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ALKALIS IN COAL AND COAL CLEANING PRODUCTS

ALKALIA W WĘGLU I PRODUCTACH JEGO WZBOGACANIA

In the coking process, the prevailing part of the alkalis contained in the coal charge goes to coke. The content of alkalis in coal (and also in coke) is determined mainly by the content of two elements: sodium and potasium. The presence of these elements in coal is connected with their occurrence in the mineral matter and moisture of coal. In the mineral matter and moisture of the coals used for the coke production determinable the content of sodium is 26.6 up to 62. per cent, whereas that of potassium is 37.1 up to 73.4 per cent of the total content of alkalis. Major carriers of alkalis are clay minerals. Occasionally alkalis are found in micas and feldspars. The fraction of alkalis contained in the moisture of the coal used for the production of coke in the total amount of alkalis contained there is 17.8 up to 62.0 per cent. The presence of sodium and potassium in the coal moisture is strictly connected with the presence of the chloride ions. The analysis of the water drained during process of the water-extracting from the flotoconcentrate showed that the Na to K mass ratio in the coal moisture is 20:1. Increased amount of the alkalis in the coal blends results in increased content of the alkalis in coke. This leads to the increase of the reactivity (CRI index), and to the decrease of strength (CSR index) determined with the Nippon Steel Co. method.

Keywords: coal, cleaning process, alkalies, mineral matter

W procesie koksowania przeważająca część zawartych we wsadzie węglowym alkaliów przechodzi do koksu. Zawartość alkaliów w węglu, a co za tym idzie i w koksie determinowana jest głównie zawartością dwóch pierwiastków: sodu i potasu. Obecność tych pierwiastków w węglu wiąże się z występowaniem ich w substancji mineralnej i wilgoci węgla. W substancji mineralnej oraz wilgoci węgli stosowanych do produkcji koksu, oznaczona zawartość sodu wynosi od 26.6 do 62.9%, a zawartość potasu od 37.1 do 73.4% alkaliów ogółem. Głównymi nośnikami alkaliów w substancji mineralnej są minerały ilaste, sporadycznie też miki oraz skalenie. Udział alkaliów zawartych w wilgoci węgli stosowanych do produkcji koksu w ogólnej ilości zawartych w nim alkaliów wynosi dla badanych węgli od 17.8 do 62.0%. Obecność sodu i potasu w wilgoci węgla związana jest wyłącznie z obecnością w niej jonów chlorkowych. Wyniki analizy wody odprowadzanej z procesu wirowania flotokoncentratu wskazują, że stosunek masowy Na do

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K w wilgoci węgla wynosi 20:1. Wzrost zawartość wilgoci w koksie będący wynikiem ich zwiększonej ilości w mieszance węglowej prowadzi do wzrostu reaktywności (wskaźnik CRI) oraz spadku wytrzymałości (wskaźnik CSR) oznaczonych metoda Nippon Steel Co.

Słowa kluczowe: węgiel, proces wzbogacania, substancja mineralna

1. Introduction

In the modern blast furnace technology, the cost of pig iron production as well as the coke consumption can be reduced due to the use of alternative fuels. In recent years, in the world iron and steel industry, the majority of production of pig iron is concentrated in the blast furnaces of 3000-5000 m^3 capacity where the Powder Coal Injection (PCI) technology is applied. PCI in quantity of approximately 200 kg/t of the pig iron reduces the coke consumption down to 300 kg/t of pig iron (Rooney et al., 1987). Reduction in coke consumption is related to the increase of its quality requirements (Diez et al., 2001; Karcz & Strugała, 2008) These requirements focuses not only on conventionally used parameters, such as reactivity, mechanical properties, ash content, moisture, but also content of sulphur, phosphorus, chlorine, and alkali compounds. These compounds worsen the pig iron quality, cause certain disturbances in the blast furnace operation decreasing its productivity and increase coke consumption.

