Changes in the Properties of Coke in Blast Furnace

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Synopsis

Changes in the properties of coke in the blast furnace have been investigated by using the samples taken out of Higashida No. 5 B.F., Hirohata No. 1 B.F. and Kukioka No. 4 B.F., which were water-quenched under the normal operating condition and subsequently dissected. The results obtained by the present work are summarized as follows:

1. The properties of coke are changed considerably at lower levels of the shaft.
2. Microscopic observations reveal that the changes in the properties of coke are probably due to the selective carbon solution reaction with particular coke textures, resulting in the decrease of strength and the generation of fine particles.
3. The above phenomenon is particularly conspicuous around the raceways.

I. Introduction

A comprehensive understanding on the changes in the properties of coke during its descending to the tuyeres in the blast furnace is essential to the stabilization of blast furnace operation, the elucidation of the coke quality requirements for the blast furnace use and to the control of coke plant operation so as to produce coke having adequate properties for these requirements. The blast furnaces under normal operating condition, Higashida No. 5 B.F. (inner volume 646 m³), Hirohata No. 1 B.F. (inner volume 407 m³), and Kukioka No. 4 B.F. (inner volume 1279 m³) were shut down and dissected after water quenching in 1968, 1970 and 1971, respectively. This report presents the results of the study on how the changes of coke in the blast furnaces are influenced by the variation of the coke properties and the blast furnace operating practices.

II. Changes of the Coke Properties in the Blast Furnace

1. Size

As a general trend for three blast furnaces, as shown in Fig. 1, the coke size is reduced gradually as the coke descends from the lower part of the shaft to the bosh, but it is rapidly reduced at the region tuyeres from the bosh to the tuyeres. In the radial direction of the furnace, on the other hand, the change of coke size is considerably influenced by the blast furnace operating practice, and hence it varies with the operating condition. In addition, the size of coke charged seems to have some influence on the size changes even in the vicinity of tuyeres. Since the operating practice adopted and the inner volume are not the same for these three blast furnaces, no simplified comparison can be made, but, in our view, if the coke of lower cold strength is used, the charging of large size coke seems to prevent the degradation of coke to a certain degree.

In the actual blast furnace operation, there is a tendency to adopt large undersized coke. One of the outstanding observations as to the degradation of coke is a highly concentrated distribution of -15 mm fine coke at the tuyeres in the peripheral zone, as shown in Fig. 2. In analyzing this phenomenon as a part of

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Fig. 1. Changes of coke properties in blast furnaces

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the investigation on the changes of coke properties in the blast furnace, it seems to be necessary to study the change in the vicinity of raceway and its effect on the whole furnace condition in further detail.

2. Strength

The DI_{15}^{98} indices of coke for these three blast furnaces are shown in Fig. 1. It indicates distinctly that:

(1) The coke used for Higashida No. 5 B.F. shows a large change of strength in the furnace, though the cold strength before charging is high.

(2) The decreasing rate of coke strength for Hirohata No. 1 B.F. is markedly small as a whole. Generally, the samples of coke before charging shows that the finer the mean coke size is, the higher the DI_{15}^{98} index becomes. The samples taken out of the furnace indicate the same trend as changes of coke before charging in the coke properties. The finer the sample coke is, the lower the DI_{15}^{98} index becomes. From the above observation, it can be concluded that coke is slightly affected by the temperature and gas at about the middle of the shaft. It can also be concluded that the DI_{15}^{98} value is lower as the coke size becomes smaller in the lower part of the shaft and below the shaft because the smaller the coke size is, the larger the surface area affected by the reaction with gas becomes.

On the other hand, the strength of the cell wall of coke (micro-strength) tends generally to increase by the influence of furnace temperature but it is not necessary to consider seriously. Judging, however, from the fact that the micro-strength indices of −15 mm samples are lower than those of lump samples, the surface layer of coke in furnace reduces its cell wall due to the reaction with gas, and, consequently, it becomes brittle. The generation of coke fines resulting from the deteriorated coke surface can be considered to exert an influence on the blast furnace condition.

3. Reactivity

The change of coke reactivity commences at the lower part of the shaft. Generally, when coke is treated at a high temperature, the cell wall is hardened and the reactivity is reduced. As far as the coke samples taken out of the furnaces are concerned, the reactivity becomes higher though it turns out a better quality coke with the progress of graphitization by the thermal history of the furnace and by the hardening of cell wall. The change in the radial direction of the furnace given in Fig. 3 as an example agreed well with that of the alkali distribution to be described later in this report. It is, therefore, logically concluded that the high reactivity of coke in the furnace must be due to the presence of alkali over the coke surface. The changes in the coke properties of these three blast furnaces are shown in Fig. 3 as compared with each other. The degree of change is largest for Higashida No. 5 B.F., and is remarkably small for Hirohata No. 1 B.F. The difference between the two blast furnaces seems to be resulted from the difference in the concentration distribution of alkali, the gas atmosphere and the temperature distribution brought about by different blast furnace operating practices.

