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The relationship between CRI and CSR indices and other quality parameters of coking coal from the Pniówek deposit (SW part of the USCB, Poland)

Introduction

At the end of the 1980s, the CRI reactivity index and the CSR strength after reaction index were introduced by Nippon Steel Co (NSC). These indices now serve as the basic criterion in coal quality determination. In all coal classifications, both domestic and international, the quality parameters of coking coal also give consideration to the classic coking indices such as the Roga caking power (RI), the free swelling index (FSI) and the dilatometric indices: dilatation (b) and contraction (a). In the production of high-quality coke, the identification of other parameters is also required, including, among others, the content of chlorine (Cl) and phosphorus (P).

These pursuits, based only on the knowledge of values of coking parameters, do not always meet the expectations relating to the predictions of the quality of coke produced from

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coking coal. The conducted studies usually encompass the acquisition of data regarding the quality of the coke (the product) and the quality of the coking coal which serves the production of coke. This process may be considered as the development of own databases regarding the quality of the product (coal/coke).

Quoting (Coin and Broome 1997): "Considering the amount of money spent on coal each year by coke producers, there appears to be very little spent on sound data analysis studies to try and optimize production and quality." Recently, however, an increasing engagement in the study of relations between CRI, CSR and the remaining coal quality parameters has been observed. Moreover, the parameters include more than the typical coking indicators (Chiu et al. 1985; Li et al. 2015; Sakurovs et al. 2012). A significant role is also played by the subject of quality prediction in view of the coke itself as well as the coking blends (Álvarez et al. 2007; Díez et al. 2002; Gupta et al. 2012; MacPhee et al. 2013; Menendez et al. 1999; Ryan and Price 1992; Tiwari et al. 2013; Wang et al. 2016). The participation of Polish researchers may also be noted in the studies related to the topic and domestic experiments pertaining to the subject matter have not neglected the Polish coking coal deposits (Komorek et al. 2010; Koszorek et al. 2009; Krzesińska et al. 2009; Morga et al. 2015; Probierz and Marcisz 2010b; Pusz and Buszko 2012; Pusz et al. 2009). An attempt to present the global comprehensive results included in the professional literature/publications was made in (North et al. 2018a, b).

Several notes might be made on the Polish publications regarding CRI and CSR. The focus on the correlations of these parameters to the vitrinite reflectance and the content of individual maceral groups (petrographic composition) may be noted, among others, in the papers (Probierz and Marcisz 2015; Pusz and Buszko 2012; Pusz et al. 2009).

The study were provided in the Jastrzębie area (SW part of the Upper Silesian Coal Basin), where coking coal deposits exploited by Jastrzębska Spółka Węglowa SA – the largest business entity preoccupied with the exploitation of such coal in Poland – are located (Fig. 1). Among all the deposits exploited by JSW SA the "Pniówek" deposit is of fundamental significance to the Polish reserves of coking coal. It is, however, the least mentioned as the subject matter of conducted studies regarding CRI and CSR, which may result from the fact that the deposit is characterized by considerable variability of quality and coal rank and the CRI and CSR indices have only been registered there since August of 2009. The knowledge of the geological structure of the deposit in the Jastrzębie region should provide significant data allowing for a more precise prediction of the quality of coke produced from the coal. The above, of course, may pertain to any coking coal deposit in the world.

The problem of the variability of the coal quality in the "Pniówek" deposit, as well as its dependence on the structure and the petrographic composition of seams, has been the subject of numerous papers, including (Probierz and Marcisz 2010a).

The study area is characterized by a high variability of coal quality and a unique coexistence of anthracite among coking coal at an area of ~0.24 km^2. The anthracite resources of ~1 million tons are surrounded by inertinite-rich coking coal characterized by decreased

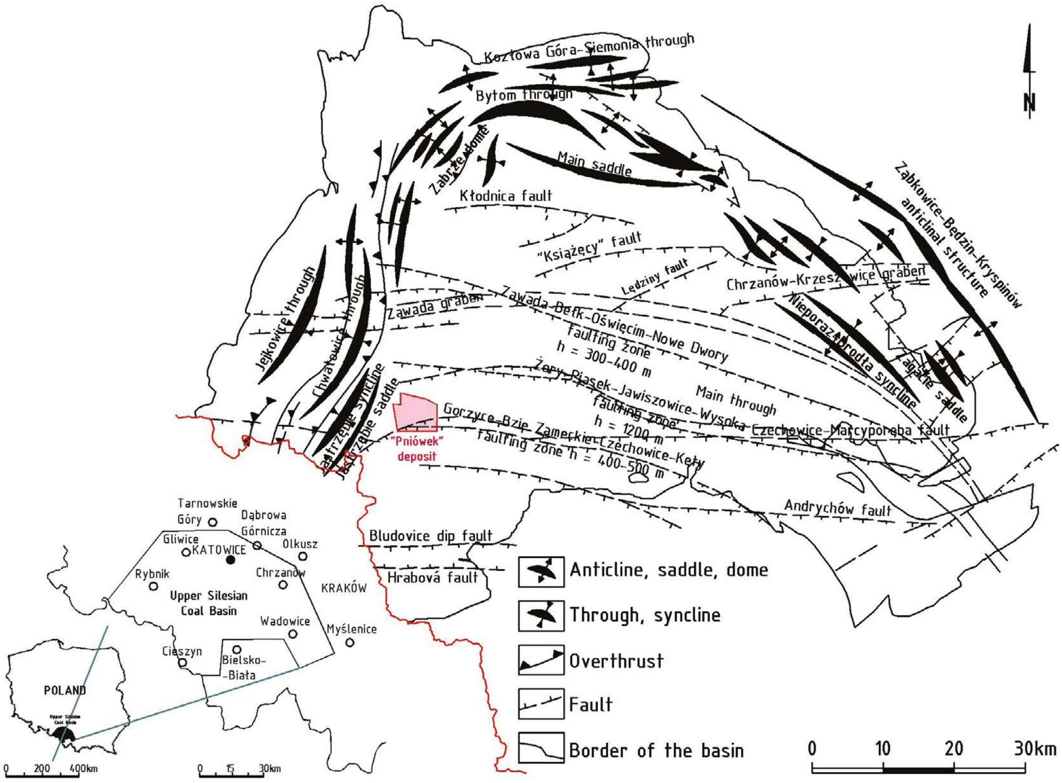

Fig. 1. The studied area against the background of Upper Silesian Coal Basin tectonics

Rys. 1. Obszar badań na tle tektoniki Górnośląskiego Zagłębia Węglowego

coking properties (Fig. 2). Their identification was only possible due to detailed petrographic as well as chemical and technological tests. The identification of such a co-occurrence of anthracite and coking coal in underground conditions was difficult. Only the change in the quality of the mined material, noted by the processing plant, caused detailed geological and petrographic studies to be initiated. Thermal metamorphism was identified as the cause of the coexistence of anthracite and coking coal (Gabzdyl and Probierz 1987).

The subject of dependence of the variability of coal quality, its coalification level and the petrographic composition from the possibility of thermal metamorphism processes was also raised in (Komorek et al. 2010).

The variability of the CRI and CSR indices has also been identified in the deposit area and the vertical and horizontal variations of these indices have been analyzed (Probierz and Marcisz 2015).

Considering the above as the substantiation of taking up the research described in this paper, an analysis of the correlation between the values of the CRI and CSR indices and the values of the remaining coal quality parameters determined in the channel samples acquired in the workings, was conducted. The presented results should facilitate the prediction of the quality of coking coal characterized by parameters desired by the coking industry.

Considering the paper of (North et al. 2018b) as a list of world experiences in the prediction of coal quality, it seems to be a reasonable comparison of the variability of CRI and CSR parameters of coal in the "Pniówek" deposit, an elementary basis of Polish coking coal against the background of results placed in this work. One of the purposes of this work was also making of preliminary statistical analysis enabling presentation of position of coal from the "Pniówek" deposit against the background of world coal deposits.