Alkali compounds participate in the so-called circulation of alkalis in the blast furnace and are responsible for their accumulation in the bottom part of the furnace. This leads to creation of slug crusts, the removal of which leads to clogging and burning of the blowing nozzles (Ronney et al., 1987; Buzek & Dankmayer-Łączny, 1999).

In the blast furnace process the main source of alkali compounds is coke, which is responsible for the introduction of approx. 40 per cent of all alkalis circulating in the blast furnace. According to the current requirements, the content of the alkali compounds in coke, considered as the sum of Na_2O+K_2O, should not exceed 0.2-0.3 per cent (Wasilewski et al., 2000). The high level of the content of the alkali compounds in coke increases its reactivity in relation to CO_2 determined by the coke reactivity index (CRI) and decreases coke strength (determined by the coke strength after reaction index (CSR), according to the Nippon Steel Co method

The alkalis occurring in coke can by divided into three groups (Groβpietsch, 2000; Strugała & Bytnar, 2004):

- alkali compounds includedin the mineral matter of the coals used for coke production,
- chlorides of the alkali metals dissolved in watercontained in coals used for coke production,
- salts of alkali metals introduced to coke at wet quenching or utilised by adding them directly to the coked mixture

In this paper we present the quantitative and qualitative characteristics of alkali compounds contained in the selected coals from the Upper Silesia Coal Basin and in products obtained after processing these coals.

2. The forms of occurrence of alkali compounds and chlorine in coal

The coal mineral matter is a mixture of various chemical compounds. Only small part of these compoundsis bounded or forms a homogenous mixture with coal organic matter. The major part of the mineral matter in coal forms isolated clusters of different sizes.

The analysis of the mineral matter composition for Polish and foreign coals shows that the major components containing alkali elements are: three-dimensional aluminosilicates, layered aluminosilicates, sulphates, halogens and trace and dispersed elements (Table 1) (Kuhl, 1980; Strugała, 1998).

TABLE 1

Examples of minerals containing alkali compounds occurring in Polish hard coals (Strugała, 1998)

Name and description		Chemical formula
Potassium feldspars:		
orthoclase (medium temperature variety)		$K[AlSi_3O_8]$
microcline (low-temperature variety)		$K[AlSi_3O_8]$
Sodium & Potassium feldspars (plagioclases):		
albite		$Na[AlSi_3O_8]$
mixed plagioclases (albite + anorthite)		-
anorthite		$Ca[Al_2Si_2O_8]$
Micas:		
dioctahedral structure:		
ordinary micas:	muscovite	$KAl_2[(OH)_2/AlSi_3O_{10}]$
	hydromuscovite	$(K,H_3O)Al_2[(OH)_2/AlSi_3O_{10}]$
hydromicas	illite	$(K,H_3O)Al_2[(OH)_2/AlSi_3O_{10}]$
trioctahedral structure:		
ordinary micas:	biotite	$K(Mg,Fe, Mn)3(OH,F)_2 /AlSi_3O_{10}]$
Montmorillonites:		
dioctahedral structure:		
montmorillonites	montmorillonite	$(Al_{1.67}Mg_{0.33})[(OH)_2[Si_4O_{10}]Na_{0.33}(H_2O)_4$
	beidelite	$Al_2[(OH)_2 [Al_{0.5}Si_{3.5}O_{10}]\cdot (Ca, Na)_{0.5}(H_2O)_4$
Anhydrous chlorides:		
halite		NaCl
sylvine		KCl
Anhydrous sulphates:		
alunite		$KAl_3(OH)_6/[(SO_4)_2]$
natroalunite		$NaAl_3(OH)_6/[(SO_4)_2]$
jarosite		$KFel_3(OH)_6/[(SO_4)_2]$
natrojarosite		$NaFe_3(OH)_6/[(SO_4)_2]$

The properties of these components including behaviour in high temperatures, as well as thermal alternations they undergo have been discussed inter alia (Hodges et al., 1983; Strugała & Bytnar, 2004). Because the temperature of the decomposition of alkalis contained in the mineral coal matter is higher than the temperature of coking, these compounds go almost entirely to coke. The presence of alkali compounds in coal is closely connected with the presence of chlorine in it. Chlorine may occur in coal in three forms: as chloride ions in the coal moisture, as inorganic chlorides, and as organic combinations. However, the presence of the latter ones is questioned by many authors (Hodges et al., 1983; Manzoori & Agarwal, 1992).

