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OPTIMIZATION OF OXIDATION DESULPHURIZATION OF POWER-GENERATING COAL

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Abstract. An experimental statistic model of power-generating coal oxidative desulphurization has been developed and its adequacy has been proved. On the basis of the proposed model the desulphurization optimal conditions for three samples of power-generating coal were determined. The low-sulphur solid fuel with the content of general sulphur of 1.0–1.5 mas % was obtained under established optimal conditions.

Keywords: oxidative desulphurization, coal, sulphur, pyrite, modeling.

1. Introduction

It is well-known that coal is one of the main energy sources among hydrocarbon raw materials. Its deposits (to calculate for equivalent fuel) were 67 % in 2008 as compared to oil deposits of 18 % and gas – of 15 % [1].

An average content of sulphur in coal is within 1.0–1.5 mas % [2]. However there are a lot of deposits which may be referred to sulphuric or high-sulphuric ones [2-4]. Burning of such coals at heat power plants (HPP) leads to pollution of environment by sulphur dioxide and to corrosion of equipment.

We are working out a technological basis of coal desulphurization using oxidation method. Its application in the power engineering, for example, at HPP, would allow using high-sulphuric coal and reducing the pollution of environment by SO_2. The essence of the process is the treatment of the raw material by steam-air mix. Pyrite sulphur, which is the main part of total sulphur in coal, converts sufficiently selectively. During the process the following products are obtained: solid low-sulphuric fuel; resin formed during thermal decomposition of the organic part; desulphurization gases with high (compared to HPP gases) content of sulphur-containing components, which may be converted or utilized using the known methods [5] with the production of liquefied sulphur dioxide, sulphuric acid, *etc.*

In our previous works [3, 4] we used high- and low-metamorphized coal, because it is the main raw material for HPP. Optimal conditions suggested for various types of coal were different. Therefore the aim of this work is to develop a universal experimental-statistic model which would adequately describe the desulphurization process of all investigated types of black coal.

2. Experimental

Desulphurization was carried out in a perfect-mixing reactor under the conditions close to the isothermal ones. The detailed procedure is described in [6]. The oxidant flow rate and sizes of coal grains under investigated temperatures ensure proceeding of sulphur conversion in a kinetic area.

The coal was analyzed using the standard methods [7-11]. The content of sulphur dioxide in the desulphurization gases was determined using chromatography [12].

Conversion of total or pyritic sulphur was calculated without taking into consideration its loss with the resin by the following formula (%):

$$\Delta S_x^a = \frac{S_{x0}^a \cdot 100 - S_x^a \cdot x_C}{S_{x0}^a} \qquad (1)$$

where S_{x0}^a – content of total or pyritic sulphur in the initial coal to calculate for analytical sample, mas %; S_x^a – content of total or pyritic sulphur in desulphurized coal to calculate for analytical sample, mas %; x_C – desulphurizated coal yield, mas %.

The black coal, whose characteristics are given in Table 1, was taken form the mines and concentrating mills of Donetsk and Lviv-Volyn coal basins, as well as from Dobrotvir HPP (Lviv region).

Table 1

Characteristics of the initial samples of coal

Coal type (symbol)	Moisture content, W^a, mass %	Ash, A^d, mass %	Volatiles yield, V^{daf}, mass %	Sulphur content for dry mass, mass %				Relative content of sulphur different types, %		
				total, S^d_t	pyritic, S^d_p	organic, S^d_o	sulphate, $S^d_{SO_4}$	S^d_p/S^d_t	S^d_o/S^d_t	$S^d_{SO_4}/S^d_t$
Candle (C)	4.85	11.36	37.67	3.98	2.34	0.54	1.1	58.79	13.57	27.64
Gas (G1)	1.21	16.80	40.91	7.95	7.20	0.52	0.23	90.57	6.54	2.89
Gas (G2)	1.17	20.82	41.11	7.18	6.05	0.60	0.54	84.23	8.31	7.46
Lean (L)	1.00	43.10	16.53	4.48	3.81	0.54	0.13	85.04	12.05	2.90
Anthracite (A1)	3.50	6.22	2.00	2.85	1.41	1.21	0.23	49.47	42.46	8.07
Anthracite (A2)	6.40	19.44	5.70	3.07	1.94	0.92	0.20	63.41	29.97	6.62
Run-of-mine (R)	2.83	24.42	35.80	1.92	1.52	0.3	0.10	79.14	15.51	5.35

