Effective Utilization Technique for Coal Having High Fluidity and Long Maximum Permeation Distance by Coal Size Adjustment

Yusuke DOHI,* Kiyoshi FUKADA, Tetsuya YAMAMOTO, Takashi MATSUI, Hiroyuki SUMI and Izumi SHIMOYAMA

Steel Research Laboratory, JFE Steel Corp., 1 Kokan-cho, Fukuyama, Hiroshima, 721-8510 Japan.

(Received on August 15, 2019; accepted on October 28, 2019; originally published in Tetsu-to-Hagané, Vol. 104, 2018, No. 10, pp. 525–534)

In our previous paper, we proposed a new measurement method for coal thermoplasticity called the “permeation distance method,” in which the permeation distance of thermally plastic coal into a glass bead layer adjacent to the coal sample is measured as a unique caking property. Although the maximum permeation distance measured by this method is roughly correlated with Gieseler fluidity, large deviation is observed, especially for high fluidity coals. Moreover, that study revealed that high MF coal having a longer maximum permeation distance forms thinner pore-wall structures in coke and that coke strength deteriorates when the coal blend includes longer maximum permeation distance coal. Therefore, a technique for reducing the adverse effects of long permeation distance coal on coke strength is necessary so as to utilize the coal more efficiently.

In this paper, the influence of the grain size of mainly high MF coal on permeation distance and coke strength was investigated to clarify the possibility of controlling the permeation distance. As a result, it was found that the measured maximum permeation distance became shorter with decreasing coal size. Moreover, the coke strength deterioration caused by long permeation distance coal in a coal blend was suppressed as the size of the long permeation distance coal became smaller. Consequently, coal grain size design and control techniques for more effective utilization of long permeation distance coal were proposed.

KEY WORDS: cokemaking; coal thermoplasticity; permeation distance; coal size adjustment; preparation technique.

1. Introduction

Caking properties of coal such as maximum fluidity (MF) and total dilatation (TD), which are provided in JIS M 8801,1) are conventionally the most important factors in coal blending theory for controlling coke strength.2) However, in some cases, the coke strength predicted based on these conventional properties does not match the actual coke strength. In our previous study,3) a novel measurement method for coal thermoplasticity, namely the “permeation distance method,” was developed to solve the problems of the conventional method. In the developed method, glass beads are placed on a coal sample arbitrarily to simulate the surrounding voids. Under heating conditions simulating an actual coke oven, thermally plastic coal particles permeate into the glass bead layer. The distance that the thermally plastic coal permeates into the glass bead layer is defined as the permeation distance. The maximum value of the permeation distance during the thermally plastic stage (referred to here as maximum permeation distance), which is a newly developed index, has a roughly positive correlation with the conventional caking property (MF). However, large deviation from that correlation was observed, especially in the case of high fluidity coals whose log MF exceeds about 3.0. Therefore, the maximum permeation distance is a unique index representing thermoplasticity different from the conventional caking properties. In addition, coke strength was decreased by blending coals that have a long maximum permeation distance of 15 mm or more (descriptively referred to here as long permeation distance coal), because the coal forms distorted pores and thin pore walls in the resultant coke structure. Therefore, coke strength can be increased by reducing the blending ratio of long permeation distance coal. As described above, for precise prediction of coke strength, permeation distance is effective as an evaluation index of caking property, particularly for high MF coal.

Recently, production of high strength coke has been strongly required for low reducing agent ratio operation of blast furnaces. On the other hand, from the viewpoint of effective utilization of carbon resources, it is desirable to use long permeation distance coals, even if this may cause a reduction in coke strength. Therefore, it is necessary to develop a technology capable of reducing the adverse effect of long permeation distance coal on coke strength.

* Corresponding author: E-mail: y-dohi@jfe-steel.co.jp
DOI: https://doi.org/10.2355/isijinternational.ISIJINT-2019-518

© 2020 ISIJ
In our previous study, we showed that fine crushing of a coal blend is effective for reducing the adverse effect of long permeation distance coal on coke strength. However, the mechanism responsible for this improvement has not been fully studied. Therefore, the aim of the present study is to clarify the mechanism by which fine crushing of the coal blend reduces the adverse effect of long permeation distance coal on coke strength. Finally, effective methods for adjusting the coal particle size for practical use were proposed.