3. Methods of investigation

Investigation was conducted on samples of raw hard coals from the mines labeled as: A, B, C, D located in the Upper Silesia Coal Basin. Also products obtained from raw hard coal cleaning were investigated.That included concentrates, by-products as middlings and tailings, and wastes. Coal types were selected by taking into account the differentiated content of the alkalis in these coals and the variety of the technological solutions of the cleaning processes used.

All mines have the full technology of coal cleaning and comprehensively carry out the processing of the entire coal output (Nycz & Zieleźny, 2004; Kowalczyk & Strzelec, 2004)

As far as the B, C and D mines are concerned the cleaning process includes three operations:

– cleaning in suspension washer (for grain class > 20 mm),
– cleaning in jig washer (for grain class 20-0.5 mm),
– cleaning by floatation (for grain class < 0.5 mm).

In case of the A mine, the cleaning process goes without producing intermediate products and it is limited to two processes:

– cleaning in jig washer (for grain class 70-0.5 mm),
– cleaning by floatation (for grain class < 0.5 mm).

The processing plants in these mines have closed water-slurry circuits and a separate post-flotation waste management system. In the D mine, the spinning water-extraction systems of sediment-flotation type are used for the dewatering of the flotoconcentrate. In addition, the process of coal thermal drying is used there whereas in other coal mines the coal thermal drying process is only used for the dewatering of the flotoconcentrate (Kowalczyk & Strzelec, 2004; Nycz & Zieleźny, 2004)

The technical analysis for individual samples has been performed in order to determine the moisture content (PN-80-G-04511), ash content (PN-80-G-04512 and PN-G-04560:1998 with TGA-501 automatic analyser, by Leco) and the content of volatile matter (PN-G-04516:1998). Furthermore, the chemical analysis to determine chlorine according to PN-G-04534:1999 and PN-ISO 587:2000as well as sodium and potassium dioxide in the ash obtained after burning these samples according to PN-G-04528-10:1998 (with AFP-100 flame photometer, by BIOTECH) has been carried out.

The selected samples have also undergone investigation with the use of X-ray diffractometry using the Debye-Sherrer powder method as well as microscope examination. X-ray photographs have been recorded with the Philips roentgen diffractometer of X'Pert PW 3020 type, while microscope investigations have been performed in the passing light with the POLMI A polarisation microscope and additionally with an OLYMPUS microscope. Preparations were made in the form of polished, thin plates made of the chipping material of the investigated samples after hardening them with araldite.

4. Results of investigations

The results of the investigations of the distribution of alkalis in the cleaning processes of the coals originated from selected mines have been discussed with reference to the individual products of this process Flow charts of the cleaning of the coals of various types as well as the results of the determination of Na, K, and Cl content in both raw coals and various products coming from coal cleaning processes were presented in figures 1-4.

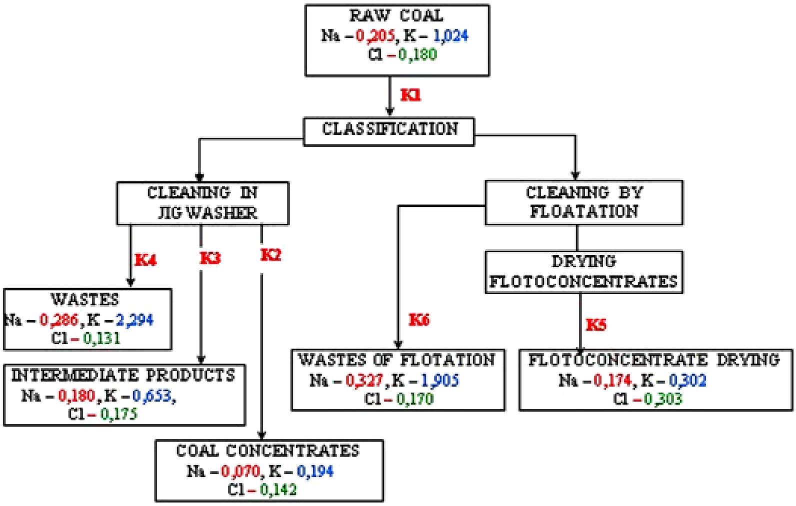

Fig. 1. The chart of coal cleaning process and the distribution of contaminants in the A mine