To describe the main characteristics of the process the following response functions were used: Y_1 – fractional conversion of pyrite sulphur (FCPS), %; Y_2 – content of sulphur dioxide in the desulphurization gases (CSD), vol %. The following process parameters were taken into consideration during the model development: X_1 – temperature, C; X_2 – process time, min; X_3 – content of water steam in the oxidant, vol %; X_4 – repetition factor of oxidant flow rate (RFO), m^3/h/kg of coal. The analysis of the previous results [3, 4] shows that in the range of the investigated coals the main characteristics of the process (response functions) are practically independent of the quality of coal organic mass. On the other hand, sulphur content (primarily pyrite sulphur) affects SO_2 amount in the desulphurization gases and mostly the character of parameters influencing the fractional conversion of pyrite sulphur. Therefore, the content of pyrite sulphur in the initial coal was chosen as another parameter of the process that indicates the influence of the raw material quality (X_5, S^d_p, mas %).

To estimate the adequacy of the obtained regression equation we substituted the given experimental parameters (X_1–X_5) and found the expected (regressive) values of response functions. In accordance with the obtained values we calculated the remainders:

$$\Delta Y_{ij} = Y_{ij}^{reg} - Y_{ij} \qquad (2)$$

where Y_{ij} – values observed during the experiments; Y_{ij}^{reg} – values of response functions calculated using the regression equations; i – number of response function (criterion, parameter; ($i = \overline{1,2}$)); j – number of experiment.

The estimation of model adequacy was conducted using five parameters: bar charts and probit-diagrams built on the basis of the remainders; mean relative errors of approximation (ε_i); coefficient of determination (R_1^2); Fisher criterion (F_i) and criterion of statistics (F_{ri}).

The value of mean relative error of approximation was calculated by the formula:

$$e_i = \frac{1}{n}\sum_{j=1}^{n}\left|\frac{Y_{ij} - Y_{ij}^{reg}}{Y_{ij}}\right| \qquad (3)$$

where n – sample collection (number of experiments).

To check the adequacy of multiple-factor regressive model Fisher criterion was used. It was calculated by the formula:

$$F_i = \frac{S^2_{reg_i}}{S^2_{resit_i}} \qquad (4)$$

where $S^2_{reg_i}$ – dispersion of experimental response functions relative to their mean values; $S^2_{resit_i}$ – residual dispersion of response functions.

$$S^2_{reg} = \frac{1}{n-1}\sum_{n-1}^{n}(Y_{ij} - \overline{Y}_i)^2 \qquad (5)$$

where $\overline{Y}_i$ – average experimental value of response function.

$$S^2_{resit} = \frac{1}{n-m_i}\sum_{j-1}^{n}(Y_{ij}^{reg} - \overline{Y}_{ij})^2 \qquad (6)$$

where m_i –number of coefficients in the regression equation.

In accordance with the mentioned calculations Fisher criterion should be greater than the table value at significance level α and freeness (n-1) and (n-m_i). In this case it shows the number of times the results scattering relative to the line of obtained regression equation changes as compared to the scattering relative to a mean value [13].

Coefficient of determination characterizing the significance of response functions dependence on process parameters has values from 0 to 1. It was determined using the standard procedures [14].

It is proved that F_{ri} may be expressed by means of coefficient of determination [15]. The criterion of statistics, which is a measure of regressive model adequacy, is also a measure of statistic significance R_i^2. Therefore, to check the significance of the coefficient of determination F_{ri} was used. For this purpose zero ($R_i^2 = 0$) and alternative ($R_i^2 \neq 0$) hypotheses were stated. To check the zero hypothesis the criterion of statistics was calculated. It is connected with the coefficient of determination by the following ratio:

$$F_{r_i} = \frac{n - k_i - 1}{k_i} \cdot \frac{R_i^2}{1 - R_i^2} \qquad (7)$$

and has Fisher distribution with freenesses k_i and (n-k_i-1), where k_i – number of regression equation coefficients without a free term.

