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Geochemical characteristics of fine bituminous coal from the Upper Silesian Coal Basin (Poland) and its potentially furnace waste

Introduction

A large number of the trace elements present in coal may be released into the environment during the extraction, cleaning and thermal processing of coal. The combustion of coal, particularly of low quality coal, contributes to the accumulation of high quantities of solid waste and exhaust gases in the environment. A large number and mass of ecotoxic elements are concentrated in this waste (e.g.: Davidson 1998; Strugała et al. 2014; Vassilev et al. 2005). Poland is the largest issuer of cadmium and lead from among 33 countries assessed by the European Environment Agency. Combustion processes, including mainly the combustion of raw materials in furnaces of power plants, manufacturing plants and in household furnaces have the largest share in the emission of ecotoxic elements to the environment in Poland (Makowska et al. 2017).

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The identification of petrographic coal components that have the greatest influence on the elemental content in coal makes it easier to forecast the efficiency of various procedures for purging ecotoxic elements (e.g.: Bhangare et al. 2011; Mohanty et al. 1998; Pan et al. 2018; Zhou et al. 2018); it also has an influence on more conscious forecasts concerning the quantities of elements released into the environment following coal combustion (Finkelmann 2004; Parzentny and Lewińska 2006; Parzentny and Róg 2001, 2007; Xu et al. 2003). High elemental contents in combustion waste can, in turn, become a resource for their recovery in the future (e.g.: Hanak and Kokowska-Pawłowska 2006; Strugała et al. 2014; Wdowin and Franus 2014; Zhang et al. 2015). The fine bituminous coal burned in power plants is typically characterized by high ash yields and ecotoxic element content. Although the trace element content in worldwide hard coal deposits is generally known (e.g., Finkelman et al. 2018; Ketris and Yudovich 2009; Yudovich and Ketri, 2005), there should be an increased focus on achieving a better understanding of the role of the feed coal mineral and organic matter in the concentration of ecotoxic elements in future combustion waste.

The aim of the article is to determine the ecotoxic trace element content in fine coal ash and to determine the influence of fine coal mineral and organic matter on the content of these elements in potential combustion waste. This article also intends to determine the quantity of trace elements that can be separated from potential combustion waste thanks to the separation of a fine coal fraction enriched in mineral matter. The fine coal was also subjected to a general assessment of its feasibility for combustion from ecological and technical perspectives. It was assumed for this assessment that the fine coal sold by mines is an approximate equivalent of feed coal, while the ash generated as a result of fine coal combustion at a temperature of 815°C under laboratory conditions is an approximate equivalent of the waste that can be generated in household furnaces or furnaces of power plants. This goal was to be accomplished by conducting studies of fine coal intended for combustion in power plants, originating from the Upper Silesian Coal Basin (USCB), which is the source of a significant amount of thermally processed fine coal in Europe.

1. Study scope and methodology

11 samples of fine coal intended for power plants with a size composition of 0–20.0 mm collected from USCB coal mines that are currently active or undergoing closure (Anna, Chwałowice, Jankowice, Marcel, Murcki, Piast, Pokój, Rydułtowy, Silesia, Staszic, Szczygłowice) were subjected to studies. The samples of coal fine have been collected in accordance with the standards specified in ISO 18283:2006 from the pile of fines, from undefined coal seams in a given mine. In order to determine the mode of occurrence of the elements in the fine coal, air-dry and fine (~50 µm) samples of the fine coal were parted in a mixture of dimethylbenzene, tetrachloromethane and tribromomethane into 8 density

Table 1. Petrographic and selected physico-chemical quality indices of the whole fine coal and its density fractions of fine coal from the coal mine in USCB

Tabela 1. Petrograficzne i niektóre fizykochemiczne wskaźniki jakości mianu węgla i frakcji gęstościowych mianu węgla z kopalni w GZW

Indices	Density fraction ($\times 10^3 \text{ kg/m}^3$)											whole coal	
	< 1.30	1.30–1.35	1.35–1.40	1.40–1.45	1.45–1.50	1.50–1.60	1.60–2.00	>2.00					
Q_{daf} (MJ/kg)	AA*	34586	34304	33802	33498	32265	30295	28089	16869				32825
	RC**	32520–35817	32218–35692	31826–35220	31826–35220	30973–33643	30295–30295	28089–28089	16869–16869				31165–34638
RI (%)	AA*	40.8	28.1	17.0	5.2	2.9	1.0	0.0	0.0				20.3
	RC**	0–83	0–74	0–58	0–58	0–15	0–9	0.0–0.0	0.0–0.0				0–77
R_t (%)	AA*	0.75	0.74	0.75	0.75	0.74	0.72	0.74	0.74				0.80
	RC**	0.54–0.90	0.56–0.80	0.55–0.89	0.57–0.89	0.54–0.88	0.55–0.87	0.56–0.90	0.52–0.91				0.53–0.94
% vol.													
Vt	AA*	77.5	73.8	70.9	63.8	41.6	39.2	23.3	3.0				56.7
	RC**	65–87	58–88	62–78	50–70	4–78	0–70	0–57	0–9				29–70
L	AA*	7.3	5.3	5.4	3.9	3.1	2.4	1.7	0.1				3.4
	RC**	3–14	0–13	0–13	0–12	0–6	0–7	0–7	0–1				0–7
I	AA*	9.3	16.7	15.8	16.5	14.0	12.8	7.2	0.9				13.5
	RC**	4–16	6–23	9–30	12–22	8–20	3–23	0–19	0–3				4–26
Mm	AA*	4.9	4.2	7.9	15.8	41.3	45.6	67.8	96.0				26.4
	RC**	0–16	0–13	2–18	4–36	7–87	9–97	33–100	89–100				11–64
wt%													
Yield	AA*	23.67	24.60	11.55	3.92	2.92	5.19	7.37	20.79				100.0
	RC**	0.50–56.34	10.0–63.0	3.97–30.00	1.07–10.00	0.34–8.00	2.00–11.00	2.85–16.00	7.0–28.0				100.0–100.0
A ^a	AA*	2.73	4.52	10.16	15.22	19.39	28.45	44.64	81.60				26.30
	RC**	1.43–4.58	2.35–6.09	6.85–12.99	7.92–18.19	13.12–24.07	25.87–30.39	31.91–50.92	74.72–86.38				10.95–35.45
V_{daf}	AA*	36.19	34.12	33.72	34.52	35.59	38.55	na***	na***				34.99
	RC**	32.59–43.56	31.01–37.96	30.34–37.77	30.72–38.67	31.77–67.86	38.55–38.55	na***	na***				31.23–39.10

* Arithmetic average, ** range of values, *** not analyzed.

fractions, i.e.: <1.30, 1.30–1.35, 1.35–1.40, 1.40–1.45, 1.45–1.50, 1.50–1.60, 1.60–2.00 and >2.00 × 10³ kg/m³. The following was determined for these fractions and fine whole coal samples:

- ◆ vitrinite (V_t), liptinite (L), inertinite (I) and mineral matter (Mm) content; reflectance (R_r) was measured using a Zeiss Axio Imager D1m optical microscope (reflected white light, immersion oil) with an integration table. The applied microscopic procedures were in accordance with the standards specified by the International Committee for Coal and Organic Petrology and as described in ISO 7404-3 (2009a) and ISO 7404-5 (2009b). The results are shown in Table 1.
- ◆ ash yield (A^a₈₁₅) and volatile component (V^{daf}) content, heat of combustion (Q_s^{daf}), the Roga caking index (RI) and total sulfur (S_t^a) and pyritic sulfur (S_p^a) content. The analyses were carried out according to ISO/TS 12901 (2001). The results are shown in Table 1.