1. The location and the geological characteristics of the study area

The "Pniówek" deposit is located in the SW part of the Upper Silesian Coal Basin in the disjunctive tectonics zone (Fig. 1).

The deposit includes Upper Carboniferous sediments: the Paralic Series (with Poręba Beds), the Upper Silesian Sandstone Series (with Zabrze Beds and Ruda Beds) and Mudstone Series (with Załęże Beds). The overburden is represented by Miocene and Quaternary formations, which are characterized by diversified lithological characteristics and a diversified depth.

The "Pniówek" deposit is located within the borders of the Zofiówka monocline with a slight but variable direction of dip from 5 to 20° in the SEE direction. It is also characterized by intensive fault tectonics. The deposit is situated between two fault zones with a regional range: Żory–Piasek–Jawiszowice–Wysoka fault zone to the north and the Gorzyce––Bzie Zameckie–Czechowice–Kęty fault zone to the south. The deposit is characterized by complex tectonics, including numerous faults forming areas of tectonic displacement and high variation in thickness of the entire seam as well as the presence of dirt bands (numerous barren rock interlayers).

2. Data and methods

Data acquired within the "Smart coke plant meeting the requirements of the best available techniques" project, conducted in the years 2008–2014 by the Institute for Chemical Processing of Coal in Zabrze, in which the authors participated, was used in the research. The data encompass the results of analyses of the quality of coal from 25 channel samples acquired in the workings of the "Pniówek" coal mine based on directions of the Polish Standard determining the collection of such samples in underground mining workings.

The results encompass the values of 36 coal quality parameters. Values of coal parameters have been assigned according to actual domestic and international standards regulating the determination of coal quality parameters. Some values of parameters have been assigned

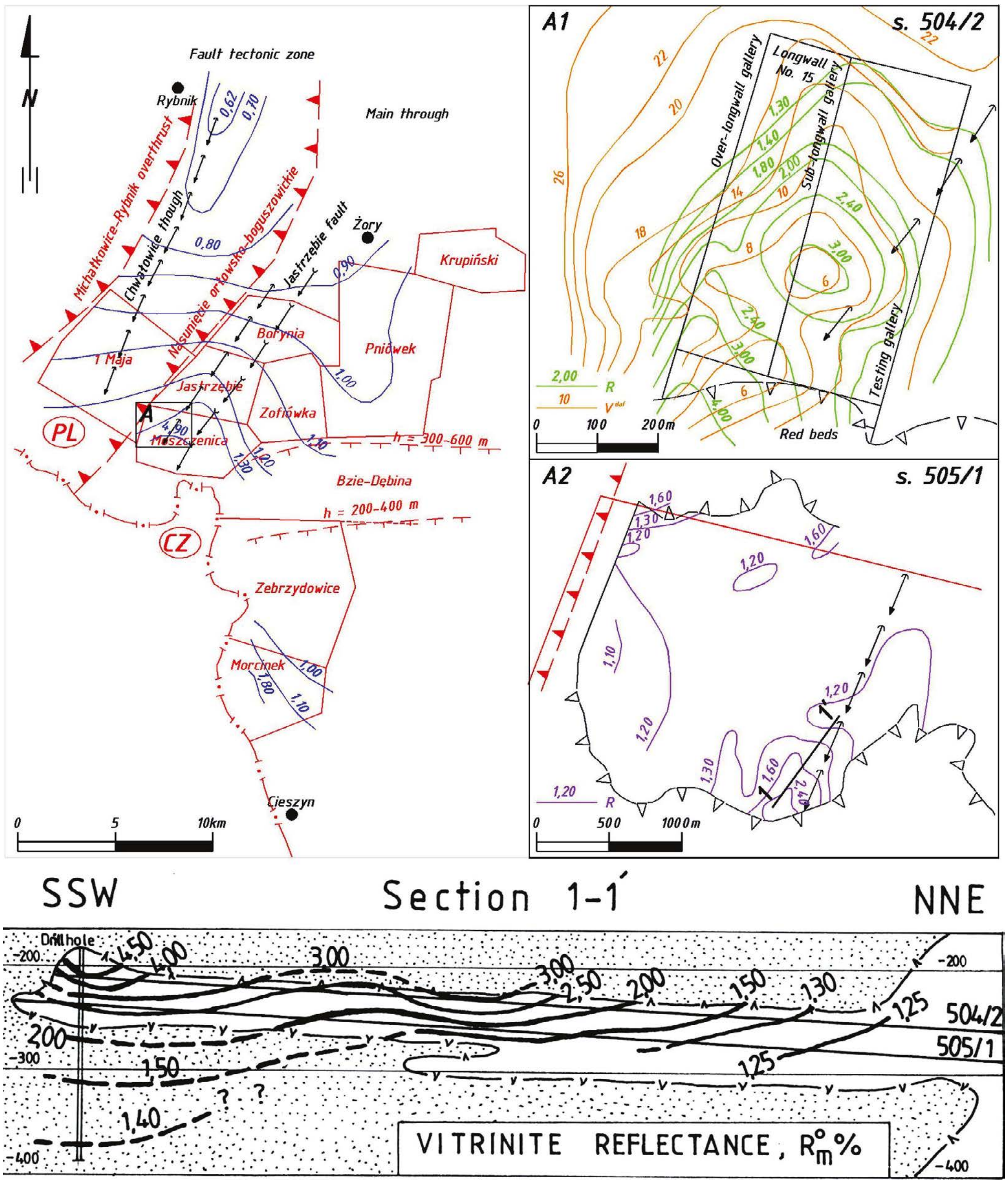

Fig. 2. A sketch of vitrinite reflectance of coal in the SW part of the USCB and the geological profile of the 504/2 and 505/1 seams with the R vitrinite reflectance plotted
A1 – R vitrinite reflectance and the content of V^{daf} volatiles in the 504/2 seam;
A2 – R vitrinite reflectance in the 505/1 seam

Rys. 2. Szkic refleksyjności witrynitu węgli SW części GZW oraz przekrój geologiczny przedstawiający pokłady 504/2 oraz 505/1 wraz z zaznaczeniem refleksyjności witrynitu R
A1 – refleksyjność witrynitu R oraz zawartość części lotnych V^{daf} w pokładzie 504/2
A2 – refleksyjność witrynitu R w pokładzie 505/1

in different states (analytic; dry; dry, ash free; mineral matter free), which is determined by the mentioned standards.

Values of CRI and CSR parameters have been obtained on the basis of the testing of coke according to Nippon Steel Co (NSC) guidelines and Polish Standards, applied at the Institute for Chemical Processing of Coal in Zabrze.

The characteristics of the variability of the CRI and CSR indices were analyzed using statistical methods. The dependencies between the CRI and CSR indices and the parameters having an impact on their values were determined using linear correlation. An attempt was also made to determine the correlations between the discussed parameters using the multiple correlation method.

The obtained results were compared to the data presented in the papers (North et al. 2018a, b) and presented against the charts included in these papers. This was done due to the fact that the data may be considered the compilation of results of globally conducted experiments in coke quality prediction. In the paper of (North et al. 2018b), published models for the prediction of various measures of coke quality, with a particular emphasis on coke strength after reaction (CSR) and the related coke reactivity index (CRI) were analyzed. The focus was placed on the coal parameters selected as model inputs, and their reported behavior with respect to the predicted coke quality. The authors ascertained that there is a limited range of model applicability beyond the specific range of coals for which each model was derived, which suggests the inconsistent utilization of key attributes contributed to these limitations.

3. Results and discussion

The results of the determination of the CRI and CSR indices as well as of the remaining quality parameters for the "Pniówek" deposit were presented in Tables 1–3.

Such a compilation of data subsequently allowed to track the mutual correlations between the CRI, CSR and the analyzed quality parameters. The achieved (r) correlation coefficient values allow to conclude that both indices usually exhibit a very low correlation (or none) with the remaining parameters (Table 4).

The value of the (r) coefficient of correlation between the CRI and CSR indices amounts to –0.9.