In the case of Higashida No. 5 B.F., the highest alkali concentration was found in the temperature range of 1000°C to 1100°C with CO₂ atmosphere. Hence, a considerable coke degradation by CO₂ as

Fig. 2. Distribution of −15 mm coke size in blast furnace (Kukioka No. 4 B.F.)

Fig. 3. Change of coke reactivity in blast furnace (Kukioka No. 4 B.F.)
well as a remarkable decrease in both the size and the strength happens in the furnace. It can, therefore, be understood that the high reactivity of coke sample taken out of the furnace is brought about both by the alkali existing in a large quantity over the coke surface and by the degradation of coke itself.

In the case of Hirohata No. 1 B.F., the highest alkali concentration distribution was found in the high temperature zone with CO atmosphere (over 1200°C). From this, it can be concluded that the decrease of coke strength in the furnace is small and that the reactivity of coke becomes not so high in spite of the adhesion of alkali over the coke surface. The difference observed in the changes of coke size and strength among the three blast furnaces seems to be caused by the phenomenon mentioned above, and therefore, it is necessary to trace the origin of this difference for the clarification of the changes of coke properties in the blast furnace.

4. Chemical Components

1. Ash and Sulfur

Difference in the ash contents of coke among the three blast furnaces is seen from the upper part of the bosh down (Fig. 1), and is probably due to the differences in the reactivities and in the temperature distributions of the furnaces. The difference in the ash contents at the hearth levels of Hirohata No. 1 B.F. and Kukioka No. 4 B.F. is attributable to the final tapping and salamander tapping prior to the quenching carried out for the one and not for the other. In contrast, the sulfur content decreases from the lower part of the shaft and shows no remarkable difference among the three blast furnaces.

2. Alkali

Although the alkali distribution in the lumpy zone is approximately twice as much as that of coke originally charged, it is rapidly increased in the softening-melting zone, but is decreased again below that zone. The reason why it shows such a change may be considered as follows. Since the boiling points of alkalis are ranging from 800°C to 900°C (K: 750°C and Na: 880°C), the volatilization of alkalis by the reduction of their oxides takes place in a reducing atmosphere at the temperature higher than their boiling points according to the following reactions.

\[ \text{K}_2\text{O} + \text{CO} \rightarrow 2\text{Na} + \text{CO}_2 \]
\[ \text{K}_2\text{O} + 3\text{C} + \text{N}_2 \rightarrow 2\text{KCN} + \text{CO} + \text{CO}_2 \]

Hence, the volatilization of alkalis by the reduction of alkali oxides begins at the softening-melting zone and is intensified in the dropping zone which is under that zone and in the high temperature region above the tuyeres transferring to the gas phase. The alkalis highly concentrated in the gas phase are again absorbed by the burden in the softening-melting zone, increasing the alkali concentration in the burden. Then, as the burden descends, a part of alkali is volatilized again. Therefore, the alkali circulates perpetually in the furnace.

The alkali concentration distributions of three blast furnaces are shown in Fig. 4. The distribution shows an inverted-V-shape in the case of Hirohata No. 1 B.F. In the case of Higashida No. 5 B.F., the highest concentration is observed in the peripheral zone, while in Kukioka No. 4 B.F. it is in the intermediate zone forming a W-shape distribution. The above difference in the alkali distributions is considered to be resulted from the difference in the operating practice adopted for each blast furnace. It can therefore be assumed that the difference in the alkali concentration distributions among three blast furnaces is directly and indirectly brought about the difference in the changes of coke properties among three blast furnaces.

Fig. 4. Distribution of alkali concentration of coke in blast furnaces (K$_2$O+Na$_2$O in coke) (%)
III. Changes in the Properties of Coke in the Vicinity of Tuyeres and in the Hearth

1. Condition in the Vicinity of Raceway

A cavity surrounded by lump coke was found in front of most of the tuyeres. This cavity may probably be formed by jostling of coke in the course of shutdown. The change in the coke condition near the raceway, detailed description is given in the former report, seems to be an important factor for the blast furnace operation, judging from the results obtained by the dissection investigations.