An X-ray fluorescence spectrometer (Rigaku ZSX Primus II, Rh anode tube power = max. 4 kW, 50 kV/60 mA and analytical crystals: PET, LiF1, Rx25, Ge) was used to determine the content of the following in the fine coal ash (whole and parted into fractions):

- ◆ oxides of major elements, i.e.: Si, Al, Fe, Ca, Mg, Na, K, S, Ti and P; the results are shown in Table 2.
- ◆ certain trace elements, i.e.: As, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb and Zn; the results are shown in Table 3.

The content by point of: As, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb and Zn in macerals and minerals was determined for the polished sections and natural surface samples of the fine coal density fractions with the greatest significance concerning the concentration of the aforementioned trace elements in fine coal ash and fine raw coal (<1.30 × 10³ kg/m³ fraction). The analysis was performed using a HITACHI SU-3500N (Analytical conditions were as follows: Acc. voltage = 15.0 kV, bse-comp = 30 Pa, image resolution = 1024 × 768, image pixel size = 0.040.27 μm, magnification = ×90–5000) universal digital scanning electron microscope. The analysis results are presented in Figure 2 and 3.

2. Obtaining the results

Using Pearson's χ^2 test and the Kolmogorov-Smirnov test ($p = 0.05$), it was determined that the values of all the aforementioned petrographic, physico-chemical and geochemical coal quality indices had normal or close to normal distribution, while the arithmetic average value was the best way to estimate the average values of the determined indices. The calculation results are presented in Table 1.

The role (% M) of each (i) density fraction in concentrating the representative and trace elements in whole fine coal ash, as well as in concentrating the total and pyritic sulfur in the fine whole coal, was determined based on the formula calculated on the basis of the formula:

Table 2. Total and pyritic sulfur (S_t^a , S_p^a) content in the fine coal as well as major element oxides content in the fine coal ash and in the density fraction ash of the fine coal from the coal mine in USCB

Tabela 2. Zawartość siarki całkowitej i pirytovej (S_t^a , S_p^a) w miale węgla oraz zawartość tlenków głównych pierwiastków w popiele mialu węgla i w popiele frakcji gęstościowych mialu węgla z kopalni w GZAW

Object	S_t^a		S_p^a		SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	P_2O_5
	wt% in coal		wt% in coal ash									
LCB coal ¹	1.96	no data	30.30	29.59	15.66	3.82	1.41	0.34	1.46	no data		
USCB coal ²	1.13	0.29–1.39	34.86	22.16	19.54	9.12	4.59	4.58	1.25	0.04		
Researched fine coal												
Whole fine coal	AA*	0.71±0.10	53.21±1.05	23.35±1.37	7.70±0.50	3.69±0.71	3.07±0.34	0.98±0.16	2.73±0.21	0.33±0.05		
	RC**	0.35–1.49	47.49–57.73	11.87–27.96	5.15–10.37	1.24–9.20	1.90–6.15	0.49–2.28	1.32–3.35	0.10–0.67		
<1.30 · 10 ³ kg/m ³	AA*	0.46±0.03	0.10±0.02	32.17±2.17	28.05±1.65	10.24±0.85	7.22±0.50	3.18±0.28	1.34±0.15	1.89±0.41		
	RC**	0.26–0.71	0.05–0.33	22.14–40.98	15.91–33.17	5.78–15.33	4.80–10.60	1.93–5.43	0.81–2.11	0.11–4.09		
1.30–1.35	AA*	0.54±0.04	0.15±0.02	35.68±2.05	27.48±1.48	9.49±1.01	6.97±0.78	3.37±0.44	1.53±0.14	2.07±0.36		
	RC**	0.32–0.82	0.09–0.25	24.27–42.61	15.67–32.64	5.23–14.20	3.87–12.80	1.65–7.13	0.80–6.75	0.72–2.21	0.26–4.09	
1.35–1.40	AA*	0.71±0.06	0.29±0.04	41.12±2.07	26.43±1.60	9.04±1.09	6.18±1.34	3.58±0.77	1.97±0.18	1.29±0.21		
	RC**	0.44–1.10	0.16–0.55	27.36–50.75	12.76–32.60	5.17–16.23	2.55–18.47	1.71–10.94	0.62–4.13	0.94–2.56	0.18–2–47	
1.40–1.45	AA*	0.80±0.07	0.40±0.06	42.93±2.25	26.13±1.64	9.03±1.10	6.04±1.72	3.78±0.93	1.22±0.23	2.21±0.20	0.91±0.15	
	RC**	0.43–1.24	0.19–0.71	25.22–53.32	12.43–31.88	5.67–10.99	1.68–22.18	1.65–12.80	0.54–3.19	0.71–2.80	0.16–1.79	
1.45–1.50	AA*	0.89±0.08	0.45±0.06	43.82±2.40	25.82±1.74	8.80±0.97	6.00±1.98	3.93±1.10	1.04±0.18	2.35±0.19	0.69±0.12	
	RC**	0.57–1.44	0.24–0.90	23.48–54.96	10.84–32.36	5.39–14.90	1.78–24.95	1.73–14.61	0.50–2.75	0.72–2.83	0.12–1.44	
1.50–1.60	AA*	1.04±0.11	0.58±0.10	46.06±1.53	25.06±1.87	8.39±0.72	5.81±1.70	3.95±0.96	0.90±0.14	2.51±0.19	0.50±0.09	
	RC**	0.64–1.91	0.25–1.32	35.97–55.60	8.69–32.03	5.43–12.71	1.60–21.34	1.66–12.94	0.39–2.12	0.86–3.20	0.10–1.17	
1.60–2.00	AA*	1.12±0.17	0.83±0.17	47.35±2.02	24.99±1.49	7.74±0.62	5.85±0.08	4.26±1.07	0.73±0.08	2.86±0.17	0.34±0.08	
	RC**	0.46–2.70	0.24–2.38	28.50–53.55	12.67–31.34	5.10–11.35	1.64–23.31	1.85–14.48	0.37–1.37	1.44–3.39	0.08–0.96	
>2.00	AA*	1.01±0.30	0.98±0.30	58.82±1.26	23.33±1.35	7.00±0.48	1.58±0.34	2.27±0.17	0.53±0.05	3.15±0.18	0.13±0.01	
	RC**	0.13–3.60	0.11–3.62	53.55–68.52	10.72–27.30	4.55–10.28	0.38–4.72	1.66–3.77	0.27–0.85	1.63–3.78	0.06–0.20	

¹ Results per Cebulak 1983, Porzycki and Zdanowski 1995; ² Results per Jureczka and Kotas 1995, Mielecki and Krzyżanowska 1961.

* Arithmetic average; ** Range of values.