The calculated value F_{ri} was compared with the critical value F_{rcri} determined by means of tables at the level of significance a and numbers of freeness k_i and (n-k_i-1). If $F_{ri} \le F_{rcri}$, then the zero hypothesis is accepted, *i.e.* regression equation is statistically insignificant. If $F_{ri} > F_{rcri}$, the zero hypothesis is rejected, *i.e.* the alternative hypothesis about statistical significance of regression equation is accepted.

On the basis of regression equations optimal conditions were determined using the method of uniform search (minimum or maximum) of response functions [16] for three types of coal: low-metamorphized (type D) high-metamorphized (type P) and ordinary (type R).

Agreement between the results obtained under optimal conditions and predicted results based on the experimental-statistic model was checked using value of mean relative error of approximation. It is calculated by the following formula:

$$e_i' = \frac{1}{n'} \sum_{j=1}^{n'} \left| \frac{Y_{ij}' - Y_{ij}^{predict}}{Y_{ij}^{predict}} \right| \qquad (8)$$

where Y_{ij}' – observed values of response functions obtained during the experiments under optimal conditions; $Y_{ij}^{predict}$ – predicted values of response functions under optimal conditions determined by regression equations; n' – numbers of experiments under optimal conditions.

3. Results and Discussion

The initial results of the experiments, which were basis for the development of the experimental-statistic model, are represented in Table 2. For both response functions (Y_1 and Y_2) the non-linear multiple regression Eqs. (9) and (10) were formed using STATISTICA software.

$$\begin{aligned} Y_1 = {} & -1126.46 - 0.003406 \cdot X_1^2 - 0.004479 \cdot X_2^2 - 0.003291 \cdot X_3^2 - 0.771913 \cdot X_4^2 + 0.551132 \cdot X_5^2 - \\ & - 0.005835 \cdot X_1 \cdot X_2 - 0.688584 \cdot X_3 \cdot X_4 - 0.177708 \cdot X_3 \cdot X_5 - 0.011040 \cdot X_1 \cdot X_3 + 0.017956 \cdot X_1 \cdot X_4 - \\ & - 0.059971 \cdot X_1 \cdot X_5 - 0.005145 \cdot X_2 \cdot X_3 + 0.079830 \cdot X_2 \cdot X_4 - 0.002756 \cdot X_2 \cdot X_5 - 0.188538 \cdot X_4 \cdot X_5 + 4.080821 \cdot X_1 + \\ & + 3.103388 \cdot X_2 + 7.188298 \cdot X_3 + 20.00892 \cdot X_4 + 25.94932 \cdot X_5 \end{aligned} \qquad (9)$$

$$\begin{aligned} Y_2 = {} & -15.9544 - 0.000019 \cdot X_1^2 + 0.002910 \cdot X_2^2 + 0.000600 \cdot X_3^2 + 0.223833 \cdot X_4^2 - 0.000631 \cdot X_1 \cdot X_2 - \\ & - 0.054981 \cdot X_3 \cdot X_4 + 0.000763 \cdot X_3 \cdot X_5 - 0.0000299 \cdot X_1 \cdot X_3 - 0.000514 \cdot X_1 \cdot X_4 + 0.001466 \cdot X_1 \cdot X_5 - \\ & - 0.000941 \cdot X_2 \cdot X_3 + 0.019109 \cdot X_2 \cdot X_4 - 0.019989 \cdot X_2 \cdot X_5 - 0.292095 \cdot X_4 \cdot X_5 + 0.029878 \cdot X_1 + 0.043360 \cdot X_2 + \\ & + 0.278156 \cdot X_3 + 0.367353 \cdot X_4 + 1.408508 \cdot X_5 \end{aligned} \qquad (10)$$

The calculated values of response functions (Y_1^{reg} i Y_2^{reg}) and relative errors (ε_1 i ε_2) are also given in Table 2.

