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IMPROVING THE TECHNOLOGY OF PREPARING COAL FOR THE PRODUCTION OF BLAST-FURNACE COKE UNDER THE CONDITIONS OF MULTI-BASIN RAW MATE-RIAL BASE. MESSAGE 2. OPTIMIZING THE DEGREE OF CRUSHING BY MEANS OF PETROGRAPHIC CHARACTERISTICS OF THE BATCH COMPONENTS

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Abstract

Our research in the coal-preparation shop of the coke plant at ArcelorMittal Krivoi Rog shows that coal crushing is not only associated with the distribution of the macerals over the size classes but also with change in composition of the coal. Thus, with increase in the degree of crushing from 55.5 to 96.2%, the vitrinite content in the batch declines from 70 to 63%, while the total content of fusinized components increases from 29 to 35%. Therefore, the change in microcomponent composition on intensity of grinding the coking batch must be studied in order to explain the coking process and to optimize batch preparation.

Keywords: coal; crushing; maceral composition; vitrinite; fusinized components.

1. Introduction

Before coking, the coal batch is crushed so as to ensure the required degree of crushing (in terms of the content of the primary < 3 mm class), the required content of the auxiliary < 0.5 mm class, the optimal granulometric composition, and the maximum packing density of the coal batch. That improves the clinkering and coking properties of the coal and ultimately the quality of the coke. In addition, the consistency of the coke properties depends on the mixing of the coal batch after crushing [1–6].

At the same time, the crushing of coal does not simply change the coal's grain size. Coke quality depends on the monitoring and adjustment of the distribution of the petrographic components over the size classes and the changes in maceral composition as a result of crushing. That will also affect the clinkering and coking properties of the coal.

Consequently, the literature provides considerable practical information on those topics. As yet, however, we lack theoretical principles explaining the decrease in vitrinite content (Vt) with an increase in the degree of crushing and the increase in the total content of lean (fusinized) components (ΣFC).

2. Results and discussions

The petrographic analysis is essential to the production of high-quality coke when using petrographically nonuniform coal and in expanding the enrichment of coals of different rank. That entails the introduction of characteristics based on the petrographic analysis in the theory and practice of coking-batch crushing.

As an example, we may consider the petrographic assessment of the technological value of coking batch on the basis of characteristics developed at the Ukrainian Coal-Chemistry Institute (UCCI)^[7-8].

Thus, the quantity of vitrinite components corresponding to intermediate metamorphic stages, as recalculated for the coal's organic mass, is expressed by the clinkering characteristic $C_{\rm b}$ of the batch $^{[7-8]}$

(1)

$$C_b = \frac{\sum (0,90 \div 1,39) \cdot V_t}{100}$$

where $\Sigma(0.90-1.39)$ is the content of vitrinite components with reflectance in the range 0.9-1.39%; Vt is the concentration of vitrinite-group macerals, %.

A second coking characteristic K_b of the batch is calculated from the formula [7-8]

$$K_{b} = \frac{\left(\Sigma(0,90 \div 1,39) \cdot Vt/100\right) + L}{\Sigma FC + \Sigma(\ge 1,70) \cdot Vt/100}.$$
(2)

where $\Sigma(0.90-1.39)$ is the content of vitrinite components with reflectance in the range 0.9– 1.39%; Vt is the concentration of vitrinite-group macerals, %; L is the concentration of liptinite-group macerals, %; ΣFC is the sum of fusinized macerals (I + 2/3Sv), %; $\Sigma(>1.70)$ Vt/100 is the content of vitrinite components with reflectance of 1.70% or more.

On the basis of C_b and K_b , we may obtain a general quantitative estimate of the batch's petrographic characteristics, including the maceral composition and the results of reflectogram analysis. With the increase in C_b , the strength of the coke produced will be higher. In turn, with an increase in K_b , the probability of wear-resistant and relatively uncrushable coke will increase, other conditions being equal ^[7].

In order to improve the preparation of coal batch for coking, we must optimize the degree of crushing (the content of the < 3 mm class), taking account of the petrographic characteristics of the batch components.

On the basis of petrographic analysis and the UCCI method we recommend determine the optimal size class of the batch (%) as a function of its actual rank composition ^[9]

$$k = \frac{75 \cdot Vt \cdot \frac{\sum(0,9-1,39)}{100} + 90 \cdot Vt \cdot \frac{\sum(0,5-0,89) + \sum(1,4-2,6)}{100} + 90 \cdot \sum FC}{100},$$
 (3)

where $\Sigma(0.5-2.6)$ is the content of the vitrinite components with reflectance in the range 0.5-2.6%, corresponding to different coal ranks; 75 is the recommended degree of crushing of Zh and K coal, %; 90 is the recommended degree of crushing of G, OS, and T coal, %; ΣFC – sum of fusinized components (I + 2/3Sv), %.