Table 3. Trace element contents in whole fine coal ash and density fraction ash of fine coal from the coal mine in USCB
 Tabela 3. Zawartość pierwiastków śladowych w popiele nierozdzielonego miatu węgla i w popiele frakcji gęstościowych miatu węgla z kopalni w GZW

Object	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	Sb	Zn	
World coal ash ¹	46±5	1.20±0.30	37±2	120±5	110±5	14±1	100±5	55±6	7.5±0.6	170±10	
Researched fine coal ash											
Whole fine coal ash	AA*	7.6±2.9	1.0±0.0	34.0±3.0	68.9±7.1	78.7±26.9	1.0±0.0	25.3±5.8	99.4±11.3	1.5±0.3	478.1±150.0
	RC**	1–28	1–1	20–53	44–115	1–224	1–1	1–56	71–207	1–4	83–1843
<1.30 · 10 ⁹ kg/m ³	AA*	40.3±10.4	2.0±0.7	312.8±47.1	287.1±37.2	695.4±126.4	7.9±3.0	767.1±114.0	270.5±33.2	28.1±23.8	6909.6±2805.5
	RC**	1–109	1–7	113–552	97–464	150–1382	1–33	363–1748	103–470	1–266	509–24063
1.30–1.35	AA*	28.5±6.5	1.5±0.5	161.7±25.6	266.4±52.3	688.0±250.5	2.9±1.9	365.5±51.3	219.2±28.3	27.4±23.9	4549.4±1707.0
	RC**	1–74	1–6	58–292	51–639	150–3027	1–22	153–627	130–470	1–266	282–12548
1.35–1.40	AA*	29.0±9.3	1.5±0.4	91.8±16.4	177.2±35.3	342.8±84.1	1.0±0.0	168.8±29.0	177.5±16.1	16.1±14.1	4613.4±1863.8
	RC**	1–98	1–6	28–205	22–419	34–805	1–1	1–331	59–257	1–157	237–14255
1.40–1.45	AA*	26.0±6.5	1.4±0.4	66.5±15.7	145.6±25.7	245.5±55.8	1.0±0.0	107.7±21.6	164.6±19.2	14.0±12.4	3475.3±1399.4
	RC**	1–69	1–5	12–199	1–287	21–606	1–1	1–230	12–233	1–138	342–11426
1.45–1.50	AA*	22.5±7.1	1.5±0.4	50.5±8.4	136.5±27.0	323.9±132.6	1.0±0.0	115.8±25.4	147.6±17.0	1.7±0.6	3016.0±1175.7
	RC**	1–76	1–6	17–109	1–326	1–1571	1–1	1–319	14–225	1–7	222–9487
1.50–1.60	AA*	25.7±10.2	1.5±0.5	40.6±7.3	102.7±15.8	172.7±43.1	1.0±0.0	58.7±13.7	132.8±14.1	1.2±0.2	3654.8±1451.3
	RC**	1–96	1–7	14–84	16–187	1–459	1–1	3–119	21–196	1–3	258–11511
1.60–2.00	AA*	16.8±6.0	1.0±0.0	40.8±12.5	71.2±12.2	297.7±107.2	1.0±0.0	31.5±8.7	122.5±14.2	1.6±0.4	3592.4±1407.0
	RC**	1–55	1–1	12–155	1–156	1–993	1–1	1–81	77–209	1–5	177–10617
>2.00	AA*	5.7±2.7	1.0±0.0	30.7±7.0	70.0±7.2	25.7±11.8	1.0±0.0	6.1±2.5	61.0±6.7	1.5±0.4	2335.3±863.8
	RC**	1–30	1–1	5–65	44–119	1–102	1–1	1–29	30–120	1–5	109–7822

¹ Results per Kettis and Yudovich 2009.

* Arithmetic average; ** Range of values.

$$(\%M) = (U_i \cdot A_i \cdot C_i) / \sum(U_i \cdot A_i \cdot C_i) \quad (1)$$

- ↗ U_i – yield of the i -density fraction (%),
 A_i – ash yield of the i -density fraction (%),
 C_i – trace element or element oxides content in ash of the i -density fraction of the analyzed fine coal (%).

The calculation results are presented in Figure 1.

In order to perform a general technical quality index assessment of the fine coal as potential feed coal, the values of three dimensionless indices were calculated, i.e.:

- ◆ the slagging index (Babcock Index)

$$BI = \frac{Fe_2O_3 + CaO + MgO + Na_2O + K_2O}{SiO_2 + Al_2O_3 + TiO_2} S_t^d \quad (2)$$

- ◆ the silica contribution index (Silica Value)

$$SV = \frac{SiO_2}{SiO_2 + Fe_2O_3 + CaO + MgO} 100 \quad (3)$$

- ◆ the heating surface fouling propensity index (Fouling Index)

$$FI = \frac{Na_2O + K_2O}{S_t^d} BI \quad (4)$$

- ↗ Fe_2O_3, CaO, \dots – representative element oxide content in fine coal ash (wt%),
 S_t^d – total sulfur content in dry coal (wt%).

After Zelkowski (1986), the following interpretation of the calculated index values was adopted:

- ◆ coal with low slagging propensity, when $BI < 0.6$ or $SV > 72$,
- ◆ coal with medium slagging propensity, where $0.6 < BI < 2.0$ or $65 < SV < 72$,
- ◆ coal with high slagging propensity, where $2.0 < BI < 2.6$ or $SV < 65$,
- ◆ coal with very high slagging propensity, where $BI > 2.6$,
- ◆ no coal boiler fouling propensity, where $FI < 0.6$,
- ◆ high coal boiler fouling propensity, where $0.6 < FI < 40.0$,
- ◆ very high coal boiler fouling propensity, where $FI > 40.0$.

Calculation results are presented in Table 4.

In order to determine the mode of occurrence of the discussed trace elements in the whole coal fines and its density fractions, the values of the correlation coefficient between the content of trace elements in ash of the whole coal fines and in the ash of its density