Table 2

Experimental data, calculated values of response functions and relative errors

X_1	X_2	X_3	X_4	X_5	Y_1	Y_1^{reg}	Y_2	Y_2^{reg}	Coal type	Relative errors	
										ε_1	ε_2
1	2	3	4	5	6	7	8	9	10	11	12
400	60	30	2.400	2.34	69.60	78.23	0.57	1.35	C	0.1240	1.3737
425	60	30	2.400	2.34	99.25	90.53	0.97	1.38	C	0.0879	0.4239
450	60	30	2.400	2.34	99.34	98.57	1.16	1.43	C	0.0078	0.2357
400	15	30	2.400	2.34	58.90	57.35	2.06	2.25	C	0.0264	0.0939

Continuation of Table 2

1	2	3	4	5	6	7	8	9	10	11	12
400	30	30	2.400	2.34	67.44	66.32	1.12	0.64	C	0.0165	0.4250
425	15	30	2.400	2.34	91.51	76.21	3.25	2.99	C	0.1672	0.0795
425	30	30	2.400	2.34	91.73	83.00	1.62	1.15	C	0.0952	0.2929
450	10	30	2.400	2.34	84.32	88.83	4.54	4.74	C	0.0535	0.0439
450	15	30	2.400	2.34	96.65	90.82	3.70	3.75	C	0.0604	0.0146
450	30	30	2.400	2.34	98.42	95.42	1.99	1.67	C	0.0305	0.1602
450	45	30	2.400	2.34	99.20	98.00	1.34	0.90	C	0.0121	0.3302
450	30	30	1.575	2.34	83.59	90.20	2.46	2.28	C	0.0791	0.0745
450	30	30	1.950	2.34	95.96	92.70	2.09	1.96	C	0.0339	0.0605
450	30	30	2.775	2.34	99.00	97.44	1.66	1.50	C	0.0158	0.0984
425	20	4	2.400	2.34	95.41	73.15	2.04	1.65	C	0.2333	0.1907
425	20	15	2.400	2.34	95.80	76.04	2.27	1.80	C	0.2062	0.2083
425	20	30	2.400	2.34	93.99	78.70	2.74	2.23	C	0.1627	0.1858
425	20	50	2.400	2.34	87.47	79.93	3.59	3.23	C	0.0862	0.1006
425	20	70	2.400	2.34	72.37	78.54	5.72	4.71	C	0.0852	0.1770
450	30	4.5	2.400	2.34	99.09	98.37	1.44	1.53	C	0.0073	0.0594
450	30	50	2.400	2.34	96.13	90.10	2.57	2.33	C	0.0627	0.0928
450	30	70	2.400	2.34	85.06	82.16	3.68	3.47	C	0.0341	0.0566
450	30	30	2.400	2.08	99.32	96.58	1.93	1.47	C	0.0276	0.2406
400	10	4.5	1.600	7.20	52.92	52.67	9.72	8.29	G1	0.0047	0.1471
400	15	4.5	1.600	7.20	62.91	56.38	6.62	7.02	G1	0.1038	0.0605
400	30	4.5	1.600	7.20	69.77	66.17	3.83	4.08	G1	0.0517	0.0664