Thus, the correction of the degree of coal blends crushing permitting the maintenance of coke quality consistent with blast-furnace requirements (minimum strength M_{25} =90%, M_{10} =6%).

The next step in our research is to consider the changes in maceral composition as a result of crushing.

The coking properties of the coal batch (especially in the case of petrographically nonuniform coal) depend considerably on the distribution of the coal components over the size class, as noted in ^[10]. This is especially important in developing methods for batch preparation and in selecting the optimal degree of crushing. The clinkering properties of each coal grain depend on its petrographic composition and size. For the same batch, the clinkering properties will improve when the distribution of the coal over the size classes is modified by the selective crushing of the batch, instead of the traditional methods (undifferentiated crushing of the whole batch or separate crushing of each component). In the case of undifferentiated crushing, increase in the degree of crushing always results in significant differences between the large and small classes in terms of their properties, on account of the expulsion of the relatively soft vitrinite from the coal grains: the ash content of the large classes increases, while their clinkering properties are degraded.

Without a change in the composition of the batch, its coking properties may be considerably improved by establishing a more uniform distribution of its components over the size classes,

as noted in ^[11]. This is possible by the appropriate crushing of the coal batch, group crushing of the components, and careful grouping of the available coal.

The change in micro-component composition on crushing the coking batch must be studied in order to explain the coking process and to optimize batch preparation. The group that is most difficult to crush is the liptinites, which are concentrated in large classes. Vitrinites, conversely, are more brittle and concentrated in smaller classes. The microcomponent distribution also depends on the level of metamorphic development. The crushing of vitrinite is easiest for coal of moderate metamorphic development (coke coal). The resistance to crushing is greatest for vitrinite in long-flame coal and anthracite ^[12]. Coal with 50–80% vitrinite (with the same R_o) is less strong than coal with a smaller vitrinite content, as noted in ^[13].

At the same time, mineral impurities such as calcite and pyrite may increase the strength of the coal if they are present in the organic mass—in other words, if they are syngenetic. (They fill the pores of fusinite, telinite, etc.) If calcite and pyrite are found in cracks (if they are epigenetic), the strength of the coal is not increased ^[14]. Research shows that the <4 and >4 mm classes have different petrographic composition. For instance, the vitrinite content is greater in the <4 mm class than in the >4 mm class ^[15].

Many studies also indicate that coal crushing is not only associated with the distribution of the macerals over the size classes but also with the change in the composition of the coal.

The change in the composition of gas (G) coal with an increase in the content of the < 3 mm class from 35 to 91% was studied in ^[16]. With 35.4% of the < 3 mm class, the vitrinite content is 76.9%; with 57.1% of the < 3 mm class, the vitrinite content is only 72.1%. With 90.8% of the < 3 mm class, the vitrinite content falls to 64.9%. Thus, the vitrinite content is 12% different for 35.4% and 90.8% crushing. The liptinite content declines correspondingly from an initial value of 13.3% to 14.2, 17.3, and 22.7%, respectively.

The change in the petrographic composition of the batch is accompanied by deterioration of the clinkering properties. For example, the plastic-layer thickness y is 17 mm when the content of the <3 mm class is 35.4% and 12% when the degree of crushing is 90.8%. The Rog index declines from 57.7 to 38.2.

Research in the coal-preparation shop of the coke plant at ArcelorMittal Krivoi Rog also shows that the petrographic composition of the batch changes on crushing. Table 1 presents the change in the vitrinite content and the total content of fusinized components before and after crushing of the batch sent to coke batteries 1-6.

It follows from Table 1 that crushing of the batch changes its composition. However, the change in the vitrinite content on crushing may be attributed to the errors of the method. According to State Standard GOST 9414.3-93 (the ISO 7403-3-84 standard), the consistency of the method is 5.1% when the vitrinite content in the sample is 80%.

	Vitrinite Vt, %		∑FC	, %	
Sample	before	after	before	after	
	crushing	crushing	crushing	crushing	
		Batteries 1-3			
1	75	70	23	27	
2	75	68	23	30	
3	74	72	24	27	
4	74	64	24	32	
5	74	65	24	32	
Battery 4					
1	75	68	23	29	
2	74	69	24	28	
3	74	62	24	36	
4	74	66	24	31	
5	74	63	24	34	
		Batteries 5-6			

Table 1. Petrographic indices of coal batch

1	75	68	23	30
2	75	65	23	33
3	75	64	23	33
4	74	63	23	34
5	74	73	24	25
6	75	67	23	31

At the same time, it is evident from these results and from literature data that this error is exceeded in many cases; values of 7-12% may be obtained.