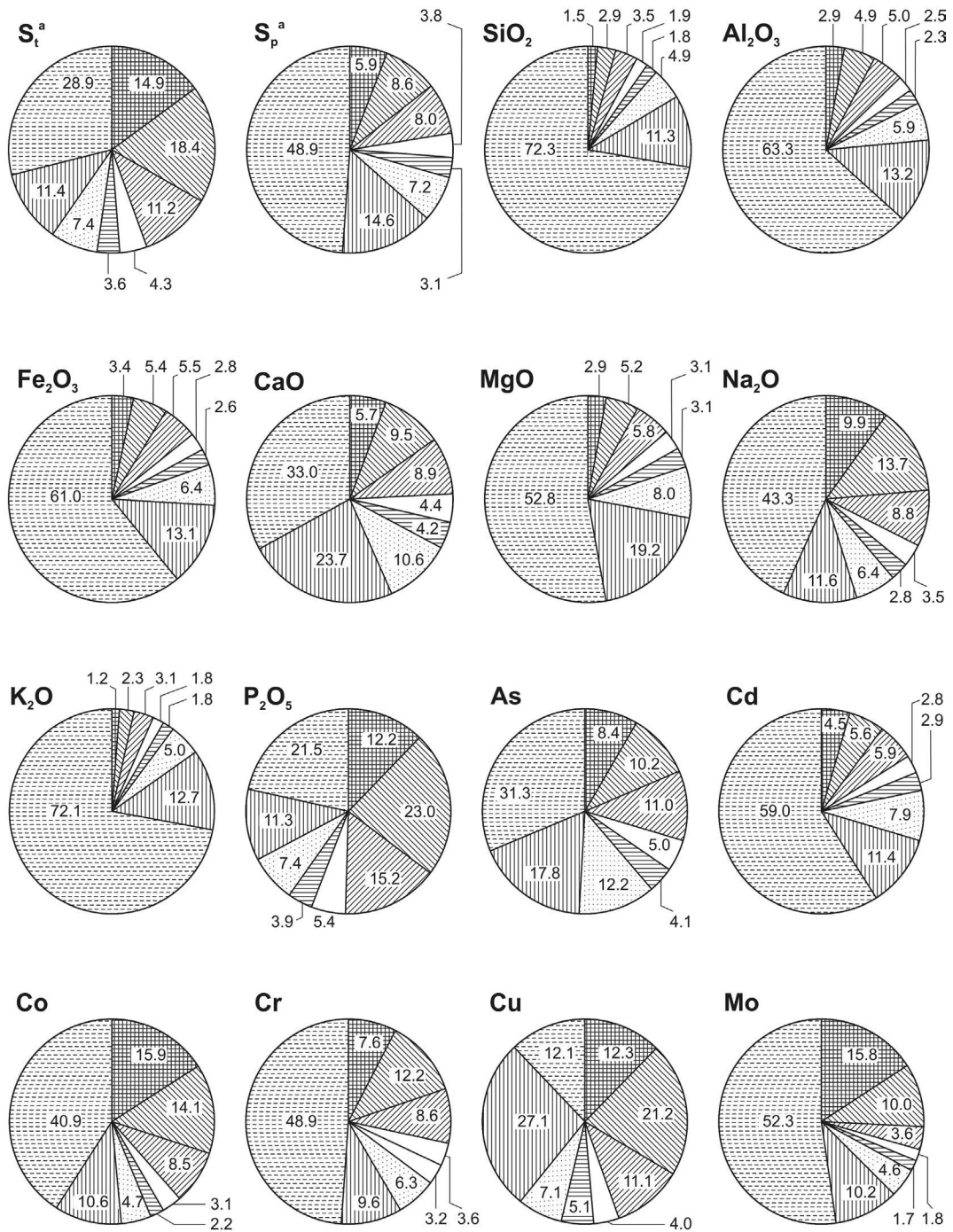


Fig. 1.

Rys. 1.

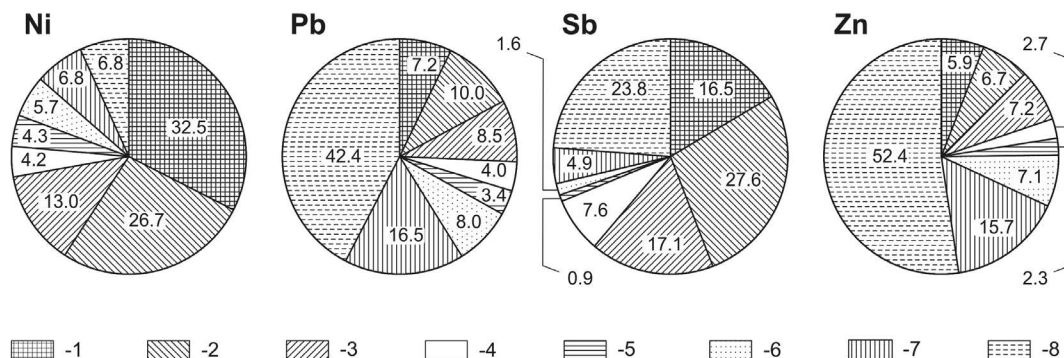


Fig. 1. Contribution (wt%) of the examined density fractions to the concentration of major element oxides and ecotoxic trace elements in whole fine coal ash as well as sulfur in whole fine coal from coal mines in USCB

* Density fractions ($\times 10^3 \text{ kg/m}^3$): 1–<1.30, 2–1.30–1.35, 3–1.35–1.40, 4–1.40–1.45, 5–1.45–1.50, 6–1.50–1.60, 7–1.60–2.00, 8–>2.00

Rys. 1. Udział (% wag.) badanych frakcji gęstościowych w koncentracji tlenków głównych pierwiastków i ekotoksycznych pierwiastków śladowych w popiele nierozdzielonego mialu węgla oraz w koncentracji siarki w nierozdzielonym miale węgla z kopalń w GZW

* Frakcje gęstościowe ($\times 10^3 \text{ kg/m}^3$): 1–<1,30, 2–1,30–1,35, 3–1,35–1,40, 4–1,40–1,45, 5–1,45–1,50, 6–1,50–1,60, 7–1,60–2,00, 8–>2,00

fractions and the content of petrographic components of coal fines and the content of the main elements oxides in coal ash and in the ashes of its density fractions. The calculation results have been presented in Table 5.

Table 4. Quality index of the whole fine coal and density fractions of the fine coal from the coal mine in USCB

Tabela 4. Wskaźniki jakości mialu węgla i gęstościowych frakcji mialu węgla z kopalń w GZW

Factor	Density fraction ($\times 10^3 \text{ kg/m}^3$)								
	< 1.30	1.30–1.35	1.35–1.40	1.40–1.45	1.45–1.50	1.50–1.60	1.60–2.00	> 2.00	whole coal
BI*	0.19	0.20	0.23	0.25	0.28	0.31	0.33	0.18	0.17
SV*	60.88	64.27	68.61	69.49	70.05	71.73	72.62	84.43	78.63
FI*	1.86	1.51	1.14	1.09	1.06	1.02	1.05	0.64	0.87

* BI – slagging index (Babcock Index), SV – silica contribution index (Silica Value), FI – heating surface fouling propensity index (Fouling Index).

Table 5. Correlation coefficients (r) of trace elements in fine coal ash with petrographical components of fine coal and sulfur in fine coal and major oxide elements in fine coal ash from mine coal in USCB ($p < 0.05$)

Tabela 5. Współczynnik korelacji (r) dla zależności między zawartością pierwiastków śladowych w popiele miazła węgla, a składem petrograficznym miazła węgla, zawartością siarki w miazle węgla i zawartością tlenków głównych pierwiastków w popiele miazła węgla z kopalń w GZW ($p < 0,05$)

Element	Fraction ($\times 10^3$ kg/m ³)	Vitrynite (V _t) liptynite (L) inertynite (I) mineral m.(M _m)	Pyritic sulfur (S _p) Fe ₂ O ₃	CaO MgO P ₂ O ₅	SiO ₂ Al ₂ O ₃ Na ₂ O K ₂ O
As	<1.30	rV _t = 0.726		rMgO = 0.795	
	1.30–1.35	rV _t = 0.865			rSiO ₂ = 0.766
					rK ₂ O = 0.805
Cd					
Co	1.35–1.40				rSiO ₂ = 0.738
	1.40–1.45				rSiO ₂ = 0.824
					rAl ₂ O ₃ = 0.771
					rK ₂ O = 0.787
	1.45–1.50				rSiO ₂ = 0.749
					rAl ₂ O ₃ = 0.852
					rK ₂ O = 0.804
	1.50–1.60	M _m = 0.745			
Cr	1.30–1.35				rSiO ₂ = 0.716
					rAl ₂ O ₃ = 0.709
	1.35–1.40				rSiO ₂ = 0.722
					rAl ₂ O ₃ = 0.833
	1.40–1.45				rSiO ₂ = 0.753
					rAl ₂ O ₃ = 0.892
	1.45–1.50				rAl ₂ O ₃ = 0.889
	1.50–1.60				rAl ₂ O ₃ = 0.894
					rK ₂ O = 0.707
	1.60–2.00				rSiO ₂ = 0.733
				rAl ₂ O ₃ = 0.968	
				rK ₂ O = 0.795	
Cu	<1.30	rI = 0.747	rFe ₂ O ₃ = 0.732		
	1.30–1.35		rSp = 0.781		
	1.45–1.50	rL = 0.832			
	1.60–2.00	rV _t = 0.764			
	whole coal	rL = 0.702			
Mo					

Table 5. cont.

