (3.0 g/Mg), 389 (3.9 g/Mg) i 387 (5.8 g/Mg).

3.3. Effect of organic and mineral matter on the content of Bi and Sb in coal

The calculated values of the Pearson correlation coefficient – equal to: -0.50 and -0.60 – indicates relationship between the increase in the content of Bi and Sb in coal ash, and

decrease of the the ash content of coal (Tab. 3). While the p level of significance for t-statistic values lower than of 0.05 means that the values of the correlation coefficient are significantly different from zero. The correlation coefficients – equal to: -0.20 and -0.06 and the significance level $p > 0.05$ – indicate an insignificant relationship between the content of Bi and Sb in coal and ash content. These dependencies may indicate a dominant influence of organic matter on the content of Bi and Sb in the examined LCB coal. This assumption is confirmed by the results of the RS function, according to which the 100% of Bi content and 97% of Sb content in coal is

TABLE 3

Bi and Sb distribution between the organic and mineral matter of coal from LCB, determined using RS function and Pearson correlation coefficient (r) between ash content (A) and Bi Sb content in coal and coal ash (C_{Bi} , C_{Sb})

Element	RS-Organic matter		RS-Mineral matter		$r (A/C_{Bi}, C_{Sb})$
	g/Mg	%	g/Mg	%	
In coal ash					
Bi	4.56	100.00	0.00	0.00	$-0.50 (p = 0.005)$
Sb	16.54	97.40	0.44	2.60	$-0.60 (p = 0.000)$
In coal					
Bi*	0.63	100.00	0.00	0.00	$-0.20 (p = 0.289)$
Sb*	2.30	97.46	0.06	2.54	$-0.06 (p = 0.761)$

* Correlation coefficient (r) for the other depending: $r_{Bi - As} = 0.48$; $r_{Bi - Fe} = 0.70$; $r_{Bi - Mn} = 0.60$; $r_{Bi - Mo} = 0.48$; $r_{Sb - Cu} = 0.48$; $r_{Sb - Mo} = 0.48$

* Współczynniki korelacji (r) dla innych zależności: $r_{Bi - As} = 0.48$; $r_{Bi - Fe} = 0.70$; $r_{Bi - Mn} = 0.60$; $r_{Bi - Mo} = 0.48$; $r_{Sb - Cu} = 0.48$; $r_{Sb - Mo} = 0.48$