To confirm the change in the petrographic composition of coking batch on crushing, we also conduct a direct experiment. Batch is taken from the crushers in the coal-preparation shop, carefully homogenized, and split into thirds. One part is used as the uncrushed baseline, with 55.5% of the < 3 mm class; the other two parts are passed through a laboratory mill to obtain 75.5 and 96.2% of the <3 mm class, respectively (Table 2).

Table 2. Granulometric composition of batch

Commis	Granulometric composition of batch by size class, (mm)				
Sample	>6	6-3	3-0,5	≤0,5	≤3 mm
1	32,8	11,7	27,1	28,4	55,5
2	5,0	19,5	43,8	32,7	75,5
3	0,2	3,6	52,9	43,3	96,2

Table 3 present the petrographic composition of the three samples. The experiment indicates that, with an increase in the degree of crushing from 55.5 to 96.2%, the vitrinite content falls from 70 to 63% ($\Delta = 7\%$), while the content of fusinized components increases from 29 to 35%, with deterioration in the clinkering and coking properties of the coal batch.

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		Sample	
Characteristic	1	2	3
Content of 0-3 mm class, %	55,5	75,5	96,2
Vitrinit Vt, %	70	66	63
ΣFC, %	29	33	35

Many such examples may be presented, but what is the underlying mechanism? We know from the literature that the disintegration of coal depends on its microstructure and molecular structure. Note that the coal fractures along the boundary between microlayers of lithotypes, so that the coal is divided between the vitrinite group and the inertinite group [17-18].

The crushing of coal not only reduces the particle size and increases the specific surface but also changes its structure, as shown in ^[19]. In addition, the molecular interactions are disrupted, and chemical bonds in the organic macromolecules will be broken ^[20].

In crushing, coal does not simply disintegrate but becomes chemical active, as noted in ^[21]. Changes in the pore structure facilitate oxidation. Quantitative data for peroxides in the fractions of G coal indicate that, for technological applications, coal should not be crushed beyond 0.5 mm, because smaller particles are much more rapidly oxidized ^[22].

In crushing, the molecular structure of the coal is broken down ^[18]. In particular, the length of the chemical chains is reduced, while the compounds become more aromatic. The main structural changes due to stress occur not only in the framework of molecules and macromolecules but also at their periphery—that is, in the space between and above the molecules. Note the difference in strength not only of carbon-hydrogen bonds but also of carbon-carbon and carbon-oxygen bonds. The structural strength of the micro-components in the coal will depend on the number of such bonds in the molecules and macromolecules and on their relative positions.

The relation between the degree of crushing and the structural changes (in terms of the atomic ratio C/H) was established in ^[19]. For example, on crushing coal, C/H tends to decline in the early stages; later on, it increases somewhat. On crushing, the interplane distance increases, while the height of the stacks of carbon layers declines. This indicates disruption of the coal structure such that disordered carbon appears

We may speak of a vitrinite macromolecule with a specific spatial position. It consists of repeating structural units, corresponding to a set of condensed aromatic nuclei with different numbers of rings (mainly 2-8 rings)^[23].

Analysis of the IR and NMR spectra for the crushing products of G coal indicates destruction of the arylalkyl ester bonds and the ester bonds within the aliphatic structures, together with rupture of the ester bonds with the aromatic rings and the formation of phenol groups ^[24].

3. Conclusions

In order to improve the clinkering and coking properties of the coal and ultimately the quality of the coke, we must optimize the degree of crushing (the content of the < 3 mm class), taking account of the petrographic characteristics of the batch components.

Experimental data from many sources indicate that, in crushing, the distribution of the petrographic micro-components over the size classes is modified. We may also assume that, with an increase in the degree of crushing, the petrographic composition of the coal batch is changed: the vitrinite content falls.

For coal with 50-85% vitrinite, which is less strong than coal with smaller vitrinite content, crushing is probably accompanied by disruption of the molecular interactions and rupture of the chemical bonds in the organic macromolecules. That leads to the partial destruction of the vitrinite structure, which is brittle, especially in the intermediate stages of metamorphism ($R_0 = 0.9-1.39\%$).

Further study of this topic is required. More precise and comprehensive experiments must be conducted in order to explain the influence of crushing coal composition. Thus, the theory and technology of coal crushing still require the focused attention of coking specialists.