Tabela 5. cd.

Element	Fraction ($\times 10^3$ kg/m ³)	Vitrynite (V _t) liptynite (L) inertynite (I) mineral m.(M _m)	Pyritic sulfur (S _p) Fe ₂ O ₃	CaO MgO P ₂ O ₅	SiO ₂ Al ₂ O ₃ Na ₂ O K ₂ O
Ni	1.30–1.35				rSiO ₂ = 0.805
	1.35–1.40				rSiO ₂ = 0.745
					rAl ₂ O ₃ = 0.910
	1.40–1.45				rSiO ₂ = 0.827
					rAl ₂ O ₃ = 0.960
					rK ₂ O = 0.813
	1.45–1.50				rAl ₂ O ₃ = 0.922
					rK ₂ O = 0.791
	1.50–1.60				rAl ₂ O ₃ = 0.834
1.60–2.00				rAl ₂ O ₃ = 0.820	
Pb	1.35–1.40				rSiO ₂ = 0.752
					rAl ₂ O ₃ = 0.924
	1.50–1.60				rSiO ₂ = 0.789
					rAl ₂ O ₃ = 0.970
					rK ₂ O = 0.919
	>2.00		rFe ₂ O ₃ = 0.706		
	whole coal		rFe ₂ O ₃ = 0.632		rNa ₂ O = 0.759
		rSp = 0.886			
Sb	<1.30			rP ₂ O ₅ = 0.747	
	1.30–1.35			rP ₂ O ₅ = 0.719	
Zn	<1.30	rL = 0.834			
		rI = 0.931			
	1.30–1.35	rL = 0.829			
	1.35–1.40	rL = 0.861			
	1.40–1.45	rL = 0.947			
	1.45–1.50	rV _t = 0.968			
		rL = 0.902			
	1.50–1.60	rV _t = 0.897			
		rL = 0.959			
	1.60–2.00	rV _t = 0.984			
		rL = 0.838			
		rI = 0.955			
	>2.00	rV _t = 0.926			
	whole coal	rL = 0.639	rFe ₂ O ₃ = 0.619		rNa ₂ O = 0.752
		rSp = 0.796			

3. Results and their interpretation

3.1. Fine coal petrographic and physico-chemical quality index assessment

Regarding the random reflectance (R_r) of vitrinite (Table 1), feed coals originating from the LCB and the USCB are classified as ortho-bituminous coal. The average ash yields for the studied feed coals (Table 1) are similar to or slightly higher than those of the most commonly used coals burned in Poland (12–30% according to [Dubiński et al. 2005](#); 23.82–29.17% according to [Smółka-Danielowska 2013](#)). When compared to flame coal, which according to [Gabzdyl \(1999\)](#) is the most commonly burned coal in Poland ($V^{\text{daf}} = 36\text{--}43\%$, $Q_s^{\text{daf}} = 29.1\text{--}33$ MJ/kg), the average volatile component content in the studied fine coal is low ($V^{\text{daf}} = 35\%$), while the average heat of the combustion value ($Q_s^{\text{daf}} = 32.8$ MJ/kg) is similar. The studied fine coal Roga caking index ($RI = 20.27$) is generally low compared to the USCB fine power coal ($RI = 29.1$; per [Mielecki et al. 1963](#)), which indicates the suitability of the fine coal for combustion and its unsuitability for coking.

The total sulfur content in the studied fine coal is $S_t^a = 0.71\%$ (Table 2) and is similar to the feed coal burned in power plants in Poland ($S_t^a = 0.6\text{--}1.2$ wt%; [Dubiński et al. 2005](#)). The (Table 1) fraction enriched with vitrinite with a density $1.30\text{--}1.35 \times 10^3$ kg/m³ and the fraction enriched with mineral matter with a density of $1.60\text{--}2.00 \times 10^3$ kg/m³ (Fig. 1) has the largest share in sulfur concentration (S_t^a , S_p^a) in the discussed fine coal ash (apart from the fraction with a density of $>2.00 \times 10^3$ kg/m³). The purification of the examined fine coal from the mineral matter to be burned may cause a reduction of sulfur release from the coal combustion chamber by approx. 40%. This would be an advantage especially for the environment and for reducing the rate of corrosion of the heating devices in the power plant.

A large range of changes in the values of petrographic and physical-chemical indexes of coal fines (Table 1), as well as the content of the element content, discussed below, in fine coal ash (Table 2 and 3) are noteworthy. This coal fine variability is due to the fact that every sample of fine coal is a mixture of coal from not one but most often from several coal deposits operated in a given mine. Each mine in which samples of coal have been collected also operate in coal seams from various lithostratigraphic members in the Upper Silesian Coal Basin. Therefore, coal often has a very different suitability for combustion and other thermal processing methods. The standardization of the quality of fine coal targeted for combustion has been the goal of lignite and hard coal mining in Poland for years ([Dubiński et al. 2005](#); [Kurczabiński 1996](#); [Uliasz-Bochenczyk and Mokrzycki 2007](#)).

3.2. Fine coal geochemical characteristics

In the chemical composition of fine coal ash SiO_2 predominates over Al_2O_3 , and the combined content of both oxides is definitely higher than the content of Fe, Ca, Mg, Na,

K and P oxides in fine coal ash (Table 2). Compared to the results of raw coal from LCB and USCB, the content of SiO_2 , K_2O and P_2O_5 in the discussed fine coal ash is higher, and the content of Fe_2O_3 is clearly smaller. The largest share in the concentration of: Si, Al, Fe, Ca, Mg, Na, K and P oxides in coal fine ash, and thus in future furnace waste, is found in (except for the mineral matter with a density $> 2.00 \times 10^3 \text{ kg/m}^3$) the coal fine fraction enriched with mineral matter with the density of $1.60\text{--}2.00 \times 10^3 \text{ kg/m}^3$ (for oxides of: Si, Al, Fe, Ca, Mg and K) and the coal fine fraction enriched with vitrinite with the density of $1.30\text{--}1.35 \times 10^3 \text{ kg/m}^3$ (for Na and P oxides) (Fig. 1). It is presumed that the: Al, Fe, Ca, Mg, Na and K in the USCB coal vitrinite are most commonly connected with scattered syndimentary minerals and with aliphatic and heterocyclic compounds originating from coal-forming vegetation (Fabiańska and Parzentny 1993, Lewińska-Preis et al. 2008; Zubovic et al. 1964). It may be forecasted that the cleaning of coal fines from mineral matter before its combustion will reduce the content of: Si, Al, Fe, Ca, Mg, Na, K and P oxides in fine coal ash and thus in future furnace waste by app 33% to 85 %, and thus also the mass of furnace waste generated will decrease.

The calculated average index values for BI (0.17), SV (78.63) and FI (0.87) indicate a low propensity of the studied fine coal for slagging furnaces and fouling power boilers with sediments (Table 4). The SV index reaches >72 for fine coal components with densities over $1.50 \times 10^3 \text{ kg/m}^3$, while an SV value of <65 was noted for the $<1.35 \times 10^3 \text{ kg/m}^3$ -density fraction enriched in vitrinite. These empirical tendencies indicate that there is no sense in burning fine coal devoid of mineral matter due to its high slagging propensity, which has already been discussed by, among others, Collot (2006), and Hamala and Róg (2004).