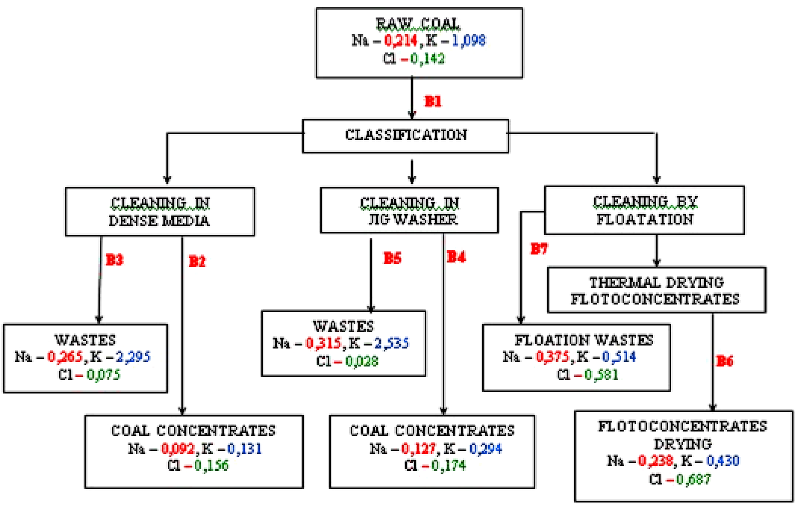

Fig. 2. The chart of coal cleaning process and the distribution of contaminants in B mine

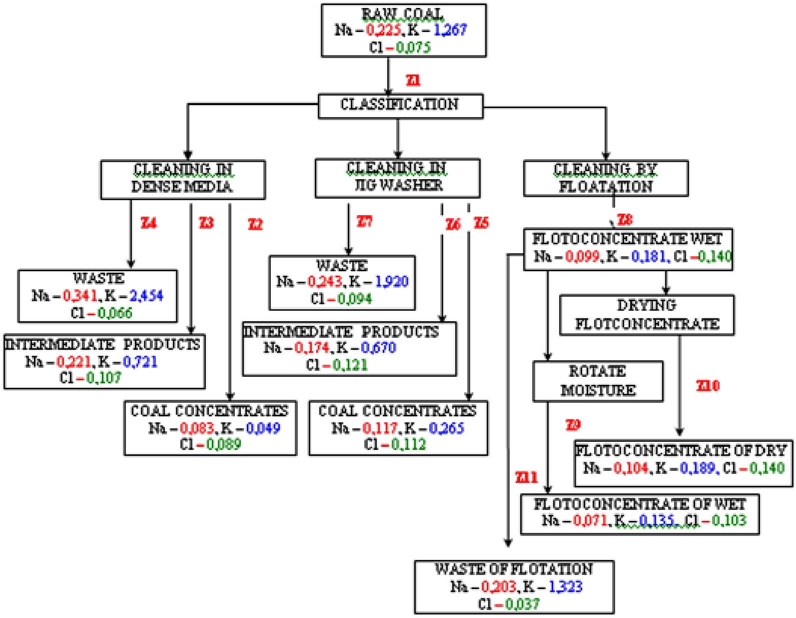

Fig. 3. The chart of coal cleaning process and the distribution of contaminants in the C mine