Table 4 indicates that the CRI index is less correlated with the quality parameters than it is in the case of the CSR index. In case of the CRI index, only two cases have been noted in which the (r) value equaled or exceeded 0.5, reaching –0.6 for the correlation with the hydrogen content (H_t^a) and 0.5 for the correlation with the actual density of coal (d_r^a). A more common correlation (7 cases) in an analogous compilation was exhibited by the CSR index, reaching 0.6 for the correlation with the hydrogen content (H_t^a), 0.5 in case of the correlation with Na_2O Al_2O_3 and SiO_2 and –0.5 for the correlation with Mn_3O_4 and the density of coal – both the actual (d_a^a) and apparent (d_r^a) one.

Table 1. The results of elemental and technical analyses of coal samples from "Pniówek" deposit

Tabela 1. Wartości wskaźników analizy technicznej i elementarnej węgli ze złoża „Pniówek"

No.	W^a	A^d	V^{daf}	GCV^{daf}	CV^{daf}	S_t^d	Cl^a	P^a	C_t^a	H_t^a	N_t^a	O_t^a
	%	%	%	MJ/kg	MJ/kg	%	%	%	%	%	%	%
1	0.8	9.7	28.1	37.3	36.1	0.8	0.11	0.023	84.7	5.0	1.3	0.7
2	0.4	5.7	27.8	36.2	35.0	0.7	0.10	0.036	86.5	5.0	1.5	0.3
3	0.7	6.0	30.9	35.3	34.1	0.5	0.15	0.034	84.8	5.1	1.4	1.6
4	0.6	5.8	27.3	35.4	34.2	0.5	0.13	0.045	86.0	4.8	1.3	1.0
5	0.7	5.8	29.0	36.4	35.2	0.5	0.11	0.047	85.3	5.0	1.5	1.3
6	0.5	6.2	26.4	36.5	35.3	0.4	0.11	0.122	82.8	5.1	1.4	3.5
7	0.6	3.9	26.4	36.7	35.6	0.5	0.12	0.017	88.3	5.1	1.5	0.1
8	0.6	7.1	31.6	36.0	34.8	0.5	0.14	0.095	83.7	5.1	1.4	1.7
9	0.8	7.2	32.9	35.5	34.3	0.6	0.13	0.028	83.4	4.8	1.4	1.9
10	0.6	6.7	25.8	36.7	35.5	0.4	0.12	0.093	84.6	4.9	1.4	1.5
11	0.7	7.3	26.2	36.2	35.0	0.5	0.13	0.045	84.0	4.9	1.4	1.2
12	0.6	5.8	27.4	36.1	35.0	0.5	0.27	0.033	85.4	5.1	1.3	1.4
13	0.9	3.8	26.8	36.4	35.2	0.4	0.12	0.030	88.7	4.7	1.5	0.1
14	0.4	3.9	27.7	36.1	34.9	0.7	0.08	0.074	88.0	4.9	1.5	0.7
15	0.6	7.2	26.1	36.1	34.9	0.5	0.12	0.190	84.5	4.8	1.4	1.0
16	1.0	8.2	28.8	35.8	34.6	0.7	0.13	0.104	83.0	4.9	1.3	0.9
17	0.5	5.4	26.1	36.2	35.0	0.5	0.11	0.085	85.8	4.9	1.4	1.5
18	1.0	11.9	27.7	39.0	37.9	0.8	0.13	0.062	86.6	4.6	1.4	1.1
19	0.6	6.5	29.0	35.9	34.7	0.5	0.10	0.033	85.7	4.8	1.4	0.6
20	0.8	5.1	30.8	35.7	34.5	0.5	0.15	0.057	85.4	4.9	1.4	2.0
21	0.6	7.6	26.2	35.7	34.6	0.5	0.10	0.079	84.2	4.8	1.4	1.0
22	0.6	7.6	30.8	34.7	33.6	0.8	0.09	0.025	83.4	4.8	1.4	1.4
23	1.0	5.7	26.5	35.6	34.4	0.4	0.12	0.052	86.4	5.0	1.4	0.2
24	0.7	3.6	29.3	36.1	34.9	0.5	0.12	0.049	87.0	5.2	1.4	1.6
25	1.1	5.1	29.7	36.0	34.8	0.7	0.17	0.083	85.8	5.1	1.4	0.9
Min.	0.4	3.6	25.8	34.7	33.6	0.4	0.08	0.017	82.8	4.6	1.3	0.1
Max.	1.1	11.9	32.9	39.0	37.9	0.8	0.27	0.190	88.7	5.2	1.5	3.5
Mean	0.7	6.3	28.2	36.1	35.0	0.6	0.13	0.062	85.4	4.9	1.4	1.2
Standard deviation	0.19	1.87	1.99	0.80	0.80	0.13	0.04	0.039	1.61	0.14	0.05	0.73

W^a – moisture content (analytic state), A^d – ash content (dry state), V^{daf} – volatile content (dry, ash free state), GCV^{daf} – gross calorific value (dry, ash free state), CV^{daf} – calorific value (dry, ash free state), S_t^d – sulphur total (dry state), Cl^a – chlorine content (analytic state), P^a – phosphorus content (analytic state), C_t^a – carbon content (analytic state), H_t^a – hydrogen content (analytic state), N_t^a – nitrogen content (analytic state), O_t^a – oxygen content (analytic state).

Table 2. Chemical composition of ash of coal samples from "Pniówek" deposit

Tabela 2. Wartości parametrów składu chemicznego popiołu z węgli ze złoża „Pniówek"

No.	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	K_2O	CaO	TiO_2	Mn_3O_4	Fe_2O_3
	%	%	%	%	%	%	%	%	%	%	%
1	1.0	2.2	33.9	44.2	0.8	1.4	2.3	2.0	1.0	0.1	9.1
2	1.3	2.8	31.9	41.7	1.5	2.9	3.0	3.6	1.0	0.1	9.6
3	1.5	2.8	30.8	35.4	1.3	3.5	1.9	3.7	0.9	0.2	17.4
4	1.2	4.7	23.1	24.0	1.8	5.6	1.5	5.1	0.6	0.3	31.4
5	1.4	5.1	19.5	15.9	1.9	4.7	0.7	6.8	0.5	0.6	42.2
6	1.4	3.0	28.7	27.5	4.5	4.9	0.8	8.6	0.8	0.2	18.8
7	2.2	4.2	23.5	36.8	1.0	7.4	1.0	8.8	0.6	0.2	13.4
8	1.1	4.8	24.0	20.6	3.1	4.9	1.4	7.8	0.9	0.4	30.5
9	0.9	4.4	13.9	6.5	0.9	5.5	0.3	6.2	0.4	0.6	6.0
10	1.4	3.9	26.3	30.4	3.2	7.8	1.3	12.1	1.0	0.2	10.7
11	1.6	4.5	30.1	41.6	1.4	5.1	2.0	6.1	0.8	0.1	6.1
12	2.0	2.9	28.3	43.6	1.3	2.9	3.1	3.7	1.0	0.1	10.5
13	2.0	6.3	22.2	17.3	1.9	8.0	0.9	9.0	0.6	0.3	31.4
14	2.0	4.8	25.9	22.7	4.4	7.4	0.9	12.2	0.5	0.3	17.9
15	1.8	1.0	35.0	37.0	6.6	3.1	1.2	8.1	0.8	0.1	4.4
16	1.7	1.8	34.8	42.8	2.9	1.4	2.3	3.8	1.0	0.1	6.0
17	1.9	4.3	29.4	31.2	3.7	5.3	1.9	10.1	0.8	0.2	10.7
18	1.5	1.6	35.8	41.1	2.9	1.4	2.1	3.8	1.1	0.1	7.6
19	1.0	5.7	16.2	9.7	1.2	7.7	0.6	11.6	0.4	0.5	44.3
20	1.6	4.0	20.5	14.7	2.5	3.6	0.7	6.4	0.5	0.6	43.9
21	1.4	5.0	23.3	20.3	2.4	5.0	0.9	5.1	0.6	0.4	34.7
22	0.6	6.6	15.0	9.8	0.8	5.7	0.5	7.7	0.3	0.6	52.1
23	1.7	2.0	29.6	36.2	2.1	4.5	2.1	6.0	0.9	0.2	14.3
24	1.7	2.8	34.5	32.7	3.2	2.8	1.1	4.8	0.9	0.1	14.0
25	2.4	3.2	31.8	36.0	3.8	3.1	1.7	5.7	0.8	0.1	10.8
Min.	0.6	1.0	13.9	6.5	0.8	1.4	0.3	2.0	0.3	0.1	4.4
Max.	2.4	6.6	35.8	44.2	6.6	8.0	3.1	12.2	1.1	0.6	52.1
Mean	1.5	3.8	26.7	28.8	2.4	4.6	1.4	6.7	0.7	0.3	19.9
Standard deviation	0.4	1.5	6.45	11.8	1.4	2.0	0.8	2.8	0.2	0.2	14.4