2. Preliminary Study Regarding Influence of Coal Size on Coke Strength

Figure 1 schematically shows the process of pore formation during carbonization of coal particles in a coal blend containing long permeation distance coal. Upon heating, the coal blend becomes thermally plastic at about 400–500°C and generates bubbles in the particles. At that time, particles of long permeation distance coal permeate excessively into the surrounding voids. Simultaneously, the bubbles coalesce with each other and form distorted pores and thin pore walls. It is supposed that these remain as defects in the coke and cause a decrease in the coke strength. We considered the following two cases concerning the mechanism by which the adverse effect of long permeation distance coal on coke strength can be suppressed by size refinement of a coal blend containing long permeation distance coal. In Case 1 (Fig. 1(b)), the long permeation distance coal itself is fine-grained. In Case 2 (Fig. 1(c)), coal particles not corresponding to long permeation distance coal (hereinafter, non-long permeation distance coal), i.e., coal particles around the long permeation distance coal, become finer. The effects that can appear in each case and the respective mechanisms are shown in the system diagram in Fig. 2 and are organized below.

[Case 1] The maximum permeation distance of the long permeation distance coal is shortened, and as a result, the defect area formed by the long permeation distance coal is reduced; this has the effect of suppressing the generation of defects (Effect ①) and the effect of improving homogeneity by uniform dispersion of the long permeation distance coal (Effect ②) (Figs. 1(b), 2).

According to previous studies investigating the effects of particle size on caking properties, fluidity (MF) decreases with coal size refinement. Since the maximum permeation distance is one index of caking that has a substantially positive correlation with MF, there is a possibility that the maximum permeation distance may be reduced by size refinement. Moreover, as shown in the discussion in our previous report, since the expansion pressure decreases, it can be inferred that the maximum permeation distance also decreases, as the expansion pressure is considered to be the main influential factor of the permeation phenomenon. In addition, when the long permeation distance coal itself is fine-grained, it is also considered that the permeation distance is shortened due to a simple reduction in the amount of the material to be permeated accompanying the reduction of the size. Moreover, the negative effect on coke strength would be also suppressed because the suppression of connected pore defects formed in the long permeation distance coal. It was reported that larger pore defects negatively influence on coke strength. The size of connected pores from long maximum permeation distance coal would be reduced with refining the long maximum permeation distance coal containing long permeation distance coal. In Case 1 (Fig. 1(b)), the long permeation distance coal itself is fine-grained. In Case 2 (Fig. 1(c)), coal particles not corresponding to long permeation distance coal (hereinafter, non-long permeation distance coal), i.e., coal particles around the long permeation distance coal, become finer. The effects that can appear in each case and the respective mechanisms are shown in the system diagram in Fig. 2 and are organized below.

[Case 1] The maximum permeation distance of the long permeation distance coal is shortened, and as a result, the defect area formed by the long permeation distance coal is reduced; this has the effect of suppressing the generation of defects (Effect ①) and the effect of improving homogeneity by uniform dispersion of the long permeation distance coal (Effect ②) (Figs. 1(b), 2).

According to previous studies investigating the effects of particle size on caking properties, fluidity (MF) decreases with coal size refinement. Since the maximum permeation distance is one index of caking that has a substantially positive correlation with MF, there is a possibility that the maximum permeation distance may be reduced by size refinement. Moreover, as shown in the discussion in our previous report, since the expansion pressure decreases, it can be inferred that the maximum permeation distance also decreases, as the expansion pressure is considered to be the main influential factor of the permeation phenomenon. In addition, when the long permeation distance coal itself is fine-grained, it is also considered that the permeation distance is shortened due to a simple reduction in the amount of the material to be permeated accompanying the reduction of the size. Moreover, the negative effect on coke strength would be also suppressed because the suppression of connected pore defects formed in the long permeation distance coal. It was reported that larger pore defects negatively influence on coke strength. The size of connected pores from long maximum permeation distance coal would be reduced with refining the long maximum permeation distance coal containing long permeation distance coal.

![Fig. 1. Presumed mechanism of suppression of negative effect of long permeation distance coal on coke strength by reducing grain size of coal blend (a) Mechanism of negative effect, (b) mechanism of suppression of negative effect by reducing grain size of long maximum permeation distance coal, (c) mechanism of suppression of negative effect by reducing grain size of non-long maximum permeation distance coal). (Online version in color.)](image1.png)

![Fig. 2. System diagram of mechanism of positive effect of long permeation distance coal on coke strength by reducing grain size of coal. (Online version in color.)](image2.png)
distance coal. Therefore, the negative effect of connected pores from long maximum permeation distance coal would be also reduced.