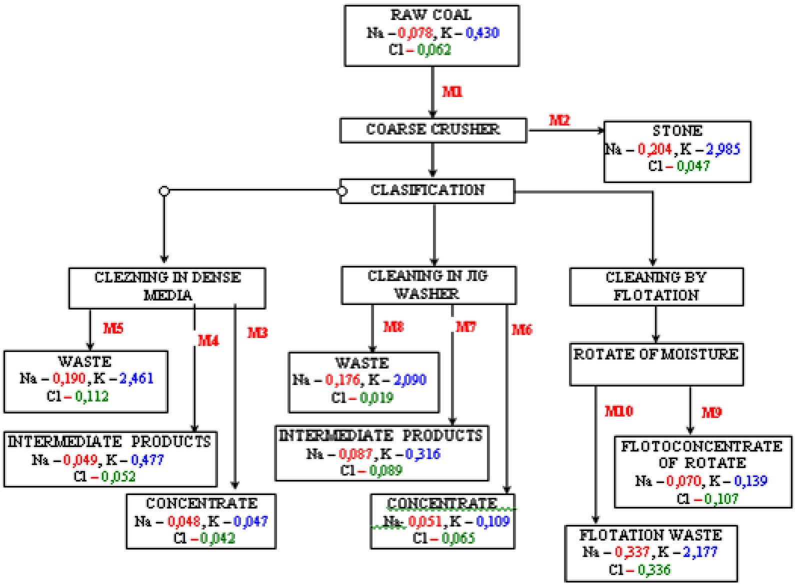

Fig. 4. The chart of the coal cleaning process and the distribution of contaminants in the D mine

4.1. Raw coals

The content of alkalis (the sum of Na and K) in raw coals amounted to 0.508% for the coal from the D mine, and up to 1.492 for the coal from the C mine. The ratio of Na to K was in general constant for all of these coals and it varied from 0.18 to 0.20 (Table 2).

TABLE 2

Specification of the selected results for raw coals

Raw coals	Na content [%]	K content [%]	Alkalis generally content [%]	Alkalis in moisture content [5]	Alkalis in mineral matter content [%]	Cl content [%]	Ash content [%]	Alkalis in ash content [%]
Mine A	0.205	1.024	1.229	0.109	1.120	0.180	38.4	3.201
Mine B	0.214	1.098	1.312	0.086	1.226	0.142	40.6	3.232
Mine C	0.225	1.267	1.492	0.045	1.447	0.075	44.0	3.391
Mine D	0.078	0.430	0.508	0.038	0.470	0.062	18.8	2.702

The distribution of alkalis in individual coals was in generalconstant; about 90 per cent of the alkalis were connected with the mineral matter and merely 10 per cent – with the moisture contained in the coal. Therefore, the content of alkalis in coals is determined by the content of the mineral matter.

In Fig. 5, Corelation between the content of alkalis in ash and that of ash in coal is presented.

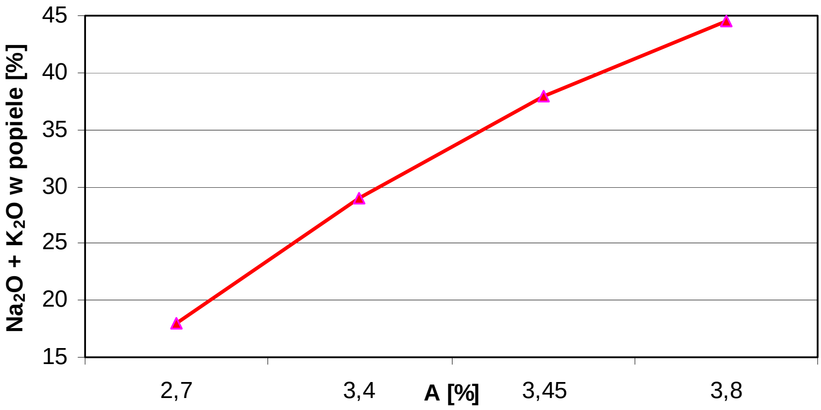

Fig. 5. Corelation between the content of alkalis in ash and that of the ash in coal

The results of the phase and mineralogical analysis show a great similarity for all the coals and confirm that clay mineral (illites and minerals of the mixed-package type as illite/smectite) and the micas of secondary occurrence (muscovite and hydromuscovite) are basic carriers of alkalis. In addition, for the coal from the B mine the admixtures of feldspars have been identified (Table 3).