400	60	4.5	1.600	7.20	76.43	79.69	2.11	2.14	G1	0.0427	0.0142
400	15	30	2.400	7.20	56.28	64.12	6.32	7.20	G1	0.1393	0.1387
400	30	30	2.400	7.20	66.88	72.90	3.71	4.13	G1	0.0900	0.1132
425	15	30	2.400	7.20	76.80	75.70	8.60	8.11	G1	0.0144	0.0566
425	30	30	2.400	7.20	85.62	82.29	4.69	4.81	G1	0.0389	0.0255
450	15	30	2.400	7.20	81.88	83.02	9.17	9.05	G1	0.0139	0.0127
450	30	30	2.400	7.20	93.13	87.42	5.17	5.51	G1	0.0614	0.0664
400	30	4.5	2.000	7.20	75.40	75.11	3.41	3.76	G1	0.0039	0.1027
400	30	4.5	2.800	7.20	79.42	92.25	2.57	3.33	G1	0.1615	0.2947
425	15	4	2.400	7.20	92.25	91.94	7.60	7.31	G1	0.0034	0.0376
425	15	15	2.400	7.20	94.02	85.61	8.92	7.55	G1	0.0894	0.1532
425	15	50	2.400	7.20	65.35	60.17	10.74	9.28	G1	0.0792	0.1360
425	15	70	2.400	7.20	47.41	42.02	12.49	10.93	G1	0.1137	0.1252
400	60	4.5	2.400	7.20	96.18	99.24	1.88	2.02	G1	0.0318	0.0757
400	60	15	2.400	7.20	99.07	93.65	2.25	1.89	G1	0.0547	0.1611
400	60	30	2.400	7.20	94.67	84.40	2.67	1.92	G1	0.1085	0.2793
400	60	40	2.400	7.20	83.07	77.41	2.60	2.10	G1	0.0681	0.1927
400	60	50	2.400	7.20	73.71	69.77	2.54	2.39	G1	0.0535	0.0576
400	60	70	2.400	7.20	42.32	52.50	2.35	3.34	G1	0.2406	0.4226
400	60	30	2.400	6.23	93.94	81.09	2.35	1.81	G1	0.1368	0.2296
430	30	50	1.600	7.20	73.82	72.48	8.10	8.39	G1	0.0181	0.0361
400	30	20	2.400	7.20	83.93	77.68	4.01	3.79	G1	0.0744	0.0541
430	60	20	1.600	7.20	87.70	88.10	3.06	3.02	G1	0.0046	0.0125
400	60	30	2.400	6.05	76.42	80.59	1.83	1.79	G2	0.0546	0.0223
425	60	30	2.400	6.05	90.23	87.33	2.11	1.95	G2	0.0322	0.0742
450	60	30	2.400	6.05	95.20	89.80	2.29	2.14	G2	0.0567	0.0648
425	15	30	2.400	6.05	54.53	73.47	5.02	6.90	G2	0.3473	0.3747
425	30	30	2.400	6.05	58.90	80.10	2.77	3.94	G2	0.3600	0.4233
425	45	30	2.400	6.05	82.36	84.72	2.55	2.29	G2	0.0287	0.1007
425	60	30	2.000	6.05	64.84	84.43	1.87	2.41	G2	0.3022	0.2876