Symbols

Vt – vitrinite, %; Sv – semivitrinite, %; I – inetinite, %; L – liptinite, %; ΣFC – sum of fusinized components, %; Ro – mean vitrinite reflection coefficient, %; M10, M25 – indices of resistance of coke abrasion and crushability, respectively, %;

References

- [1] Lyalyuk VP, Uchitel' AD, Lyakhova IA, Kassim DA, and Zaitsev GL. Preparation of coking batch. Coke and Chemistry, 2011; 54(8). 271–286.
- [2] Lyalyuk VP, Sokolova VP, Lyakhova IA, and Kassim DA. Ensuring stable quality of blast-furnace coke. Coke and Chemistry, 2012; 55(8): 304–308.
- [3] Lyalyuk VP, Kassim DA, Lyakhova IA, and Sokolova VP. Coke quality and optimization of batch composition. Coke and Chemistry, 2012; 55(12). 448-452.
- [4] Lyalyuk VP, Sokolova VP, Lyakhova IA, and Kassim DA. Quality fluctuations of coking coal. Coke and Chemistry, 2013; 56(1): 1-6.
- [5] Lyalyuk VP, Shmeltser KÖ, Lyakhova IA, and Kassim DA. Changes in granulometric composition of blast-furnace coke. Coke and Chemistry, 2013; 56(12): 456-460.
- [6] Lyalyuk VP, Shmeltser EO, Lyakhova IA, and Kassim DA. Influence of the batch properties and coking technology on the granulometric composition of coke. Coke and Chemistry, 2014; 57(10): 398-404.

- [7] Chernyshov YuA, Ovchinnikova SA, Podlubnyi AV, et al., Assessing the properties of coal and complex batch at OAO Zaporozhkoks on the basis of petrographic characteristics and new coefficients. Uglekhim. Zh., 2009; (1/2): 12-20.
- [8] Spravochnik koksokhimika. T. 1. Ugli dlya koksovaniya. Obogashchenie uglei. Podgotovka uglei k koksovaniyu (Handbook of Coke Chemistry, Vol. 1: Coking Coal, Coal Enrichment and Preparation for Coking), Borisov LN, and Shapoval YuG. Eds., Kharkov: Izd. Dom Inzhek, 2010.
- [9] Lyalyuk VP, Lyakhova IA, Kassim DA, Shmeltser EO. et al., Ukranian Patent 85803, 2013.
- [10] Fomin AP, Konyakhin AP, and Martynyuk II. Specific distribution of coal material by classes of coal charge. Koks Khim., 1981; (8): 2-4.
- [11] Aleksandrova LP, Nagornyi YuS, and Gulyaev VM. Re-distribution of the components of material composition of coals by the size classes. Koks Khim., 1992; (10): 5-6.
- [12] Koshina M, and Maglicheva A. Change of macrocomponent composition during the grinding of coal. Khim. Tverd. Topliva, 1980; (4): 12-18.
- [13] Temeeva LA. Relationship between the coal strength properties with petrographic characteristics. Khim. Tverd. Topliva, 1979; (4): 102-105.
- [14] Bykadorova VI, Matveeva II, and Polferov KYa. Influence of petrographic composition on the grind- ability of coals. Khim. Tverd. Topliva, 1970; (4): 28-33.
- [15] Korobchanskii IE, Kuznetsov MD, Eidel'man EYa. et al., Analysis of selective crushing of some coal of the Donetsk basin. Koks Khim, 1956; (6): 8-13.
- [16] Kuznetsov MD, and Lyannaya ZG. Composition and properties of large coal classes from the Donetsk basin. Koks Khim, 1960; (5): 10-13.
- [17] Mitsenko GP. Microstructure and composition of microlithotypes of coals of working layers of the Donetsk basin. Khim. Tverd. Topliva, 1991, (5): 3-6.
- [18] Ivanov VP. Structural features of coals and their mechanical destruction. Koks Khim., 2008; (9): 24-27.
- [19] Khrenkova TM, Lebedev VV, Goldenko NL, and Golovina GS. Change type of coals during grinding. Khim. Tverd. Topliva, 1975; (1): 11-17.
- [20] Lebedev VV, Khrenkova TM, Chubarova MA. et al., Change of coal properties during dispersion. Khim. Tverd. Topliva, 1977; (6): 11-16.
- [21] Lebedev VV, Golovina GS, and Cheredkova KI. Change of coal porosity during vibratory grinding. Khim. Tverd. Topliva, 1978; (5): 43-44.
- [22] Kamneva AI, Gukha Sh, and Smiryagina NA. Influence of the grinding degree on the formation of peroxide compounds in the air oxidation of coal. Khim. Tverd. Topliva, 1969; (5): 115-116.
- [23] <u>http://studopedia.ru/2 123646 osHoBnie-predstavleniya-o-himicheskoy-strukture-ugley.html</u>.
- [24] Khrenkova TM, and Goldenko NL. Analysis of mechanical destruction products of gas coal used in hydrogenation. Khim. Tverd. Topliva, 1978; (5): 44-45.

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