On the other hand, it is generally known that size refinement of coal improves homogeneity and coke strength after carbonization.\(^8,9\) When a material is virtually divided into a number of blocks having a certain size, the homogeneity of the material is defined as the degree of property variation of each block. A material with smaller variation is regarded as a material having higher homogeneity. The homogeneity of coke should mainly depend on the homogeneity of the raw material coal. However, coal is heterogeneous because it is composed of various macerals with different characteristics such as vitrinite and inertinite. Moreover, heterogeneity increases further in the coal blends which are used industrially in coke production because blends are composed by blended particles of a plurality of coal brands with different properties. For that reason, it can be understood that the homogeneity of a coal blend is governed by the maceral and particle compositions per virtual block. As the coal particles become finer, the macerals become finer as well, and the number of coal macerals and particles contained in the virtual block increases. Therefore, the compositions of the macerals and particles in a virtual block stochastically must approach the composition of the whole coal blend, which means the properties of any virtual block move in the same direction as the average properties of the coal blend. Finally, the variation in quality among blocks is reduced and homogenized. Suginobe et al. theorized the relationship between coke strength and the homogeneity and particle size composition of the charged coal by a statistical approach, assuming that coke strength is fundamentally determined by Ro and MF. It has been pointed out that coke strength is improved by crushing and uniformly dispersing coal brands whose Ro and MF have large deviation from the weighted averaged value of those properties of the whole coal blend.\(^8\) Since long permeation distance coal has higher MF than the one of commercial coal blend used in general coke plant, the effect of homogenization due to size refinement is considered to be comparatively large.

Thus, when long permeation distance coal is refined, if the effect of suppressing the formation of defects (Effect \(①\)) and homogenization (Effect \(②\)) are larger than the influence of the deterioration in fluidity, the adverse effect of the long permeation distance coal on coke strength will also be suppressed. Although Effect \(②\) is generally known, Effect \(①\) has not been fully confirmed.

[Case 2] This case assumes the effect of reducing the generation of defects because the permeability coefficient around the long permeation distance coal is reduced and, as a result, the permeation of the long permeation distance coal is suppressed (Effect \(③\)). The effect of the reduction of the generation of defects derived from the non-long permeation distance coal (Effect \(④\)), and the effect of improvement of homogeneity by uniform dispersion of the non-long permeation distance coal (Effect \(⑤\)) (Figs. 1(c), 2).

In our previous paper,\(^3\) we reported that changing the diameter of the glass beads comprising the packed bed adjacent placed on the sample coal had the effect of changing the maximum permeation distance. Since the experimental results showed that the permeation behavior of coal depends on the permeability coefficient of the permeation region, the change in the permeability coefficient of the coal packed bed accompanying a change of the coal particle size can control the maximum permeation distance. That is, when the non-long permeation distance coal around the long permeation distance coal is fine-grained, the permeation coefficient of that part decreases. It is therefore presumed that the permeation of long permeation distance coal to the periphery is suppressed, and as a result, the generation of defects is suppressed and the decrease in coke strength is mitigated (Effect \(③\)).

When the non-long permeation distance coal is refined, the effect of suppressing the formation of defects formed by the non-long permeation distance coal (Effect \(①\)) and the homogenization effect by uniform dispersion of the non-long permeation distance coal (Effect \(⑤\)) should also contribute to improvement of coke strength. As a defect derived from non-long permeation distance coal, for example, micro cracks formed around inertinites which do not have thermoplasticity can be considered. It is widely known that coke strength is improved due to the decrease in micro cracks when coal containing a large amount of inertinite is finely crushed.\(^10–13\) As inertinite is generally included in all coals and is also present in non-long permeation coals, it is thought that Effect \(①\) should occur, although the size of the effect will depend on the coal brands. Furthermore, since the non-long permeation distance coal is also composed of various macerals and thus is heterogeneous, it can be inferred that Effect \(⑤\) also occurs. Although the existence of Effects \(①\) and \(⑤\) is well known from the past, Effect \(③\) has not been sufficiently studied.

In this study, several tests were carried out based on the above considerations in order to elucidate the mechanism responsible for suppressing the decrease in coke strength caused by long permeation distance coal when the grain size is refined, and to organize the impact on coke strength. Furthermore, methods of particle size adjustment were investigated with the aim of controlling the maximum permeation distance and effectively utilizing long permeation distance coal.