Continuation of Table 2

1	2	3	4	5	6	7	8	9	10	11	12
425	60	30	2.800	6.05	95.98	89.97	1.97	1.57	G2	0.0626	0.2028
425	60	30	3.200	6.05	93.89	92.37	1.71	1.26	G2	0.0162	0.2637
425	60	30	2.400	4.84	76.94	86.70	1.25	1.77	G2	0.1269	0.4134
425	15	30	2.400	3.81	78.05	73.31	4.81	4.54	L	0.0607	0.0560
425	15	30	2.400	3.81	79.33	73.31	4.82	4.54	L	0.0759	0.0579
425	15	30	2.400	3.81	80.01	73.31	4.49	4.54	L	0.0838	0.0113
400	15	30	2.400	3.81	44.52	56.65	2.90	3.75	L	0.2724	0.2926
450	15	30	2.400	3.81	80.66	85.71	4.78	5.36	L	0.0626	0.1207
425	15	30	1.800	3.81	54.13	70.78	4.23	5.37	L	0.3075	0.2702
425	15	30	2.057	3.81	66.41	71.93	4.44	5.00	L	0.0831	0.1254
425	15	30	2.880	3.81	78.86	74.93	4.16	3.99	L	0.0498	0.0406
425	15	30	3.600	3.81	82.16	76.71	3.82	3.36	L	0.0664	0.1204
425	15	30	4.800	3.81	83.10	77.88	2.72	2.82	L	0.0628	0.0382
425	15	4	2.400	3.81	74.18	73.89	3.19	3.81	L	0.0039	0.1941
425	15	15	2.400	3.81	74.65	74.19	3.92	4.02	L	0.0062	0.0254
425	15	50	2.400	3.81	69.45	69.83	5.01	5.66	L	0.0055	0.1288
425	15	70	2.400	3.81	68.29	63.73	4.59	7.25	L	0.0668	0.5796
400	10	30	3.600	3.81	53.42	55.61	3.24	3.44	L	0.0410	0.0627
400	20	30	3.600	3.81	63.14	63.18	1.88	1.87	L	0.0007	0.0058
400	30	30	3.600	3.81	69.50	69.86	1.39	0.88	L	0.0052	0.3690
425	10	30	3.600	3.81	78.50	73.54	4.77	4.30	L	0.0632	0.0988
425	20	30	3.600	3.81	84.26	79.65	2.80	2.57	L	0.0547	0.0833
425	25	30	3.600	3.81	84.91	82.37	1.15	1.92	L	0.0299	0.6689
425	30	30	3.600	3.81	86.23	84.87	0.86	1.42	L	0.0158	0.6477
425	40	30	3.600	3.81	86.64	89.19	0.80	0.85	L	0.0295	0.0614
425	50	30	3.600	3.81	86.12	92.62	0.61	0.86	L	0.0755	0.4150
450	10	30	3.600	3.81	81.29	87.21	5.15	5.18	L	0.0728	0.0055
450	20	30	3.600	3.81	85.05	91.86	2.65	3.29	L	0.0801	0.2410
450	30	30	3.600	3.81	86.42	95.62	1.80	1.98	L	0.1065	0.1006
425	15	30	3.600	2.66	74.68	79.03	3.55	2.55	L	0.0583	0.2813
425	20	4	2.400	1.41	54.02	71.94	0.72	0.78	A1	0.3317	0.0866
425	20	15	2.400	1.41	58.97	76.64	1.01	0.92	A1	0.2997	0.0882
425	20	30	2.400	1.41	65.29	81.78	1.35	1.34	A1	0.2525	0.0046
425	20	50	2.400	1.41	80.39	86.32	2.27	2.33	A1	0.0737	0.0255
425	20	70	2.400	1.41	76.71	88.22	3.42	3.79	A1	0.1501	0.1087
400	30	62.5	1.800	1.94	99.01	87.46	4.82	3.55	A2	0.1167	0.2640
400	30	62.5	1.800	1.86	89.55	88.05	3.45	3.47	A2	0.0167	0.0069
Mean relative error of approximation (ε)										0.0859	0.1721

The nature of bar charts (Figs. 1 and 2) and probit-diagrams (Figs. 3 and 4) built from of the calculated values remainders indicates the first obligatory feature of the obtained equations adequacy, since the remainders obey the law of normal distribution with the center in the point of coordinates origin, *i.e.* the main part of the remainders $\Delta Y_{ij} = Y_{ij}^{reg} - Y_{ij}$, is around zero.

The mean relative errors of approximation calculated by (3), $\varepsilon_1 = 0.0859$ (8.59 %), $\varepsilon_2 = 0.1721$ (17.21 %) also indicate the agreement between the developed model and the experimental data because it is admitted according to [17] that at $\varepsilon = 0$–10 the prognosis accuracy is high, at $\varepsilon = 10$–20 % it is good and at $\varepsilon = 20$–50 % – satisfactory.

The calculated values of Fisher criterion (accordingly to (4)) are: $F_1 = 2.348$; $F_2 = 10.143$. In accordance with the table of critical values of Fisher criterion [18] at the level of significance $a = 0.05$ the critical values are: $F_{1kr} = F(0.05; 92; 72) = 1.452$ and $F_{2kr} = F(0.05; 92; 73) =$ $= 1.450$. They are lower than the calculated values, which also confirm the adequacy of the model.

Coefficients of determination $R_1^2 = 0.6667$ and $R_2^2 = 0.9218$ indicate that 66.67 % and 92.18 % of the changes in response functions Y_1 and Y_2 respectively are defined by the change of process parameters X_1–X_5. The fact that $R_1 = 0.82$ and $R_2 = 0.96$ are close to 1, indicates the presence of “strong” bond between Y_1 and Y_2 and process parameters X_1–X_5.