Compared to hard coal ash from worldwide deposits, the Pb and Zn contents in the studied raw fine coal ash are considerably greater, while the: As, Cd, Co, Cr, Cu, Mo, Ni and Sb contents in the raw fine coal ash are lower (Table 3). Extremely high zinc content (24 063 g/Mg) was found in several $<1.30 \times 10^3 \text{ kg/m}^3$ -density fine coal fraction ash samples. Such levels of zinc concentration were already noted in bituminous coal ash originating from deposits in Poland and worldwide; they can be considered a potential source of its recovery (e.g.: Parzentny 1990; Strugała et al. 2014; Zhang et al. 2015).

It was observed that: As, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb and Zn contents are the greatest in $<1.30 \times 10^3 \text{ kg/m}^3$ -density studied fine coal fraction ash, and the lowest in $>2.00 \times 10^3 \text{ kg/m}^3$ -density fraction ash (Table 3). Furthermore, the $<1.35 \times 10^3 \text{ kg/m}^3$ -density fraction ash has the greatest influence on the content of these elements in whole fine coal ash. Aluminosilicate and iron oxide particles are the main element carriers in the $<1.35 \times 10^3 \text{ kg/m}^3$ -density fine coal ash, and thus also in future combustion waste (Fig. 2). The particles of these phases were most likely generated as a result of $<1.35 \times 10^3 \text{ kg/m}^3$ -density fine coal fraction component combustion, i.e. mostly fusinite and aluminosilicates, as well as intergrowths of these macerals with ankerite and pyrite (Fig. 3).

Due to the high ash yield of mineral matter (Table 1), the ash of the fraction with the density of $> 2.00 \times 10^3 \text{ kg/m}^3$ has the greatest impact on the content of As, Cd, Co, Cr, Mo, Pb and Zn in ash of whole coal fines (Fig. 1). In turn, the largest impact on the content

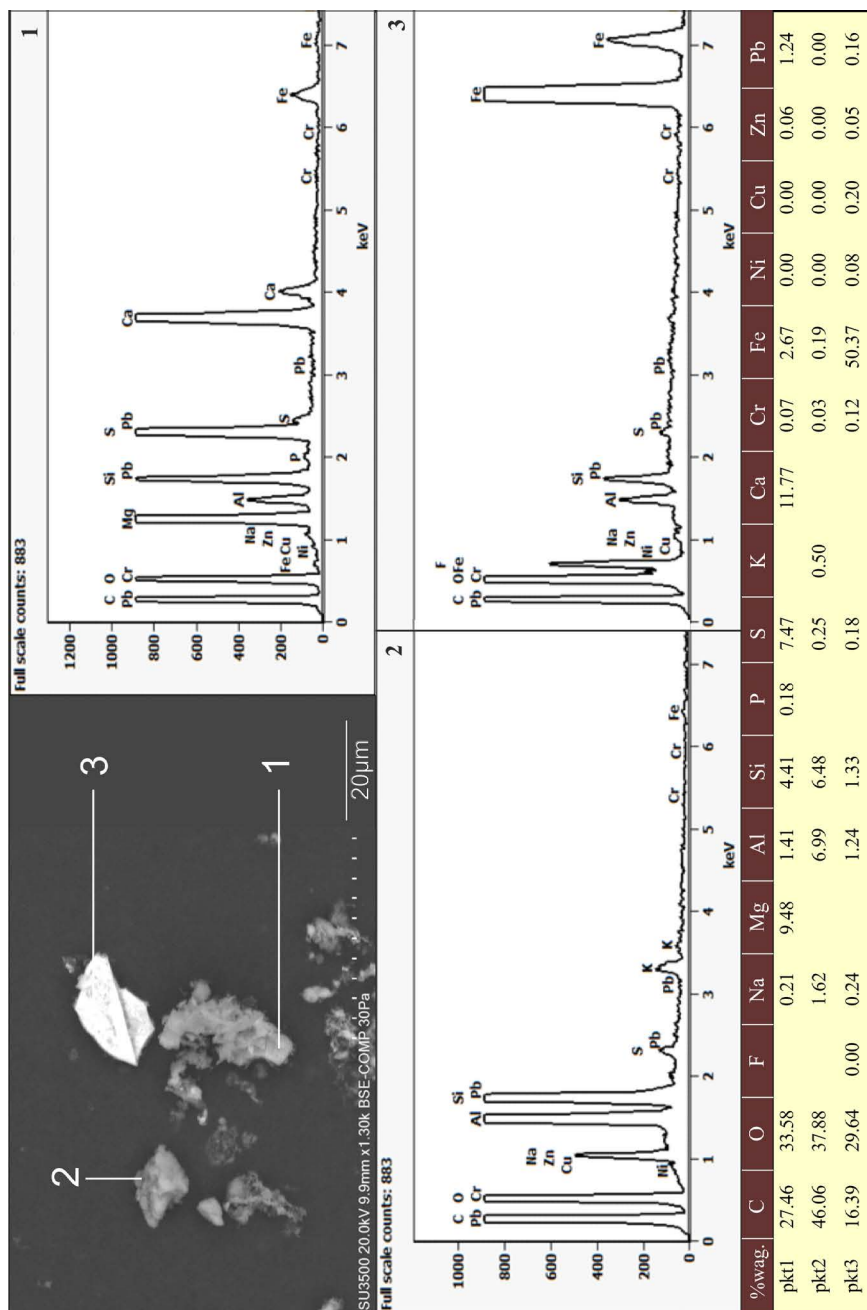


Fig. 2. SEM/EDS analysis of element content in an aluminosilicate particle with Ca and Mg oxide inclusions as well as non-fully burned macerals (1), in an aluminosilicate intergrowth with non-fully burned macerals (2), and in an iron oxide particle with non-fully burned maceral inclusions (3) in $<1.30 \times 10^3 \text{ kg/m}^3$ -density fine coal fraction ash (Piaś mine)

Rys. 2. Analiza SEM/EDS zawartości pierwiastków w cząstkach glinokrzemianu z wtrąceniami tlenków Ca i Mg, a także w niespalonych w pełni maceralach (1), w glinokrzemianach zrosniętych z nie w pełni spalonymi maceralami (2) oraz w cząstce tlenku żelaza z nie w pełni spalonymi wtrąceniami maceralów (3) w popiele miazłu węgla o gęstości $<1,30 \times 10^3 \text{ kg/m}^3$ (kopalnia Piaś)