Table 3. Coking and physical indices and petrographic composition of coal samples from "Pniówek" deposit

Tabela 3. Wartości wskaźników koksowniczych i fizycznych oraz składu petrograficznego węgli ze złoża „Pniówek"

No.	**CRI**	**CSR**	RI	FSI	a	b	d_a^a	d_r^a	HGI	R	Vt^{mmf}	L^{mmf}	I^{mmf}
	%	**%**	–	–	%	%	g/cm³	g/cm³	–	%	% vol.	% vol.	% vol.
1	**19.9**	**65.3**	83	8.5	29	164	1.3	1.3	84	1.12	80	7	13
2	**23.4**	**59.1**	85	8.5	28	199	1.3	1.4	84	1.09	81	4	15
3	**30.7**	**49.7**	84	8.5	29	139	1.3	1.3	70	1.00	72	9	19
4	**60.8**	**24.4**	84	8.5	29	138	1.3	1.4	90	1.12	78	8	14
5	**34.2**	**49.9**	87	8.5	30	161	1.3	1.4	83	1.08	74	5	21
6	**24.2**	**61.8**	87	8.5	30	175	1.2	1.4	89	1.12	72	6	22
7	**30.9**	**53.6**	88	8.5	31	180	1.2	1.3	96	1.13	70	8	22
8	31.2	47.1	86	8.5	29	172	1.3	1.4	74	1.04	77	10	13
9	**40.6**	**34.2**	80	8.5	28	112	1.3	1.4	76	0.98	70	11	19
10	**58.3**	**27.6**	84	8.5	30	161	1.3	1.4	91	1.14	81	5	14
11	**29.3**	**55.2**	86	8.5	30	188	1.3	1.3	86	1.10	64	7	29
12	**33.6**	**49.3**	84	8.5	31	120	1.3	1.4	86	1.08	74	9	17
13	**34.0**	**51.7**	87	8.5	35	141	1.3	1.4	92	1.08	74	5	21
14	**27.7**	**60.7**	86	8.5	31	160	1.3	1.4	82	1.06	60	9	31
15	**37.5**	**42.7**	86	8.5	30	180	1.3	1.4	87	1.10	70	4	26
16	**27.0**	**54.0**	87	8.5	26	140	1.3	1.4	78	1.05	80	6	14
17	**25.7**	**60.9**	84	8.5	31	186	1.2	1.3	90	1.12	74	7	19
18	58.3	26.7	87	8.5	28	144	1.4	1.4	84	1.06	74	9	17
19	**52.1**	**23.0**	86	8.5	32	178	1.3	1.4	88	1.10	85	4	11
20	**45.1**	**32.7**	87	8.5	30	108	1.3	1.4	76	0.98	74	8	18
21	**40.2**	**43.6**	85	8.5	28	140	1.3	1.4	82	1.08	65	7	28
22	**41.3**	**27.2**	84	8.5	30	184	1.4	1.4	94	1.08	81	6	13
23	**33.6**	**47.2**	80	6.0	31	58	1.4	1.4	83	1.09	78	6	16
24	**24.6**	**63.3**	86	8.5	29	127	1.3	1.3	69	0.99	65	10	25
25	**30.6**	**52.8**	84	8.5	31	191	1.3	1.4	72	0.98	74	9	17
Min.	**19.9**	**23.0**	80	6.0	26	58	1.2	1.3	69	0.98	60	4	11
Max.	**60.8**	**65.3**	88	8.5	35	199	1.4	1.4	96	1.14	85	11	31
Mean	35.8	46.5	85	8.5	30	154	1.3	1.4	83	1.07	74	7	19
Standard deviation	**11.43**	**13.30**	2	0.5	2	33	0.04	0.03	7	0.05	6	2	5

CRI – coke reactivity index, CSR – coke strength after reaction, RI – Roga index, FSI – free swelling index, a – contraction, b – dilatation, d_a^a – actual density (analytic state), d_r^a – apparent density (analytic state), HGI – Hardgrove index, R – vitrinite reflectance, Vt^{mmf} – vitrinite contetnt (mineral matter free state), L^{mmf} – liptinite contetnt (mineral matter free state), I^{mmf} – inertinite contetnt (mineral matter free state).

Table 4. The values of the (r) correlation coefficient for the dependencies between the CRI and CSR indices and the analyzed quality parameters of coal from the "Pniówek" deposit and ash from this coal

Tabela 4. Wartości współczynnika korelacji (r) dla zależności pomiędzy wskaźnikami CRI i CSR a analizowanymi parametrami jakościowymi węgli ze złoża „Pniówek"

r	W^a	A^d	V^{daf}	GCV^{daf}	CV^{daf}	S_t^d	Cl^a	P^a	C_t^a	H_t^a	N_t^a	O_t^a
	%	%	%	MJ/kg	MJ/kg	%	%	%	%	%	%	%
CRI, %	0.1	0.3	0.0	0.1	0.1	–0.1	0.0	0.0	0.0	**–0.6**	–0.3	0.0
CSR, %	–0.1	–0.3	–0.2	0.1	0.1	0.0	0.0	0.1	0.2	**0.6**	0.3	0.0
r	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	K_2O	CaO	TiO_2	Mn_3O_4	Fe_2O_3	
	%	%	%	%	%	%	%	%	%	%	%	
CRI, %	–0.3	0.2	–0.4	–0.4	–0.1	0.3	–0.3	0.2	–0.2	0.3	0.3	
CSR, %	**0.5**	–0.2	**0.5**	**0.5**	0.2	–0.3	0.4	–0.2	0.4	**–0.5**	–0.4	
r	RI	FSI	a	b	d_a^a	d_r^a	HGI	R	Vt^{mmf}	L^{mmf}	I^{mmf}	
	–	–	%	%	g/cm^3	g/cm^3	–	%	% vol.	% vol.	% vol.	
CRI, %	0.0	0.0	0.0	–0.2	0.4	**0.5**	0.3	0.1	0.3	–0.1	–0.3	
CSR, %	0.1	0.0	0.0	0.2	**–0.5**	**–0.5**	–0.2	0.0	–0.4	0.1	0.4	

Technical analysis indicators, characterizing the general properties of coal, encompassed the determination of 5 parameters (Table 1): the (W^a) moisture content ranging from 0.4–1.1% (mean of 0.7%): the (A^d) ash content ranging from 3.6–11.9% (mean of 6.3%), the (V^{daf}) content of volatiles ranging from 25.8–32.9% (mean of 28.2%), the (GCV^{daf}) combustion heat ranging from 34.7 to 30.0 MJ/kg (mean of 36.1 MJ/kg) and the (CV^{daf}) calorific value ranging from 33.6 to 37.9 MJ/kg (mean of 35.0 MJ/kg). Moreover, apart from the quantitative determination of the ash content (as a percentage) its chemical composition was analyzed – encompassing 13 compounds (Table 2): Na_2O ranging from 0.6–2.4% (mean of 1.5%), MgO ranging from 1.0–6.6% (mean of 3.8%), Al_2O_3 ranging from 13.9––35.8% (mean of 26.7%), SiO_2 ranging from 6.5–44.2% (mean of 28.8%), P_2O_5 ranging from 0.8–6.6% (mean of 2.4%), SO_3 ranging from 1.4–8.0% (mean of 4.6%), K_2O ranging from 0.3–3.1% (mean of 0.7%), CaO ranging from 2.0–12.2% (mean of 6.7%), TiO_2 ranging from 0.3–1.1 (mean of 0.7%), Mn_3O_4 ranging from 0.1–0.6% (mean of 0.3%): Fe_2O_3 ranging from 4.4–52.1% (mean of 19.9%).