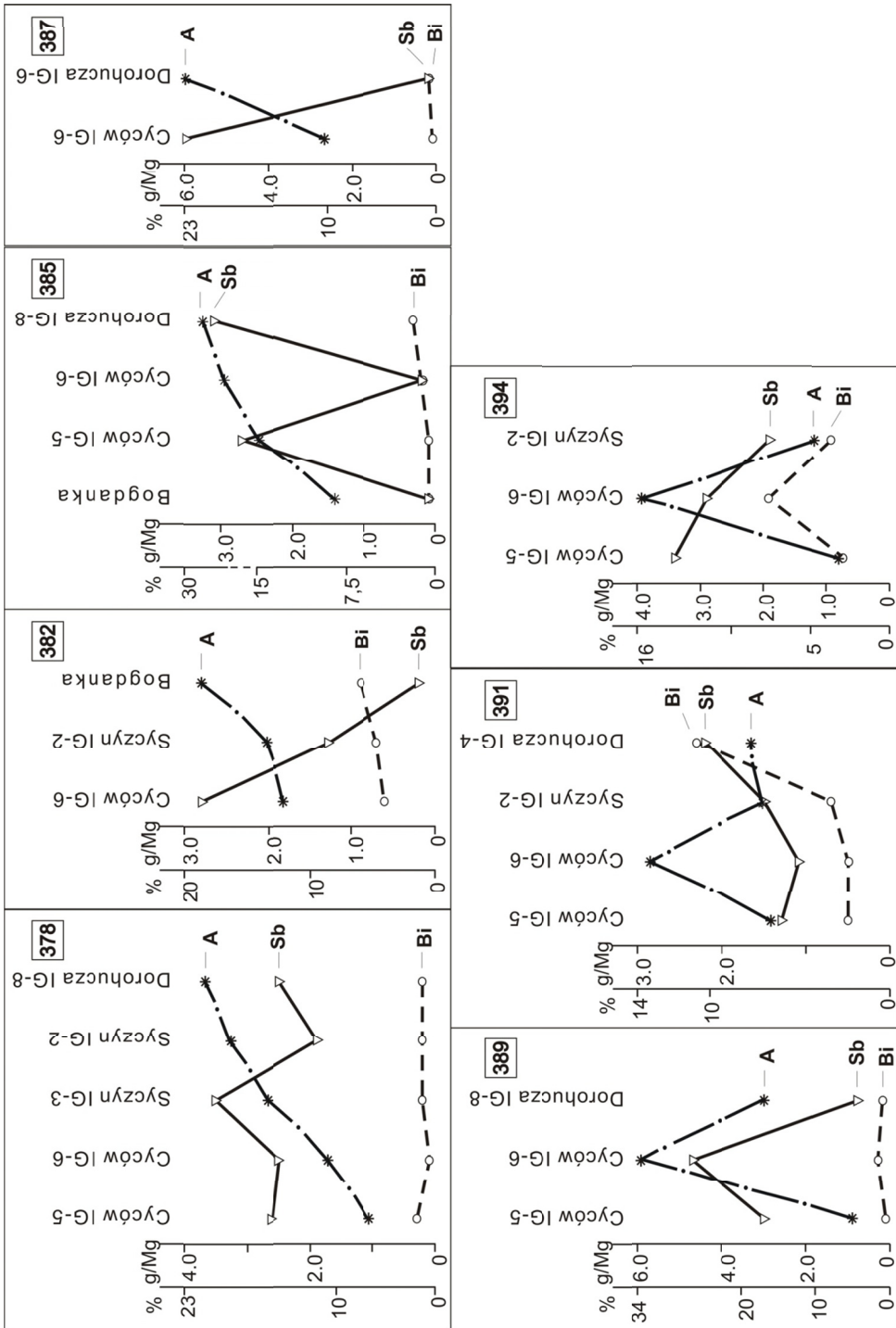


Fig. 4. Ash (A, %), bismuth (Bi, g/Mg) and antimony (Sb, g/Mg) content in coal in the along the stretch of same coal seams of LCB

derived from an organic matter. There was also a correlation relationship between the content of Bi and the content of As, Fe, Mn and Mo in the coal and between the Sb content and the content of Cu and Mo in coal. This indicates the possibility of organic matter coexistence in the LCB coal.

The observations of surface of the coal samples with an reflected light optical microscope and using a scanning electron microscope (SEM-EDS), did not reveal the presence of a Bi and Sb mineral inclusions in macerals, or admixtures of these elements in sulfide, sulfate, carbonate and clay minerals (Fig. 5). The presence of a homogeneous Bi and Sb mineral was not observed. There were only few complex peaks identifying the elements, as a result of Bi and Sb peaks compilation with the peaks of the main elements of organic matter (C, O) in a few grains of semifuzynite with fuzynite (Fig. 5). These macerals were located in a few cells with area lower than $5\mu\text{m}$ and were impregnated with pyrite or clay minerals. These findings indicate that Bi and Sb may be distributed in some macerals and minerals in coal, but because of the vast volume and