TABLE 3

Specification of the minerals identified in the samples raw coals and the products of their cleaning

		Minerals contains alkalis						Residual minerals						
		Minerals of mixed-package as illite/smectite	Illite – 2m1 $(KH_3O)Al_2Si_3AlO_{10}(OH)_2$	Albite (low) $NaAlSi_3O_8$	Halit e NaCl	Microcline $KAlSi_3O_8$	Muskovite $KAl_2(OH, F)_2AlSi_3O_{10}$	Quartz SiO_2	Kaolinite – 1md $Al_2Si_2O_5(OH)_4$	Clinochlore –1a (ferro) $(MgFeAl)(SiAl)_{4}O10(OH)_8$	Dolomite (ferroan) $Ca(Mg_{0.67}Fe_{0.33})(CO_3)_2$	Syderite $FeCO_3$	Pyrite FeS_2	Calcit e $CaCo_3$
Mine A	raw coal	•	•				•	•	•	•	•	•	•	
	intermediate products of the jig washer	•	•				•	•	•		•	•	•	
	wastes of jig washer	•	•	•			•	•	•	•	•	•	•	
	wastes of flotation	•	•				•	•	•	•	•	•		•
	coal concentrates – mix	•	•		•			•	•			•	•	•
Mine B	raw coal	•	•	•			•	•	•	•	•	•	•	
	waste of the dense liquids	•	•	•			•	•	•	•	•	•		
	wastes of jig washer	•	•				•	•	•	•	•	•	•	
	wastes of flotation	•	•		•		•	•	•	•	•	•	•	
	coal concentrates – mix	•	•		•			•	•	•	•	•	•	
Mine C	raw coal	•	•				•	•	•		•	•		
	intermediate products of the dense liquids	•	•					•	•		•	•	•	
	waste of the dense liquids	•	•	•			•	•	•	•	•	•	•	
	intermediate products of the jig washer	•	•	•			•	•	•		•	•	•	
	wastes of jig washer	•	•	•			•	•	•		•	•	•	
	wastes of flotation	•	•	•			•	•	•	•	•	•		•
	coal concentrates – mix	•			•			•	•		•	•	•	
Mine D	raw coal	•	•				•	•	•	•		•		
	stone of crushers	•	•	•		•	•	•	•	•		•		
	intermediate products of the dense liquids	•	•	•			•	•	•		•	•		•
	waste of the dense liquids	•	•	•	•	•	•	•	•	•				
	intermediate products of the jig washer	•	•				•	•	•	•	•	•	•	•
	wastes of jig washer	•	•	•			•	•	•	•	•	•	•	
	wastes of flotation	•	•	•			•	•	•	•	•	•	•	•
	coal concentrates – mix	•			•			•	•		•	•		•

4.2. Coal concentrates

The data presented in Table 4 demonstrate that, in general, the concentrates originated from the JasMos mine coal are featured with the lowest content of alkalis. This is connected with low content of alkalis in raw coal and the technology of the dewatering (hydro-extracting) of the flotoconcentrate that is used for this type of coal and that allows to remove a part of the moisture along with alkalis from it.

When comparing the concentrates produced in the different stages of coal cleaning it can be found that the most considerable decrease in the content of alkalis in relation to that in the raw coal occurs when the cleaning process is carried out in dense liquids and smaller – when the flotation cleaning process is applied. There is an exemption for the coal from the C mine for which the lowest level of the reduction of alkalis has been observed for the concentrate obtained when jig washers were used.

In comparison to raw coals, the fraction of sodium in total alkalis increases significantly and this applies in particular to both flotoconcentrates and concentrates originated from the cleaning process in dense media. This results from the higher level of the reduction of the potassium than the sodium content in different coal cleaning processes. What is interesting, in case of certain flotoconcentrates an increase of sodium content was observed in comparison with its content in raw coal (coals from the B and C mines).