The correlations between the CRI and CSR indices and the parameters in this group are very weak. The observed relations only concern the 4 cases of the weak correlation of CSR with Na_2O, Al_2O_3 and SiO_2 (r = 0.5) and with Mn_3O_4 (r = –0.5), referred to earlier and presented in Figure 3.

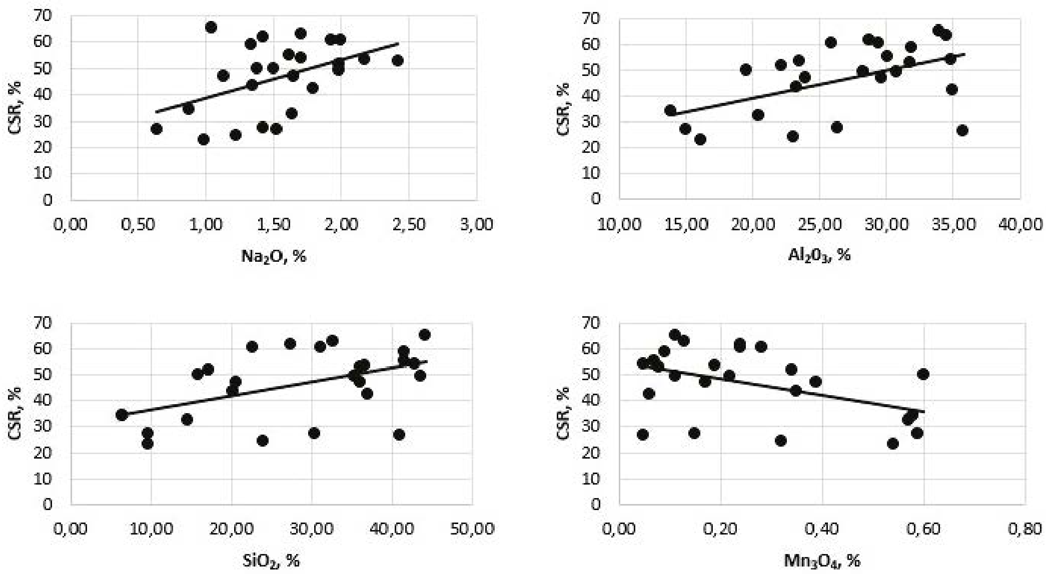

Fig. 3. Correlations between CSR index and selected compounds of ash produced from the coal samples in consideration

Rys. 3. Zależność między wskaźnikami CSR
a wybranymi związkami składu popiołu z badanych węgli

The globally conducted experiments related to this group of parameters, regarding the correlation with CRI and CSR, mostly only concern the content of volatiles. This is why it is the only parameter included in the charts by (North et al. 2018b). The marked range of values, the trend line and the mean value of volatiles content in relation to the CRI and CSR values generally appear above the curves plotted in these charts, especially in relation to the single coals (Fig. 4). The presented short trend line does not, however, deviate from the indicated curves and – in several cases – it is convergent.

The indices provided by elemental analysis, describing the content of elements, encompassed the determination of 7 parameters (Table 1): the (S_t^d) total content of sulphur ranging from 0.4 to 0.8% (mean of 0.6%), the (Cl^a) chlorine content ranging from 0.08–0.27% (mean of 0.13%), the (P^a) phosphorus content ranging from 0.017–0.190% (mean of 0.062%), the (C_t^a) carbon content ranging from 82.8–88.7% (mean of 85.4), the (H_t^a) hydrogen content ranging from 4.6–5.2% (mean of 4.9%), the (N_t^a) nitrogen content ranging from 1.3–1.5% (mean of 1.4%) and the (O_t^a) content of oxygen ranging from 0.1 to 3.5% (mean of 1.2%).

Within this group, only the (H_t^a) hydrogen content exhibits a relationship with the CRI and CSR, with the correlation coefficient amounting to r = –0.6 in case of CRI and r = 0.6 in case of CSR (Fig. 5).

The analyzed professional literature does not, however, provide information regarding the correlation between the parameters of that group and the CRI and CSR indices.

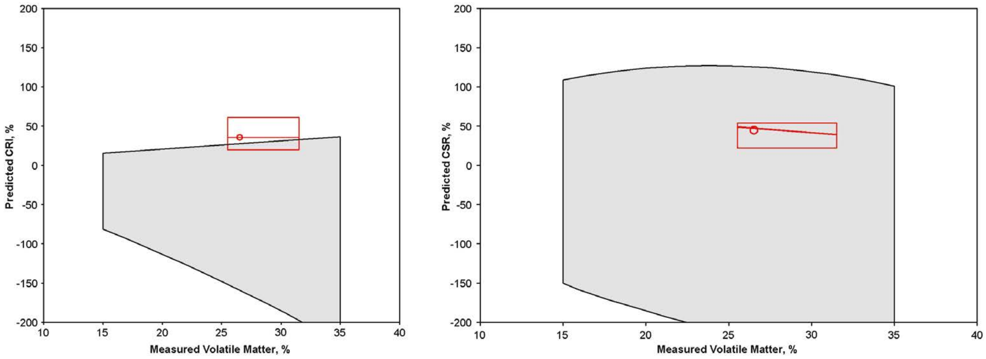

Fig. 4. Correlations between CRI and CSR indices and volatile matter content,
according to (North et al. 2018b) – grey field

Fig. 4. Zależność między wskaźnikami CRI i CSR
a zawartością części lotnych, w odniesieniu do (North i in. 2018b) – szare pole

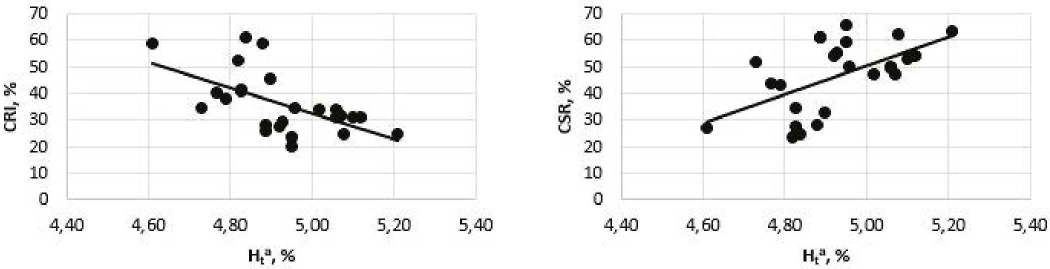

Fig. 5. Correlations between CRI and CSR indices and the hydrogen content

Rys. 5. Zależność między wskaźnikami CRI i CSR a zawartością wodoru

The indicators provided by means of coking properties analysis describe the behavior and the technological properties of coal during its thermal decomposition. The analyses encompassed the determination of 6 parameters (Table 3): the CRI index in the range between 19.9 and 60.8% (mean of 35.8%), the CSR index in the range between 23.0 and 65.3% (mean of 46.5%), the (RI) Roga caking power ranging between 80 and 88 (mean of 85), the (FSI) free swelling index in the range between 6 and 9 (mean of 8) and the dilatometric parameters – the (a) contraction in the range of 26–35% (mean of 30%) and dilatation (b) ranging between 58 and 199% (mean of 154%).