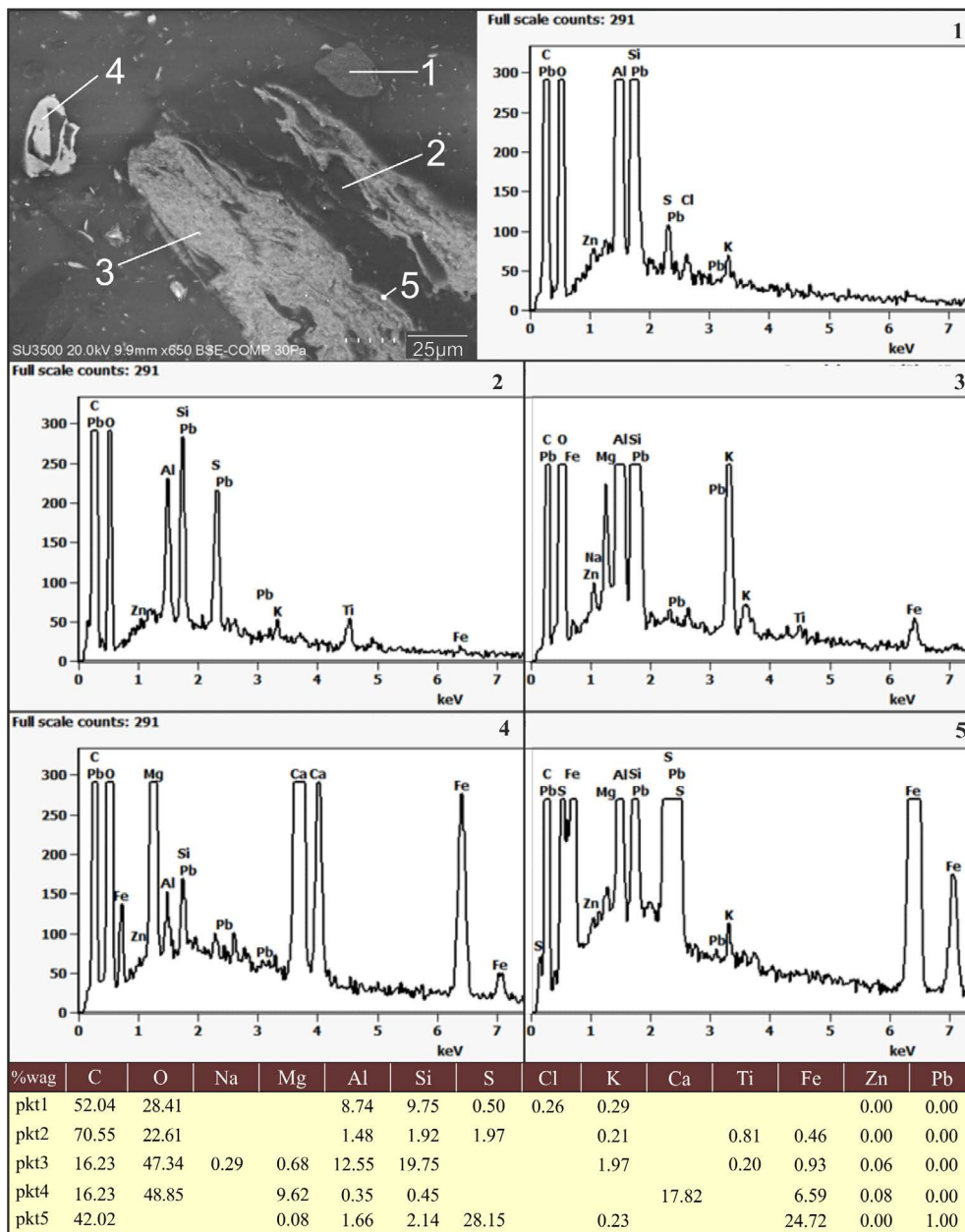


Fig. 3. SEM/EDS analysis of element content in vitrinite (1), sporinite (2), semifusinite (3), an intergrowth of Fe-dolomite with fusinite (4) and a pyrite or galena inclusion in semifusinite (5) of the $<1.30 \times 10^3 \text{ kg/m}^3$ -density fine coal fraction (Piaś mine)

Rys. 3. Analiza SEM/EDS zawartości pierwiastków w wityrynicie (1), sporynicie (2), semifuzynicie (3), zrostach Fe-dolomitu z fuzynitem (4) oraz w piryty i galenie zrośniętych z semifuzynitem (5) we frakcji miału węgla o gęstości $<1,30 \times 10^3 \text{ kg/m}^3$ (kopalnia Piaś)

of Cu, Ni and Sb in whole fine coal ash has the fraction of coal fine having the density of $1.60\text{--}2.00 \times 10^3 \text{ kg/m}^3$ (for Cu) and the fraction with the density of $<1.35 \times 10^3 \text{ kg/m}^3$ (Ni and Sb).

The assessment of the value of the correlation coefficient leads to the assumption that the content of zinc in the ash fraction with a density of $> 2.00 \times 10^3 \text{ kg/m}^3$ and copper in the ash fraction with a density of $1.60\text{--}2.00 \times 10^3 \text{ kg/m}^3$ also influences (except mineral matter) vitrite (Table 5). In turn, the content of Ni and Sb in the ash of the fraction with a density of $<1.35 \times 10^3 \text{ kg/m}^3$ is probably influenced by phosphates (applies to Sb) and clay minerals (Ni).

An unchanged large share of clay minerals in the concentration of Cr, Ni and Zn in the ash of each density fraction of whole coal fines has also been observed. In turn, along with the increase in the density of fine coal fraction, the content of Cu in the ash of this fine is probably influenced by inertinite and Fe-bearing mineral phases (in the fraction with a density of $<1.30 \times 10^3 \text{ kg/m}^3$), sulphides (1.30–1.35), liptynite (1.45–1.50), then vitrinite (1.60–2.00), and the content of Pb in fine coal ash is probably influenced by clay minerals (1.35–1.40 and 1.50–1.60) and Fe-bearing mineral phases (> 2.00) (Table 5). The above observations may be applied to the design of the petrographic and chemical composition of coal fines burned in such a way that ecotoxic trace elements may be expected in the slag, fly ash or in the absorbents purifying the gases leaving the furnace chamber. It is known that trace elements associated with organic matter of coal and sulphides, are more easily vaporized and condensed on fine particles of fly ash in the combustion chamber than when they are associated with silicates and aluminosilicates (Xu et al. 2003; Bhangare et al. 2011). In turn, knowing the distribution and forms of binding elements in the combustion residues, it will be more accurate to forecast the speed of the possible leaching of elements to soil and waters.

From the data presented in Figure 1, it also appears that the cleaning of coal fines from matter with a density $>2.00 \times 10^3 \text{ kg/m}^3$ (content Mm = 96% Tab. 1) may cause a reduction in the content of As (of about 31%), Cd (59%), Co (41%), Cr (49%), Cu (12%), Mo (52%), Ni (7%), Pb (42%), Sb (24%) and Zn (52%) and pyritic sulfur (of 49 wt %) in fine coal ash, i.e. in potential furnace wastes. This is very important information for optimizing the selection of procedures for the preparation of feed coal, for combustion, for the assessment of the suitability of coal fines for recovering or neutralizing ecotoxic elements and for assessing the probable toxicity of furnace wastes.

Conclusions

The fractions with a density of $1.30\text{--}1.35 \times 10^3 \text{ kg/m}^3$ (for Na and P) and $1.60\text{--}2.00 \times 10^3 \text{ kg/m}^3$ (Si, Al, Fe, Ca, Mg, K) have the greatest impact on sulfur content (S_t^a , S_p^a) in the coal fines from the Upper Silesian Coal Basin and: Si, Al, Fe, Ca, Mg, Na, K and P oxides in fine ash apart from mineral matter.

The content of: As, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb and Zn in the ash of coal fines fraction from the USCB with a density of $<1.30 \times 10^3 \text{ kg/m}^3$ is the largest, and in the ash fraction with a density $>2.00 \times 10^3 \text{ kg/m}^3$ is the smallest. The main carriers of elements in fine coal ash with a density of $<1.35 \times 10^3 \text{ kg/m}^3$ are grains of aluminosilicates and iron oxides resulting from the combustion of probably fusinite and semifusinite (impregnated with pyrite and aluminosilicates) and the combustion of adhesions of these macerals with dolomite, ankerite and pyrite.