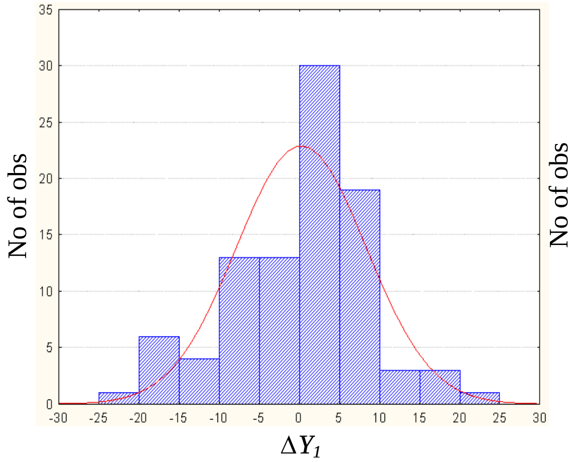

Fig. 1. Frequency distribution: residuals ΔY_1

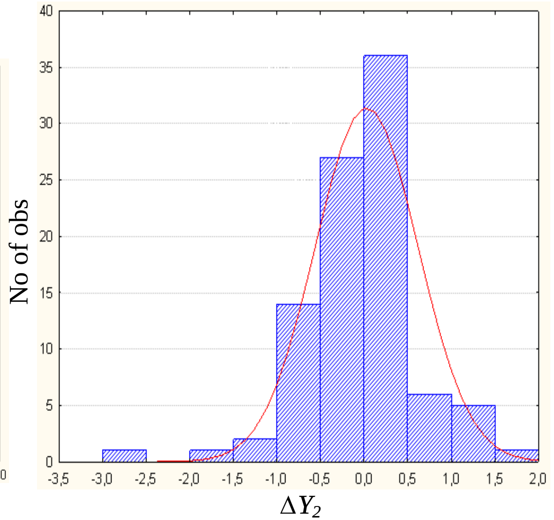

Fig. 2. Frequency distribution: residuals ΔY_2

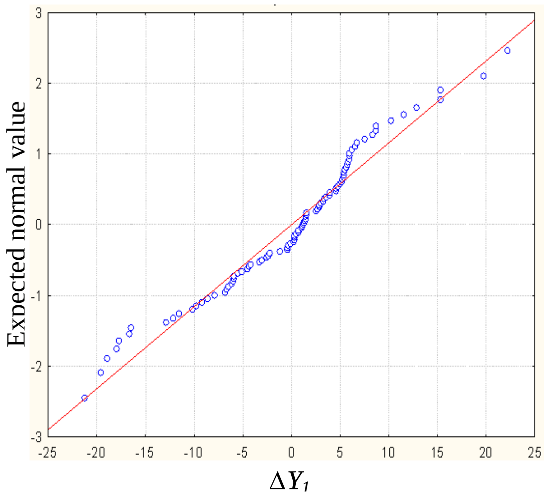

Fig. 3. Normal probability plot of residuals

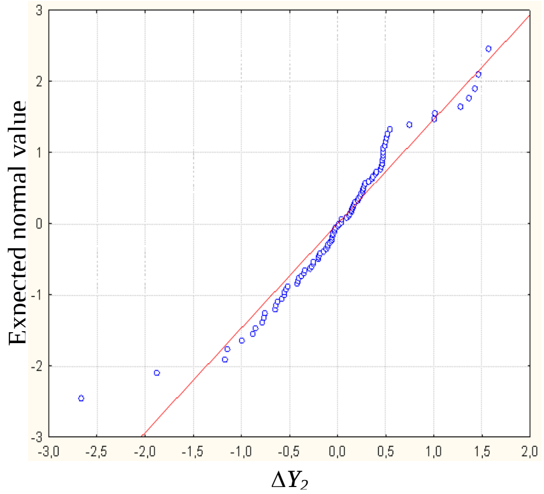

Fig. 4. Normal probability plot of residuals

Table 3

Optimal conditions for desulphurization and obtained results

Coal type	Parameter values				Sulphur content for analytical mass, mass %				Values of response functions			
	Temperature, K	Time, min	RFO, $m^3/(h \cdot kg)$	Content of water steam in the oxidant, vol %	total, S^a_t	pyritic, S^a_p	sulphate, $S^a_{SO_4}$	organic, S^a_o	Fractional conversion of pyrite sulphur, %		SO_2 content, vol %	
									Y'_1	$Y_1^{predict}$	Y'_1	$Y_1^{predict}$
Calculated values												
C	691	19.5	1.72	64.5	–	–	–	–	–	90.00	–	6.08
L	719	15.0	3.06	25.5	–	–	–	–	–	90.00	–	4.45
R	706	10.5	2.24	69.0	–	–	–	–	–	90.00	–	6.16
Experimental values												
C	693	19.5	1.72	65.0	1.36	0.40	0.48	0.48	84.82	–	6.96	–
L	718	15.0	3.06	25.0	1.11	0.38	0.15	0.58	90.84	–	3.80	–
R	708	10.5	2.24	70.0	0.59	0.26	0.12	0.21	85.42	–	5.60	–

The calculated values of the criterion of statistics (according to (7)) are: $F_{r1} = 7.20$; $F_{r2} = 45.16$. In accordance with the table of critical values of Fisher criterion at the level of significance $a = 0.05$ the critical values are: $F_{rkr1} = F(0.05;\ 20;\ 72) = 1.718$ and $F_{rkr2} = F(0.05;\ 19;\ 73) = 1.731$, which indicates the statistical significance of the determination coefficients.

Taking into account that CSD maximum values of 3–8 vol % were obtained at FCPS 80–90 % (see Table 2), the optimization task was to find such process parameters which ensure FCPS maximum under the following limitations: $Y_1 \leq 90$, $3 \leq Y_2 \leq 8$. Predicted results and results obtained under optimal conditions are represented in Table 3.Mean relative errors of predicted data approximation were calculated by formula (8): $\varepsilon'_1 = 0.0413$ (4.13 %), $\varepsilon'_2 = 0.1324$ (13.24 %). They indicate the agreement between predicted and practical data.