The correlation between the CRI and CSR indices is very strong, with the correlation coefficient value reaching r = –0.9 (Fig. 6). No other correlations between the CRI and CSR indices and the remaining parameters of this group have been identified. The observed de-

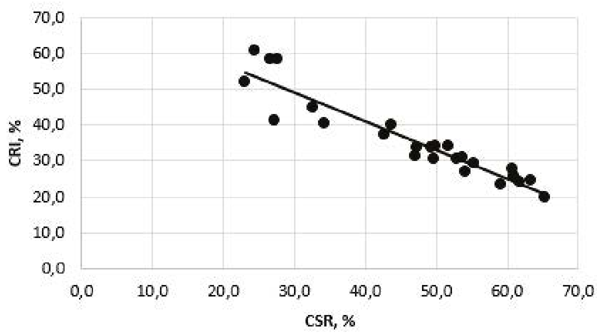

Fig. 6. Correlations between the CRI and CSR indices

Rys. 6. Zależność między wskaźnikami CRI i CSR

pendence between CRI and CSR has already been a subject of investigations done in the past (Probierz and Marcisz 2015).

To provide a comparison with the results of globally conducted experiments, the observed correlation between the CRI and CSR has also been presented in a chart by (North et al. 2018b). The range of values, the trend line and the mean value generally appear within the range of the curves plotted in these charts, especially in relation to the single coals (Fig. 7). The presented trend line is convergent to most of the plotted curves.

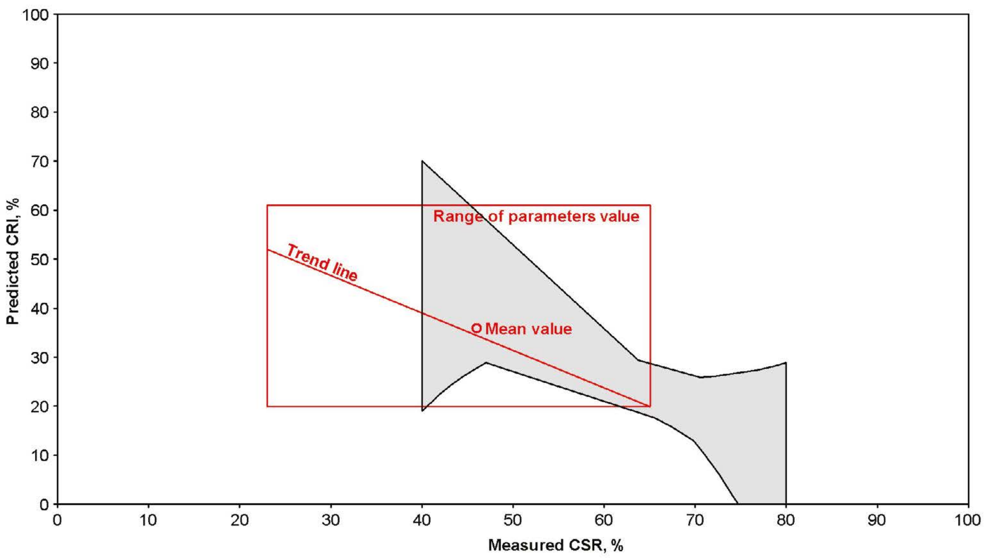

Fig. 7. Correlations between the CRI and CSR indices, according to (North et al. 2018b) – grey field

Rys. 7. Zależność między wskaźnikami CRI i CSR, w odniesieniu do (North i in. 2018b) – szare pole

The analyzed professional literature also exhibits the researchers' interest in the dependence between the CRI and CSR indices and the dilatation. In the conducted study, no such dependence was observed. However, the range of the values, the trend line and the mean values were plotted against the chart by (North et al. 2018b). In that case, however, all the elements referred to above are considerably outside the boundaries of the chart (Fig. 8), which

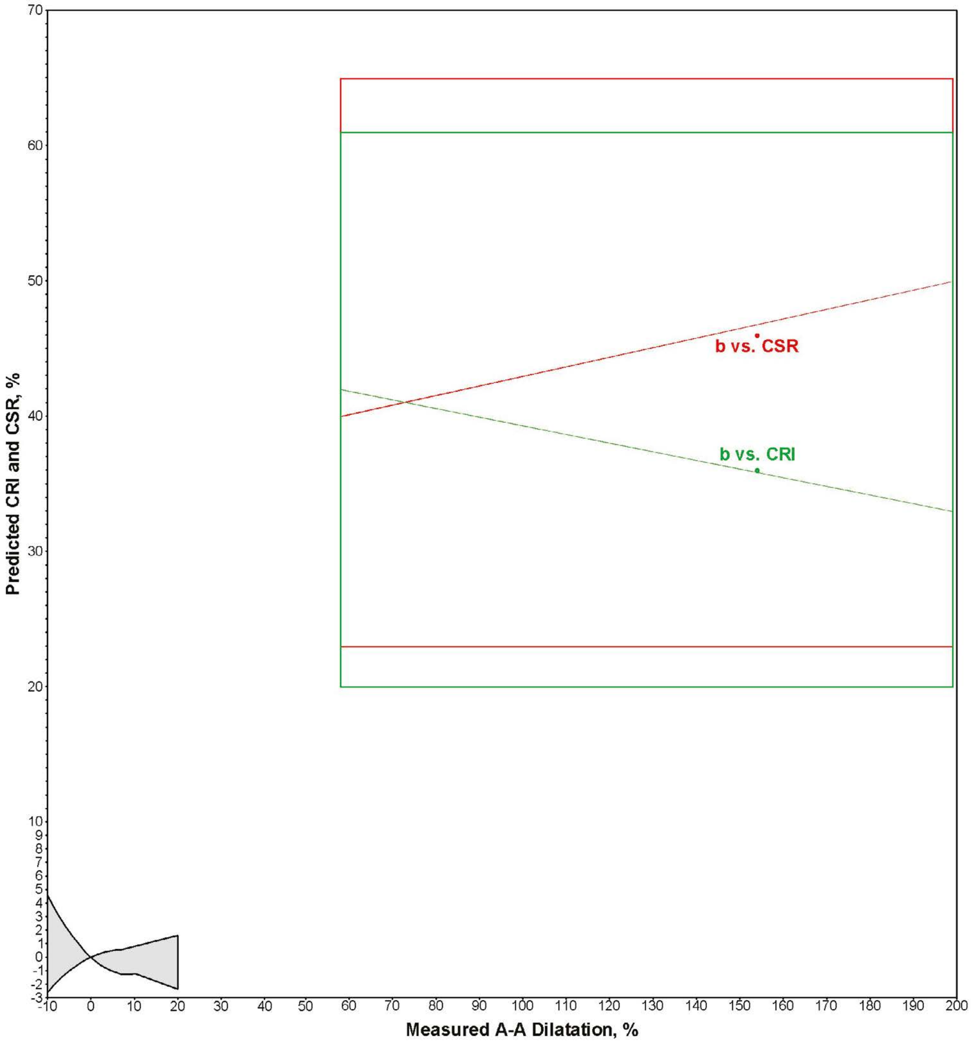

Fig. 8. Correlations between the CRI (green) and CSR (red) indices and the dilatation, according to (North et al. 2018b) – grey field

Rys. 8. Zależność między wskaźnikami CRI (na zielono) i CSR (na czerwono) a dylatacją, w odniesieniu do (North i in. 2018b) – szare pole hinders their interpretation and comparison. The spans of the values are highly different and thus the mean values are also discrepant. The trend lines also assume different shapes.

Physical indicators encompassed the determination of 4 parameters (Table 3): the ($d_a{}^a$) apparent density ranging between 1.22 and 1.40 g/cm^3 (mean of 1.30 g/cm^3), the ($d_r{}^a$) actual density ranging between 1.29 and 1.43 g/cm^3 (mean of 1.37 g/cm^3), the Hardgrove grindability index ranging from 69 to 96 (mean of 83) and the (R) vitrinite reflectance ranging from 0.98 to 1.14 (mean of 1.07%).