TABLE 4

Specification of the selected results for coal concentrates

Coal concentrates			**Na content [%]**	**K content [%]**	**Alkalis generally content [%]**	**Alkalis in moisture content [5]**	**Alkalis in mineral matter content [%]**	**Cl content [%]**	**Ash content [%]**	**Alkalis in ash content [%]**
Mine A	jig washer		0.070	0.194	0.263	0.086	0.177	0.142	9.0	2.922
	flotation machines		0.174	0.302	0.477	0.183	0.294	0.303	11.3	4.221
Mine B	dense liquids		0.092	0.131	0.223	0.095	0.128	0.156	8.1	2.753
	jig washer		0.127	0.294	0.421	0.105	0.316	0.174	12.6	3.341
	flotation machines		0.238	0.430	0.668	0.414	0.254	0.687	9.8	6.816
Mine C	dense liquids		0.083	0.049	0.132	0.054	0.078	0.089	4.6	2.870
	jig washer		0.117	0.265	0.383	0.068	0.315	0.112	10.9	3.514
	flotation machines	flot of wet	0.099	0.181	0.280	0.085	0.195	0.140	7.8	3.590
		flot of rotate	0.071	0.135	0.206	0.062	0.144	0.103	6.5	3.169
		flot of dry	0.104	0.189	0.293	0.085	0.208	0.140	7.8	3.756
Mine D	dense liquids		0.048	0.047	0.095	0.025	0.070	0.042	5.4	1.759
	jig washer		0.051	0.109	0.160	0.040	0.120	0.065	7.4	2.162
	flotation machines		0.070	0.139	0.209	0.065	0.144	0.107	9.1	2.297

The calculated values of content of alkalis in the moisture of concentrates and their mineral matter (Table 4) differ considerably from those determined for raw coals. The content of the alkalis in the mineral matter dropped significantly, whereas the content of the alkalis present in the coal moisture actually has not changed. It resulted in the significant increase of the fraction of the latter in the total amount of the alkalis in coal.

The results of the phase and mineralogical analysis, which covered the combined coal concentrates of each coal considered, show that the major carriers of alkalis are clay minerals (in the form of mixed-package minerals as illite/smectite) and rock-salt (halite). In the coal concentrates produced from C and D coals, mica flakes have also sporadically been observed (Table 3).

4.3. Intermediate products of the coal cleaning process

The results presented in Table 5 show that the content of alkalis in the coals from A and C mines is significantly lower than in raw coals. This is due to the reduced potassium content in these products only. As for the by-products of the coal from the D mine, the content of both sodium and potassium is close to that in raw coal.

When analysing the data presented in Table 5 concerning the content of the alkalis in the moisture of the cleaning by-products and the mineral matter contained there (Table 3) it can be stated that values are similar to those determined for raw coals.

The results of the phase and mineralogical analysis of the by-product originated from the cleaning process in dense media and jig washers (Table 3) only indicate of minor occurrence as possible carriers of alkalis the signs of the presence of clay minerals (illite and the minerals of the mixed-package type as illite/smectite), and for coals from the D mine - also the micas.. Moreover, in the case of coals from the C and D mines, micas and admixtures of feldspars (albite) in the by-products from jig washers can be found.

TABLE 5

Specification of the selected results for intermediate products obtained from the coal cleaning process

Intermediate products		Na content [%]	K content [%]	Alkalis generally content [%]	Alkalis in moisture content [5]	Alkalis in mineral matter content [%]	Cl content [%]	Ash content [%]	Alkalis in ash content [%]
Mine A	jig washer	0.180	0.653	0.833	0.105	0.728	0.175	24.5	-
Mine C	dense liquids	0.221	0.721	0.942	0.065	0.877	0.107	29.1	-
	jig washer	0.174	0.670	0.845	0.073	0.772	0.121	25.3	-
Mine D	dense liquids	0.049	0.477	0.526	0.032	0.494	0.052	38.9	-
	jig washer	0.087	0.316	0.403	0.054	0.349	0.089	17.5	-

4.4. Wastes from the coal cleaning process

The data presented in Table 6 show that the dominating alkali component in wastes is potassium.