After desulphurization of the three coal samples we obtain fuel with sulphur content of 1.0–1.5 mas %. It may be considered as the low-sulphuric one. At the same time fractional conversion of total sulphur (FCTS) is 69.63–77.27 %. If we assume that all sulphur dioxide contained in the desulphurization gases is recovered or converted using the known methods [5] then FCTS will be equal to the degree of environmental pollution reduction. The main pollutant is sulphuric anhydride. Hence, we may state that the proposed method gives the possibility to burn sulphuric and high-sulphuric coal, reduce sulphuric emissions by 70–80 %, and utilize free sulphur.

4. Conclusions

1. The experimental-statistic model of coal oxidative desulphurization was developed. It adequately describes the dependence of the main response functions on the process parameters and the quality of the raw material.
2. On the basis of the proposed model the optimal conditions were determined for the desulphurization of the three samples of power-generating coal. The process proceeding under optimal conditions confirmed the agreement between the experimental data and the predicted results.
3. Low-sulphuric coal was obtained (sulphur content was 1.0–1.5 mas %) and fractional conversion of total sulphur (the degree of SO_2 reduction) was 70–80 %.
4. The content of sulphur dioxide in the desulphurization gases is at the level of 3.8–7.0 vol %. Such value allows to state that it is possible to obtain liquefied SO_2 from the gases using the known methods.

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ОПТИМІЗАЦІЯ ПРОЦЕСУ ОКСИДАЦІЙНОГО ЗНЕСІРЧУВАННЯ КАМ'ЯНОГО ЕНЕРГЕТИЧНОГО ВУГІЛЛЯ

***Анотація**. Створено експериментально-статистичну модель процесу оксидаційного знесірчування кам'яного енергетичного вугілля та доведено її адекватність. З використанням запропонованої моделі знайдено оптимальні умови проведення знесірчування трьох зразків кам'яного енергетичного вугілля. Проведення процесу за цих оптимальних умов дозволило отримати низькосірчисте тверде паливо (вміст загальної сірки не перевищував 1.0-1.5 % мас.).*

***Ключові слова:** оксидаційне знесірчування, вугілля, сірка, пірит, моделювання.*