The correlations between the CRI and CSR parameters and the physical indicators are very weak. Within this group of parameters, only the apparent and actual density exhibited a relationship with the CRI and CSR: for the correlation between CRI and $d_r{}^a$, the coefficient amounted to r = 0.5, while for the CSR vs $d_a{}^a$ and CSR vs $d_r{}^a$ correlation, the coefficient amounted to r = 0.5 (Fig. 9).

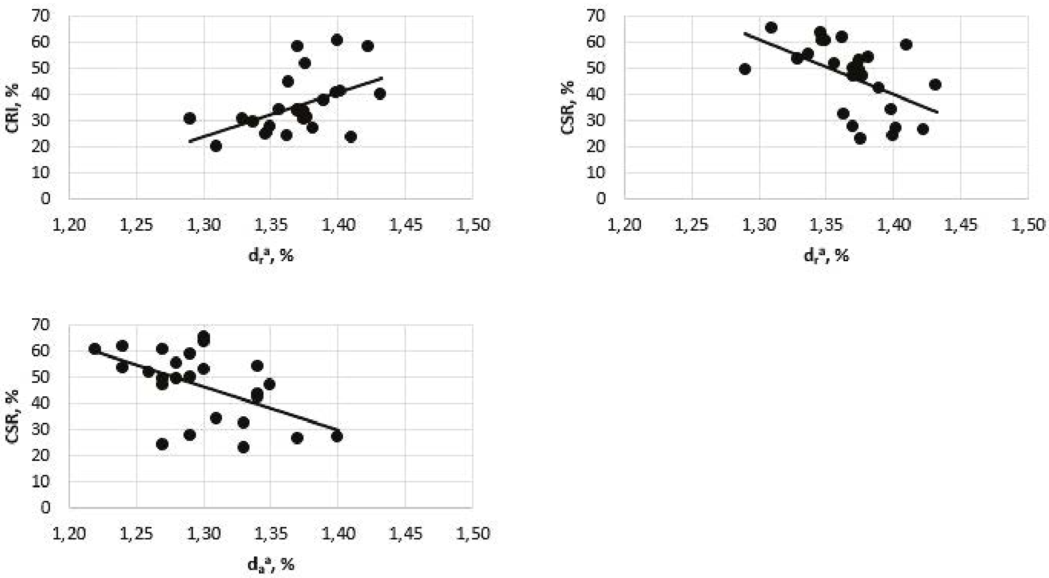

Fig. 9. Correlations between the CRI and CSR indices and the density of coal

Rys. 9. Zależność między wskaźnikami CRI i CSR a gęstością węgla

The analyzed professional literature allows to conclude that numerous studies encompassed and concerned the relations between the CRI and CSR indices and the (R) vitrinite reflectance. In the conducted research, no such dependence was observed. However, the range of the values, the trend line and the mean value were plotted against the charts by (North et al. 2018b, Fig. 10). The marked range of values, the trend line and the mean value of vitrinite reflectance, as compared to the CRI value, fits in the upper boundary of the curves

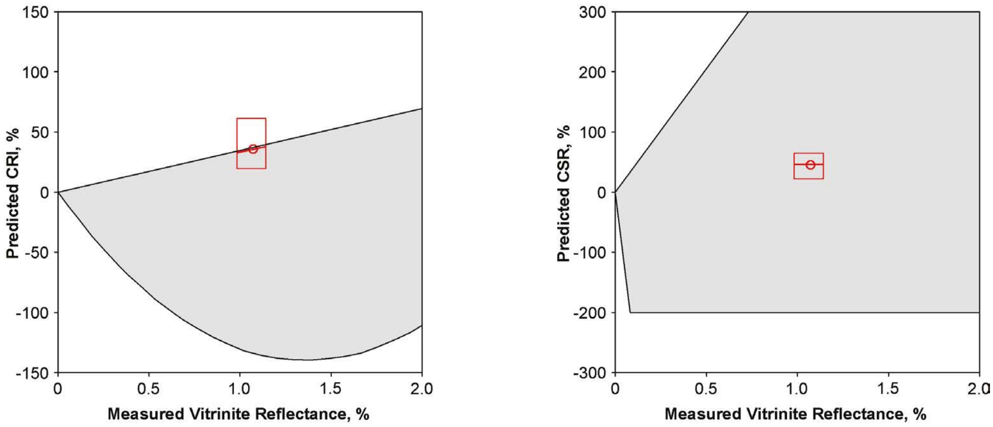

Fig. 10. Correlations between the CRI and CSR indices and the vitrinite reflectance, according to (North et al. 2018b) – grey field

Rys. 10. Zależność między wskaźnikami CRI i CSR a refleksyjnością witrynitu, w odniesieniu do (North i in. 2018b) – szare pole

plotted in the chart. The presented short trend line of reflectance is convergent only to one of the presented dependencies. In relation to the CSR value, the range of values, the trend line and the mean value of vitrinite reflectance fully fits within the curves plotted in the chart and in this case the trend line of the reflectance is convergent to several of the presented dependencies.

The study of the relationships between the CRI and CSR indices and the reflectance of vitrinite in coal from the "Pniówek" deposit has been conducted earlier in the paper (Probierz and Marcisz 2015). The observed lack of correlation between these parameters was compared to the results of research conducted by (Nakamura et al. 1977; Menendez et al. 1999) as well as (Pusz and Buszko 2012) while trying to exhibit the possible similarities (or differences) between the coal of the "Pniówek" deposit and other coal deposits around the world (Fig. 11).

Comparing position of coals of the "Pniówek" deposit with coals analyzed in the work (Nakamura et al. 1977) it may be proved that coals from the "Pniówek" deposit take place in the middle part of presented range of coalification degree (R = 0.98–1.14). The position of part of the coals from the "Pniówek" deposit differ from the chart, what may be connected with the presence of thermal metamorphism in this region. The dependence of CRI from CSR is, as it seems to be, accordant with dependence showed in the works (Menendez et al. 1999; Pusz and Buszko 2012).

The analysis of the petrographic composition of coal from the "Pniówek" deposit encompassed the determination of the content of macerals from the following groups: (Vt^{mmf}) vitrinite in the range of 60–85% vol. (mean of 74% vol.), (L^{mmf}) liptinite in the range of 4–11% vol. (7% vol.) and (I^{mmf}) inertinite in the range of 11–39% vol. (19% vol.).

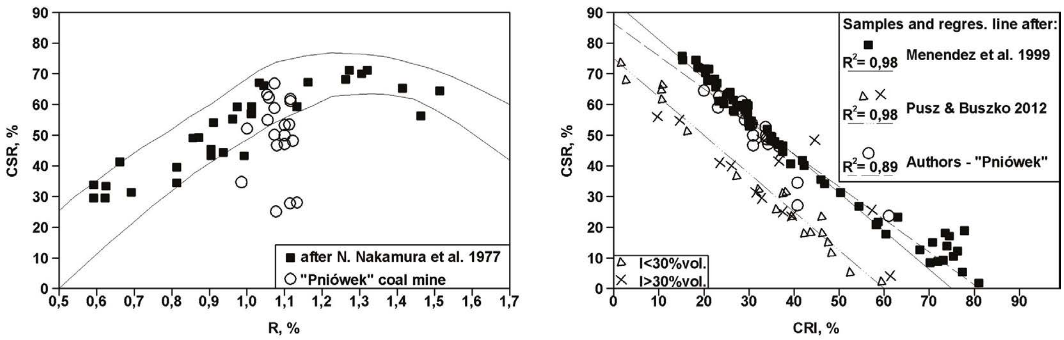

Fig. 11. The position of coals from the Pniówek deposit in view of the correlation between the CRI, CSR and the vitrinite reflectance and the results from other coal basins, according to (Probierz and Marcisz 2015)

Rys. 11. Pozycja węgli ze złoża „Pniówek" na tle zależności pomiędzy CRI, CSR oraz refleksyjnością witrynitu w świetle wyników badań węgli z innych zagłębi węglowych, za (Probierz i Marcisz 2015)

Based on the conducted analyses, no correlation was found between the CRI and CSR indices and the petrographic composition of the studied coal.