It should be emphasised that the close content of the alkalis in cleaning wastes when the process is run in dense media in the all mines considered, and the rather high differences of the alkali content for these mines as it comes for post-flotation wastes. The content of alkalis in the moisture of the wastes remaining after the coal cleaning process and the alkalis contained in their mineral matter show that the latter is the dominating type of alkalis.

The results of the phase and mineralogical analysis of the wastes coming from the cleaning process in dense media and from crushing mills (for the coal from the D mine) show that the major carriers of alkalis are clay minerals, and in the second place micas (muscovite, hydromuscovite), feldspars, as well as rock-salt (halite). For the wastes from jig washers and flotation machines, the major carriers of alkalis are clay minerals and micas. Summarising, just as in the case of for raw coals, similarity of the results of the investigation of the individual wastes from the analysed mines should be emphasised.

TABLE 6

Specification of the selected results for the wastes produced in the coal cleaning process

Waste products		Na content [%]	K content [%]	Alkalis generally content [%]	Alkalis in moisture content [5]	Alkalis in mineral matter content [%]	Cl content [%]	Ash content [%]	Alkalis in ash content [%]
Mine A	jig washer	0.286	2.294	2.508	0.079	2.501	0.131	82.0	-
	flotation machines	0.327	1.905	2.232	0.103	2.129	0.170	63.9	-
Mine B	dense liquids	0.265	2.295	2.559	0.045	2.514	0.075	86.9	-
	jig washer	0.315	2.535	2.851	0.103	2.129	0.028	80.2	-
	flotation machines	0.375	0.514	0.890	0.350	0.540	0.581	24.7	-
Mine C	dense liquids	0.341	2.454	2.795	0.040	2.755	0.066	81.9	-
	jig washer	0.243	1.920	2.163	0.057	2.106	0.094	66.9	-
	flotoconcentrate of wet	0.203	1.323	1.526	0.023	1.503	0.037	57.1	-
Mine D	crusher	0,204	2,985	3,189	0,028	3,161	0,047	94,9	-
	dense liquids	0,190	2,461	2,651	0,068	2,538	0,112	88,2	-
	jig washer	0,176	2,090	2,266	0,012	2,254	0,019	82,0	-
	flotation machines	0,337	2,177	2,514	0,203	2,311	0,336	72,0	-

5. Conclusions

In the coking process, the prevailing part of the alkalis contained in the coal charge moves to coke. Because of that, the issue of the acting of alkalis at the coal mechanical processing is very important for both the quality of the produced coke and the course of the blast furnace process.

The content of alkalis in coal (and also in coke) is determined mainly by the content of two elements: sodium and potassium. The presence of these elements in coal is connected with their occurrence in the mineral matter and coal moisture. In the mineral matter and moisture of the coals used for the coke production the content of sodium is 26.6 up to 62.9 per cent, whereas that of potassium is 37.1 up to 73.4 per cent of the total content of alkalis. The phase and mineralogical analysis results have unambiguously shown that the major carriers of alkalis are clay minerals (in the form of mixed-package minerals as illite/smectite, and sometimes also illite). Occasionally, mica flakes (muscovite, hydromuscovite) and feldspars have also been found.

The fraction of alkalis contained in the moisture of the coals used for the production of coke in the total amount of alkalis contained is 17.8 up to 62.0 per cent. A lower fraction has been found in the concentrates coming from the coal cleaning in dense media and jig washer. In case of flotoconcentrates, these fractions are definitely higher.

The presence of sodium and potassium in the coal moisture is strictly with the presence of the chloride ions in it. The presence of chlorine in the moisture is connected as much as in 90 per cent with the presence of sodium and potassium ions. The rest of chlorine is associated with calcium, magnesium, ammonia and other ions. The analysis of the water drained at the process of the water-extracting from the flotoconcentrate shows that the Na to K mass ratio in the coal moisture is 20:1.

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