2. Size

As shown in Fig. 5, the mean size of +5 mm coke in the hearth is:
1) small or near and below the raceway
2) somewhat larger in the lower part than in the upper part of the central zone (in the range from 1 m above the tuyeres to 2 m below them).

It can be assumed that the mean size distribution of coke in the hearth is maintained during the actual operation because the effect of metal drops on the coke degradation after shut-down is relatively small. Since most of the coke fines generated in the raceway do not remain there, they are probably blown out of the raceway by the blast through the voids among coke lumps, and some of them probably fall down on the raceway and scattered among the coke lumps existing in the lower part of the furnace. As a result, the coke fines are deposited along the periphery of the raceway.

Since it is difficult to ascertain how the coke fines affect on the blast furnace operation, some detailed investigations have been conducted by using an experimental lower-half blast furnace and a model combustion furnace. According to the results obtained:

(1) (as an effect of coke size distribution in the lower part of a blast furnace) COke containing much breeze, the raceway becomes to be in a slugging state, by increasing its internal pressure, and the gas flows through the part close to the furnace wall.

(2) Coke of uniform structure does not readily degrade into breeze. Ordinary blast furnace coke is strongly bonded together in a fused structure which is derived from heavy coking coal, and therefore it is hard to degrade. Coke produced from coking coal is very brittle and generates a great deal of breeze.

The above results indicate that the experiment is well representing the condition near the raceway observed by the dissection. Thus, it is understood that behavior of coke fines near the raceway plays an important role in the blast furnace operation.

3. Strength

The strength of coke obtained by drum test is largely influenced by the deteriorated surface area of sample coke. The strength of coke in the hearth seems to vary widely by the combustion temperature depending on the tuyere diameter, the rate of heavy oil injection, and the retention time in the hearth. Because of the evaporation of silicon and sulfur from coke lumps and the reaction with slag components (SiO₂, P₂O₅, MnO, K₂O, Na₂O, etc.), the internal cell wall of coke is thinned or cut, and therefore the coke strength is reduced. The progress of embrittlement of coke lumps from their surface has thus been traced.

4. Properties of Coke Existing between the Tuyeres

The followings are observed for the properties of coke existing between the tuyeres.

(1) The size of coke existing between the tuyeres is relatively large.

(2) The size of coke existing in front of the tuyeres is relatively small when the diameter of tuyere is small and no heavy oil is injected.

IV. Changes in the Micro-texture of Coke

Coke, macroscopically and microscopically, has a heterogeneous structure and the structure of each component differs in both the physical and chemical characteristics. By clarifying what structures induce the degradation of coke in the blast furnace, positive guidelines can be obtained as to the coke production and the selection of coking coal.

1. Changes in the Coke Texture by Gasification

Coke in the blast furnace is consumed and degraded
mainly by the carbon solution reaction with CO₂. For reference, the followings are given to show the relationships between the kinds of coal and the micro-textures of coke.

Low-fluidity soft coking coal - Isotropic texture
High-fluidity soft coking coal - Fine mosaic texture
Medium-heavy coking coal (Moura) - Coarse mosaic texture
Medium-volatile-matter heavy coking coal - Fibrous texture
Low-volatile-matter heavy coking coal - Leaflet texture
Inerts of all kinds of coal - Inerts

A laboratory scale experiment on the gasification of coke has been conducted in order to obtain the reference data for the investigation on the changes in the microtexture of coke in blast furnace. The experiment was carried out as follows. The sample, 10 mm cube, cut out from the top of a lump coke is placed on a thermobalance, and is gasified by CO₂ to a given weight loss. The sample cut from the middle of the same lump coke is ground and polished, and then subjected to the polarization structure analysis of the polished face with the point counter method.

The results obtained are shown in Table 1.

1) The isotropic texture and the fine mosaic texture, both derived from weak coking coals, decrease their amounts with the increase of the rate of gasification.
2) The fusite-like texture among the inert constituents decreases its quantity with the progress of gasification.
3) The fibrous texture and the leaflet texture, both derived from heavy coking coals, are relatively intensified by the progress of gasification and maintained the cell wall of coke to a gasification rate of 50%, beyond which the cell wall of coke also degrades into fragments.
4) The coarse mosaic texture and the fragment texture (micrinite and sclerotinite in coal) among the inerts are relatively intensified by the progress of gasification, and at a gasification rate above 60%, they are embrittled to such an extent that they are easily broken by the finger pressure. Therefore, they are found in the state covering the lump coke surface with a several millimeters thick or contained in the coke breeze in a blast furnace.