There is probably a statistical tendency indicating an unchanging high share of clay minerals in concentrating Cr, Ni and Zn in the ash of each density fraction of whole coal fines. In turn, along with the increase in a density of coal fine fraction, the content of Cu in the ash of this fine is probably influenced by inertinite and Fe-bearing mineral phases (in the fraction with a density of $<1.30 \times 10^3 \text{ kg/m}^3$), sulphides (1.30–1.35), liptynite (1.45–1.50), then vitrinite (1.60–2.00), and the content of Pb in fine coal ash is probably influenced by clay minerals (1.35–1.40 and 1.50–1.60) and Fe-bearing mineral phases (> 2.00).

The fraction ash with a density $> 2.00 \times 10^3 \text{ kg/m}^3$ has the greatest impact on the content of As, Cd, Co, Cr, Mo, Pb and Zn in whole coal fines from the USCB. In turn, the largest impact on the content of Cu, Ni and Sb in the whole fine coal ash has the fraction of coal fine having a density of 1.60–2.00 $\times 10^3 \text{ kg/m}^3$ (for Cu) and fraction with a density $<1.35 \times 10^3 \text{ kg/m}^3$ (Ni and Sb).

The purification of coal fine from the matter of a density $>2.00 \times 10^3 \text{ kg/m}^3$ (mainly mineral matter) will reduce the total sulfur content in the coal fine (by 40%), and will also reduce the content of oxides Si, Al, Fe, Ca, Mg, Na, K and P (from 33% to 85%) and the reduction of As (about 31%) Cd (59%), Co (41%), Cr (49%), Cu (12%), Mo (52%), Ni (7%), Pb (42%), Sb (24%) and Zn (52%) in coal ash and thus in future furnace wastes.

Due to the relatively low mineral matter and sulfur content in the fine coal and low ash yield, the low Al, Fe, Ca, Mg, Na, K and P oxide content and high SiO_2 content in the fine coal ash, the low Roga caking index, the low fine coal propensity for furnace slagging and fouling boilers with sediments, from a technical perspective the studied fine coal constitutes a favorable power plant and household fuel and is simultaneously unsuitable for coking. Due to the above-average Pb and Zn content in the ash, USCB fine coal ash can be disruptive to the environment.

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GEOCHEMICAL CHARACTERISTICS OF FINE BITUMINOUS COAL FROM THE UPPER SILESIA COAL BASIN (POLAND) AND ITS POTENTIALLY FURNACE WASTE

Keywords

trace elements, fine coal, furnace waste, USCB

Abstract

The research involved coal from 11 coal mines in the USCB in Poland, intended for combustion in power plants and for home furnaces. It has been stated that the content of As, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb and Zn in the ash of coal fines from the USCB with a density of $<1.30 \times 10^3 \text{ kg/m}^3$ is the largest, and in the ash fraction with a density $>2.00 \times 10^3 \text{ kg/m}^3$ is the smallest. The fraction ash of coal

fine with a density $> 2.00 \times 10^3 \text{ kg/m}^3$ has the greatest impact on the content of As, Cd, Co, Cr, Mo, Pb and Zn in whole coal fines from the USCB. In turn, the largest impact on the content of Cu, Ni and Sb in whole fine coal ash has the fraction of coal fine having a density of $1.60\text{--}2.00 \times 10^3 \text{ kg/m}^3$ (for Cu) and fraction with a density $< 1.35 \times 10^3 \text{ kg/m}^3$ (Ni and Sb). The main carriers of elements in fine coal ash, thus in future furnace waste, are the grains of aluminosilicates and iron oxides resulting from the combustion of probably fusinite and semifusinite and the combustion of adhesions of these macerals with dolomite, ankerite and pyrite. The purification of fine coal from the matter with a density $> 2.00 \times 10^3 \text{ kg/m}^3$ may reduce the sulfur content (by 40%), the content of main element oxides (from 33% to 85%) and the content of ecotoxic elements (from 7% to 59%) in fine coal ash, i.e. in potential furnace wastes. Due to the small content of mineral matter, ash and sulfur in coal, small content of Al, Fe, Ca, Mg, Na, K, P oxides and high content of SiO_2 in coal ash, low value of the Rogi sinterability index, small inclination of coal fine to slag the furnaces and boiler fouling by sludge, the investigated coal was favorable for technological reasons, fuel in power plants and for home furnaces

GEOCHEMICZNA CHARAKTERYSTYKA MIAŁU WĘGLA KAMIENNEGO Z GÓRNOŚLĄSKIEGO ZAGŁĘBIA WĘGLOWEGO (POLSKA) I JEGO POTENCJALNYCH ODPADÓW PALENISKOWYCH

Słowa kluczowe

pierwiastki śladowe, miał węgla, odpady paleniskowe, GZW

Streszczenie

Badaniom poddano miał węgla z 11 kopalń w GZW, przeznaczony do spalania w zakładach energetycznych. Stwierdzono, że w popiele frakcji miału węgla z USCB o gęstości $< 1,30 \times 10^3 \text{ kg/m}^3$ jest największa, a w popiele frakcji o gęstości $> 2,00 \times 10^3 \text{ kg/m}^3$ jest najmniejsza zawartość As, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb i Zn. Popiół frakcji miału węgla o gęstości $> 2,00 \times 10^3 \text{ kg/m}^3$ ma największy wpływ na zawartość As, Cd, Co, Cr, Mo, Pb i Zn w popiele miału węgla z USCB. Z kolei największy wpływ na zawartość Cu, Ni i Sb w popiele miału węgla ma frakcja miału węgla o gęstości $1,60\text{--}2,00 \times 10^3 \text{ kg/m}^3$ (dotyczy Cu) i frakcja o gęstości $< 1,35 \times 10^3 \text{ kg/m}^3$ (Ni i Sb). Głównymi nośnikami pierwiastków w popiele miału węgla, a zatem i w przyszłych odpadach paleniskowych, są ziarna glinokrzemianów i tlenków żelaza powstałe w wyniku spalania prawdopodobnie fuzynitu i semifuzynitu oraz spalania zrostów tych macerałów z dolomitami, ankerytem i pirytem. Ze względu na ponadprzeciętną zawartość Pb i Zn, popiół miału węgla z USCB może okazać się uciążliwy dla środowiska. Oczyszczenie miału węgla z materii o gęstości $> 2.00 \times 10^3 \text{ kg/m}^3$ może zmniejszyć zawartość siarki (o 40%), zawartość tlenków głównych pierwiastków (od 33 % do 85%) i zawartości ekotoksycznych pierwiastków (od 7% do 59%) w popiele miału węgla, czyli w potencjalnych odpadach paleniskowych. Ze względu na małą zawartość substancji mineralnej, popiołu i siarki w miale węgla, małą zawartość tlenków Al, Fe, Ca, Mg, Na, K, P i dużą zawartość SiO_2 w popiele węgla, małą wartość indeksu spiekalności Rogi, małą skłonność miału węgla do żużlowania palenisk i zanieczyszczania kotłów osadem, badane miały węgla są korzystnym, ze względów technologicznych, paliwem w zakładach energetycznych i dla palenisk domowych.