It should be mentioned that the dependencies between the CRI and CSR indices and the petrographic composition of the "Pniówek" deposit coal have been the subject of previous research (Probierz and Marcisz 2015). In the paper, it has been concluded that the CRI is positively correlated with the (Vt^{mmf}) vitrinite content, while it is inversely proportional to the (I^{mmf}) inertinite content. The characteristics of these correlations is opposite in case of the CSR index. It has also been established that the dependence relating to the inertinite group indicates a necessity to conduct further, more detailed research encompassing the relations between the CRI and CSR indices and the inertinite, focusing on the macerals of this group. The results of the CRI and CRS values, the petrographic composition and the coalification degree of the coal in the "Pniówek" deposit coal, obtained in the quoted paper, have been subject to interpretation in view of coal from other coal basins. This was the result of a lack of such determinations regarding Polish coking coal deposits.

Conclusions

The knowledge of the distribution of the CRI and CSR parameter values in a coking coal deposit is extremely significant to the coking industry due to the possibility of producing coke characterized by the highest quality parameters that fulfil the customer's expectations.

The research, encompassing statistical analyses, was conducted for a deposit that is substantial to the Polish coking coal reserves, characterized by high variability in terms of the quality and coalification level.

The tests encompassed 36 quality parameters determined for channel samples acquired from the workings of the "Pniówek" mine.

Conducted preliminary statistical analysis showed some interesting relations. It is obvious that all the obtained results require further research.

No clear dependence of the CRI and CSR indices was exhibited in case of most of the analyzed quality parameters, which is supported by low correlation coefficients of $r < 0.5$. The statistical analysis exhibited only 9 cases of correlation between CRI and CSR and other quality parameters, where the correlation coefficient was $r \geq 0.5$, that is: H_t^a, Na_2O, Al_2O_3 and SiO_2, Mn_3O_4, d_a^a and d_r^a.

As it was established in the paper (Probierz and Marcisz 2015), the lack of correlation may be connected to the small number of samples. It should be highlighted that the low value of the correlation coefficient does not mean that no relationship exists between random variables. This relationship may exist but it might be different from the linear (rectilinear) one. Similarly, if the correlation coefficient is close to unity, it does not mean that a causal effect connection exists between random variables. This only pertains to strong stochastic dependencies (it might be an incidental similarity of two samples). The value of the correlation coefficient of a sample may also be underrated in case of the occurrence of measuring errors of one or two variables (there are statistic measures which take such errors into account).

The cause of the aforementioned lack of correlation can also be the insufficiently broad range of variability of the analyzed parameters in the examined population of samples. In such a case, the variance caused by the variability of the examined parameters is too small in relation to the variance caused by measurement errors, and, therefore, the results of the statistical analysis cannot confirm the analyzed significant correlations, which, obviously, does not exclude the existence of such correlations.

In view of the research conducted worldwide, according to (North et al. 2018b), the V^{daf} and R values (the parameters describing the coalification level) of the coal of the "Pniówek" deposit fit within the range for the CSR index. In case of the CRI index, the values of these parameters are at the levels around the upper range presented in the quoted paper. The value of dilatation of the coal from the "Pniówek" deposit exceed the ranges provided by (North et al. 2018b) both in case of CRI and CSR.

The observed dependencies indicate the need to conduct further, more detailed research, including the relationship between the CRI and CSR indices and the remaining quality parameters. The different characteristics of coal from the "Pniówek" deposit have been exhibited multiple times, which should be related to its specific coalification processes, especially the impact of thermal metamorphism. The lack of distinct correlations between the particular parameters can be connected with high probability with the above mentioned symptoms (Fig. 2) indicated.

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THE RELATIONSHIP BETWEEN CRI AND CSR INDICES AND OTHER QUALITY PARAMETERS OF COKING COAL FROM THE PNIÓWEK DEPOSIT (SW PART OF THE USCB, POLAND)

Keywords

CRI and CSR indices, coking coal, quality parameters, Upper Silesian Coal Basin

Abstract

The paper presents the correlation between the CRI (Coke Reactivity Index), CSR (Coke Strength after Reaction) and the remaining 36 quality parameters of coking coal from the Pniówek deposit (SW part of the USCB). The test results were obtained for a region of fundamental importance to the Polish reserves of coking coal, characterized by highly variable coalification and quality parameters. The tests related to the determination of relationships of the CRI and CSR indices to other parameters were based on 25 channel samples acquired from active workings. The characteristics of the variability of the CRI and CSR indices were analyzed using statistical methods. The dependencies between the CRI and CSR indices and the parameters having an impact on their values were determined using linear correlation. An attempt was also made to determine the correlations between the concerned parameters using the multiple correlation method. The obtained results have been presented and compared to the results of globally conducted experiments in the form of charts presented by (North et al. 2018b). No clear dependence of the CRI and CSR indices was exhibited in case of most of the analyzed quality parameters, which is supported by low correlation coefficients of $r < 0.5$. The statistical analysis exhibited only 9 cases of correlation between CRI and CSR with other quality parameters, where the correlation coefficient was $r \geq 0.5$, that is: H_t^a, Na_2O, Al_2O_3 and SiO_2, Mn_3O_4, d_a^a and d_r^a.

This confirms the different characteristics of coal from the studied area, exhibited multiple times, that should be related to the specific coalification process, especially the occurrence of thermal metamorphism.

ZALEŻNOŚĆ MIĘDZY WSKAŹNIKAMI CRI I CSR A INNYMI PARAMETRAMI JAKOŚCI WĘGLA KOKSOWEGO ZE ZŁOŻA PNIÓWEK (SW CZĘŚĆ GZW, POLSKA)

Słowa kluczowe

CRI, CSR, węgiel koksowy, parametry jakościowe, Górnośląskie Zagłębie Węglowe

Streszczenie

W artykule przedstawiono zależności korelacyjne pomiędzy wskaźnikami koksowniczymi CRI (*Coke Reactivity Index*) i CSR (*Coke Strength after Reaction*) a pozostałymi 35 parametrami jakości węgli koksowych w złożu Pniówek (SW część GZW). Wyniki badań uzyskano w obszarze o podstawowym znaczeniu dla polskiej bazy zasobowej węgli koksowych, która charakteryzuje się znaczną zmiennością jakości i uwęglenia. Badania związane z określaniem związków i powiązań wskaźników CRI i CSR z innymi parametrami, oparto na podstawie 25 próbek bruzdowych pobranych z czynnych wyrobisk górniczych. Charakterystykę zmian wartości wskaźników CRI i CSR analizowano z użyciem metod statystycznych. Zależności pomiędzy wskaźnikami CRI i CSR a parametrami mającymi wpływ na kształtowanie się ich wartości określono z użyciem korelacji prostoliniowej. Próbowano również określić współzależności pomiędzy analizowanymi parametrami metodą korelacji wielorakiej. Uzyskane wyniki przedstawiono na tle doświadczeń światowych zestawionych w postaci wykresów zaprezentowanych przez (North i in. 2018b). Wykazano brak wyraźnej zależności wskaźników CRI oraz CSR od większości analizowanych parametrów jakościowych o czym świadczą niskie wartości współczynników korelacji $r < 0{,}5$. Analiza statystyczna wykazała jedynie 9 przypadków korelacji CRI i CSR z pozostałymi parametrami jakościowymi, dla których wartość współczynnika korelacji $r \geq 0{,}5$, tj.: H_t^a, Na_2O, Al_2O_3 i SiO_2, Mn_3O_4, d_r^a i d_a^a. Potwierdza to wielokrotnie wykazywaną odmienność węgli z obszaru badań, którą należy wiązać ze specyficznym przebiegiem procesów uwęglenia, a szczególnie z oddziaływaniem metamorfizmu termalnego.