2. Changes of the Coke Structure in the Blast Furnace

When some physicochemical actions are given to the coke existing in the blast furnace, the structure having higher chemical reactivity first reacts, degrades, gasifies and falls away. Also, the structure which is mechanically weak first pulverizes and falls away, thus embrittling the whole coke lump. Therefore, by the observation of the structural changes of coke remaining in the furnace, it is possible to trace what part of coke first degraded.

The results of the polarization structure analysis of coke remained in Kukioka No. 4 B.F. is shown in Table 2. Characteristic aspects observed are described below.

1) No change was observed in the upper part of the shaft, but in the middle part and below that part, the fusite-like texture and the isotropic texture derived from soft coking coal degraded partially only on the surface by the carbon solution reaction. In the lower part of the shaft, the surface degradation progressed extensively, and the pores were enlarged by the selective reaction in the layer about 3 mm from the surface.
2) In the bosh, the surface degradation of coke remarkably advanced, and inside the coke lumps the fusite-like texture, derived from soft or medium-heavy coking coal, selectively fell away (like the isotropic texture), leaving cavities in the cell wall of coke. Consequently, it formed the coke structure which is weak for the mechanical impact.
3) Between the tuyeres, the degradation of coke advanced to a degree similar to that in the bosh. The characteristic observed at this level was a spotted structure produced in the isotropic texture due probably to the reducing volatilization of SiO₂.
4) In the raceway, coke carbon fell away selectively from not only the coke surface but from the inside, making the coke porous and brittle.
5) In the hearth, it was found that the cell wall of coke thinned to varying degrees at different loca-

<table>
<thead>
<tr>
<th>Rate of gasification (%)</th>
<th>Microscopic texture</th>
<th>Isotropic texture</th>
<th>Mosaic texture</th>
<th>Fibrous and leaflet texture</th>
<th>Fragment and fusite-like texture</th>
<th>Mineral matter</th>
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<tr>
<td></td>
<td></td>
<td></td>
<td>Fine mosaic</td>
<td>Coarse mosaic</td>
<td>Fragment</td>
<td>Fuse-like</td>
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<td></td>
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<td>60</td>
<td>4.9</td>
<td>5.5</td>
<td>29.2</td>
<td>5.1</td>
<td>3.4</td>
<td>36.5</td>
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Condition of gasification: Sample 10 mm cubic, Temp. 1200°C, Gas CO₂ 100%
Table 2. Microscopic analysis of coke (% VOL) (Kukioka No. 4 B.F.)

<table>
<thead>
<tr>
<th>Sampling position</th>
<th>Microscopic texture</th>
<th>Isotropic texture</th>
<th>Mosaic texture</th>
<th>Fibrous and leaflet texture</th>
<th>Fragment and fusite-like texture</th>
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<tr>
<td></td>
<td></td>
<td>Fine mosaic texture</td>
<td>Coarse mosaic texture</td>
<td>Fibrous texture</td>
<td>Leaflet texture</td>
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<tr>
<td>Wall side</td>
<td>Shaft upper</td>
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<td>57.9</td>
<td>8.1</td>
<td>14.2</td>
</tr>
<tr>
<td></td>
<td>&quot; middle</td>
<td>10.3</td>
<td>54.1</td>
<td>15.5</td>
<td>20.1</td>
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<tr>
<td></td>
<td>&quot; down</td>
<td>15.6</td>
<td>49.2</td>
<td>21.8</td>
<td>13.4</td>
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<td>53.8</td>
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<td>12.5</td>
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<tr>
<td></td>
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<td>25.7</td>
<td>17.8</td>
</tr>
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<td>16.7</td>
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<td>34.8</td>
<td>10.8</td>
<td>28.3</td>
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<td>Melting zone</td>
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<td>41.0</td>
<td>8.4</td>
<td>34.0</td>
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<td>Small size coke (top of tuyere)</td>
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<td>40.8</td>
<td>10.6</td>
<td>36.0</td>
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<td>Lump coke (top of tuyere)</td>
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<td>34.8</td>
<td>9.8</td>
<td>36.3</td>
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<td></td>
<td>Circumference of raceway</td>
<td>26.2</td>
<td>39.8</td>
<td>18.6</td>
<td>15.4</td>
</tr>
<tr>
<td></td>
<td>Circumference of raceway (-5 mm size coke)</td>
<td>10.8</td>
<td>56.8</td>
<td>14.4</td>
<td>18.0</td>
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</table>

Photo. 3. Changes of coke texture (Kukioka No. 4 B.F.) (×200 oil) (×4/5)
tions because of the intrusion of slag.