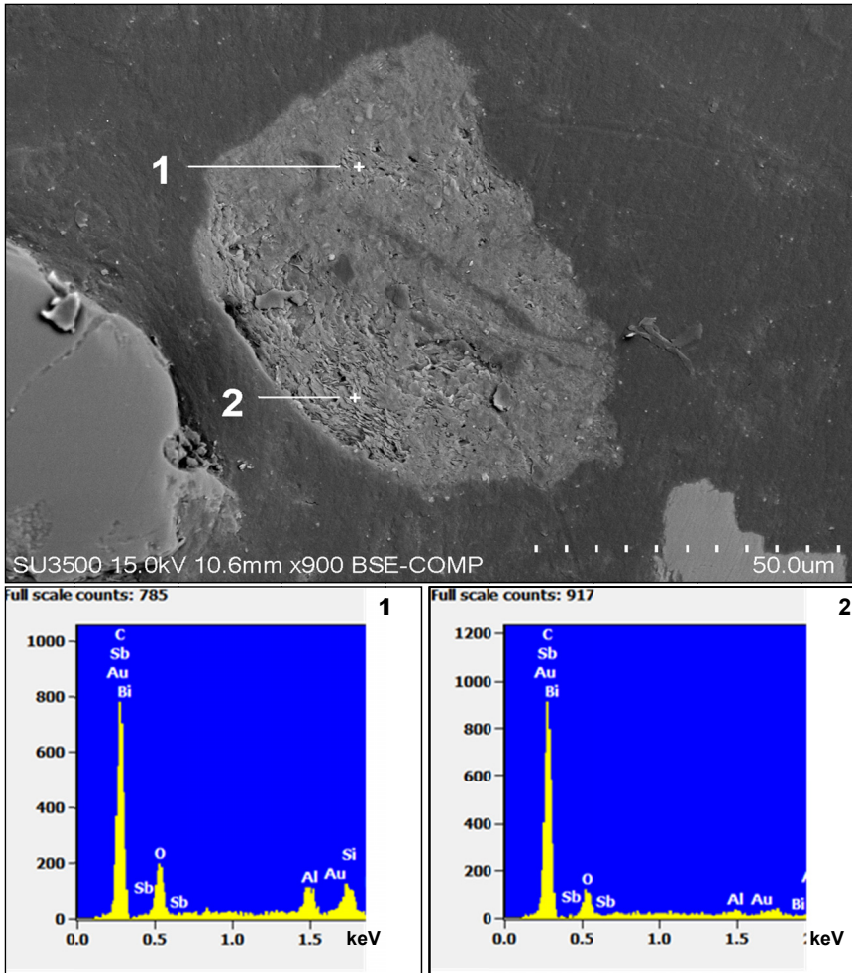


Fig. 5. Bi and Sb in semifuzynite (1) and fuzynite (2) determined by SEM-EDS

weight of the coal's macerals over the mineral substance, the organic substance has the greatest or only impact on the content of these elements in the coal.

The compound of elements with the organic matter is generally detrimental to the environment when using this coal for combustion. Discussed Bi and Sb elements in the temperature range 327°C-1127°C easily evaporates in furnaces and rapidly condenses on the particles and enriches the phase of lithophile elements as, among others: K, Ca, Na, Mg, Ti (Ratafia-Brown, 1994). Elements Bi and Sb are characterized by a relatively low value of ionization potential and a low melting and evaporation point. Therefore, they can be easily released into the atmosphere as volatile compounds and also Bi and Sb condenses on the smallest fly ash particles in the atmosphere and may be included in the content of particulate matter

4. Conclusions

1. The average contents of Bi and Sb in LCB coal are generally low and are close to the Clark value for bituminous coal:
 - 1.1. the content of Bi i Sb in LCB coal generally increases from younger to older seams;
 - 1.2. the content of Bi in coal for the roof part of seam is generally higher and the ash and Sb content in coal are roughly lower than in coal from the middle part of seam;
 - 1.3. the content of Bi in coal through 378, 382, 385, 387 and 389 seams has a low diversity. The values of Bi content between different parts of basin does not exceed 0.5 g/Mg. Only the differences between the Bi content in coal from 391 and 394 seam reach value of 1.7 g/Mg. The dispersion of Sb content in coal on the extent of LCB increase in the order of seams: 391 (1.7 g/Mg), 378 (1.6 g/Mg), 382 (2.6 g/Mg), 385 (3.0 g/Mg), 389 (3.9 g/Mg) i 387 (5.8 g/Mg).
2. The biggest influence on the content of Bi and Sb in LCB coal is probably organic matter in which these elements are scattered and do not form homogeneous minerals.

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