These characteristic features are shown in Photos. 1 to 3. As seen in Photo. 1, the structural network of coke remains unchanged in the shaft, but the cell wall of coke becomes thin and the pores are enlarged in the vicinity of tuyere and in the hearth. Photograph 2 shows the coke condition in the bosh. A cavity marked with the letter "A" is observed in the cell wall of coke. The coke condition around the tuyeres is shown in Photo. 3, where the characteristic spotted-structure generated is marked with the letter "B."

3. Discussions

Based on the observations and the results of these dissective investigations, some points which are contributive to the coke production or the blast furnace operation will be discussed below.

(1) The isotropic reactives and fusite (except the fusite surrounded by the melting components of coal is hard to gasify), derived from soft coking coal, are highly reactive with CO\(_2\) and are selectively gasified, being responsible for the brittleness of coke. This is caused either by the sensitivity of carbon of isotropic reactivates for the chemical reactions or by a large reactive surface area owing to the pores generated during the period of coking with more volatile matter. Fusite is also easy to gasify in an oxidizing atmosphere because of its porous charcoal-like structure.

(2) For the blast furnace operation, the reactivity of coke is thought to give an influence on the balance between the coke ratio and the quantity of heat generated. Although it is difficult to discuss the degree of reactivity here, the generation of large quantity of coke breeze due to the reduction of coke strength by the gasification is undesirable for the blast furnace condition. Considering that the coke strength after the reaction at a temperature in the range of 1200° to 1450°C plays an important role for the blast furnace operation, it is logically concluded that the lower the coke reactivity is, the better the operation becomes. In the temperature range shown above, the rate determining step of the reaction between coke and CO\(_2\) may be the intermediate one of the chemical reaction controlled and the diffusion controlled. Since the structural change of coke by the reaction with the gas takes place in the layer several millimeters deep from the surface, more suitable coke can be produced by improving the coke size and density.

(3) The degradation of coke is more prominent in the region from the bosh to a level near the raceway.
Especially, a marked difference in the size of coke above and below 15 mm as the boundary line is observed among coke samples taken out of the blast furnaces. Apart from the question whether this phenomenon is brought about by the breakage into fines of -15 mm or by the intense gasification, -15 mm coke fines are degraded throughout. Because the rate of reaction between coke and CO₂ gas happen above 1450°C is controlled by the pore diffusion, or by the gas film diffusion the effect of outside surface area, namely, the coke size, is naturally large. Therefore, by the adoption of large size coke, the size degradation and the pulverization of coke can be prevented to some extent.

(4) Since the isotropic reactivates and fusite, derived from soft coking coal, are sensitive to the reaction with CO₂ gas as mentioned previously, a method which is pulverized and dispersed them in the cell of coke is developed in order to minimize the effect even when they react with the gas in the furnace. Or, the method which is increasing the ratio of melting components in coke is adopted so that the embrittlement of coke by

![Photo. 2. Changes of coke texture (Kukioka No. 4 B.F.) (×200 oil) (×4/5)]
the selective gasification of the above two constituents of coke in furnace is depressed to some extent. (The latter method will be particularly effective in the case that Australian coal is used a high blending ratio.)

V. Summary

(1) Changes of the coke properties in the furnace may be started in the lower part of the shaft. The degree of the change depends on the inner volume of furnace, the operating practice adopted, and the coke properties before charging.

(2) Coke existing in the upper part of the hearth degrades mostly from the surface. Especially, the degradation of coke is more prominent in the vicinity of raceway, and the gasification advances appreciably even in the inner part of coke.

(3) Coke structure becomes porous by the selective carbon solution reaction.

(4) Coke structure weakens as the porosity increases and coke is pulverized possibly by the abrasion during the descending of coke through the furnace and by the impact in the raceway.

(5) Constituents of coke identified by the polarization structure analysis are enumerated below in the order of high chemical sensitivity to the carbon solution reaction.

i) Isotropic reactives derived from soft coking coal

ii) Porous fusite derived from soft and medium-heavy coking coke

iii) Mosaic texture

iv) Inerts completely coated with anisotropic re-actives, micrinite, fibrous texture and leaflet texture

Thus, it can logically be concluded that coke existing in the blast furnace becomes porous by the selective carbon solution reaction of the constituents with CO₂ gas, weakening its structure, and the embrittled coke pulverizes due to the abrasion during the descending of coke through the furnace and by the impact in the raceway.

REFERENCES