### 3. Experiments

A total of four tests were conducted in this study (Tests 1–4). In Test 1, the effects of the coal particle size on the maximum permeation distance were studied. Tests 2 to 4 were conducted to investigate the influences of changes in the particle size of the coal blends including highly caking coal, which was mainly a long permeation distance coal, on coke strength. Table 1 shows the property values of the single coal brands used in each test. The numbers after the sample names indicate the differences in lots. The mean maximum reflectance of vitrinite (Ro), total amount of inerts (Ti), maximum fluidity (MF) and proximate analysis values were measured respectively according to JIS M 8816,\(^15\) M 8801\(^1\) and M 8812.\(^2\) The maximum permeation distance was measured in accordance with the method described in our previous paper.\(^3\) That is, 2.5 g of air-dried whole sample coal was crushed so that all particles passed through 2.0 mm sieve, and the maximum
permeation distance of each sample was evaluated under the standard measurement conditions (particle diameter of glass bead: 2.0 mm, weight load: 1.6 kg) by measuring the weight of glass beads in their layer into which the sample coal permeated during the test and calculating based on the relationship between the weight and height of glass beads layer.

In Test 1, in order to clarify the relationship between the coal particle size and permeation distance, the permeation distance measurement was carried out with different coal particle sizes. For Coal A1, B1, C1 and D1, whose whole coal particle size was changed to 0.25, 1.0 and 2.0 mm or less, the maximum permeation distances of the sample coals were measured under the standard measurement conditions in the previous report except for the condition of coal size.

In Test 2, the change in coke strength was investigated when the particle sizes of coal blends including and not including long permeation distance coal was varied. In this test, some of the aforementioned effects may possibly occur as the grain size is reduced. That is, in a coal blend containing long permeation distance coal, there is a possibility that all of the above Effects ①–⑤ may occur. On the other hand, in a coal blend not containing long permeation distance coal, Effects ④ and ⑤ are likely to appear.

Table 2 provides the blending ratio and property values

| Test 2 | Blend 1 | Blend 2 | Blend 3 | Blend 4 |
|--------|---------|---------|---------|---------|
| CoalE1 | 0.0     | 0.0     | 10.0    | 10.0    |
| CoalF1 | 15.0    | 15.0    | 5.0     | 5.0     |
| CoalG1 | 15.0    | 15.0    | 15.0    | 15.0    |
| CoalH1 | 20.0    | 20.0    | 20.0    | 20.0    |
| CoalD2 | 25.0    | 25.0    | 25.0    | 25.0    |
| CoalI1 | 10.0    | 10.0    | 6.0     | 6.0     |
| CoalJ1 | 15.0    | 15.0    | 19.0    | 19.0    |
| Ro (%) | 1.00    | 1.00    | 1.01    | 1.01    |
| logMF (log/ddpm) | 2.8 | 2.8 | 2.8 | 2.8 |

Table 2. Coal blending and size conditions (Test 2).

Table 1. Single coal properties and measurement results.

| Test No. | Coal brand | Ro*1 (%) | logMF*2 (log/ddpm) | TI*3 (%) | Ash (wt% d.b.) | VM*4 (wt% d.b.) | Maximum permeation distance (mm) |
|----------|------------|-----------|---------------------|----------|----------------|----------------|-------------------------------|
| Test 1   | CoalA1     | 0.79      | 3.96                | 13.5     | 7.9            | 37.2           | 21.3                          |
|          | CoalB1     | 0.63      | 4.36                | 20.6     | 7.6            | 41.5           | 10.6                          |
|          | CoalC1     | 0.90      | 3.39                | 14.9     | 7.3            | 34.5           | 20.0                          |
|          | CoalD1     | 0.99      | 2.84                | 35.0     | 8.7            | 29.1           | 6.8                           |
|          | CoalE1     | 0.71      | 4.78                | 8.0      | 0.4            | 43.6           | 32.4                          |
|          | CoalF1     | 0.72      | 4.09                | 14.3     | 9.5            | 40.1           | 14.9                          |
|          | CoalG1     | 0.90      | 3.48                | 29.3     | 8.2            | 31.1           | 11.5                          |
|          | CoalH1     | 0.96      | 2.67                | 34.9     | 9.5            | 28.0           | 7.5                           |
|          | CoalD2     | 1.00      | 2.61                | 34.9     | 8.4            | 27.8           | 6.3                           |
|          | CoalI1     | 1.11      | 3.04                | 31.1     | 8.8            | 24.5           | 9.7                           |
|          | CoalJ1     | 1.37      | 1.04                | 44.3     | 7.2            | 19.3           | 0.9                           |
| Test 2   | CoalA2     | 0.80      | 3.17                | 14.5     | 10.0           | 35.9           | 20.8                          |
|          | CoalB2     | 0.66      | 3.55                | 18.3     | 5.4            | 44.0           | 8.2                           |
|          | CoalD3     | 1.02      | 2.48                | 33.9     | 8.1            | 27.7           | 6.3                           |
|          | CoalI2     | 1.31      | 1.26                | 45.6     | 7.2            | 20.4           | 0.9                           |
|          | CoalK1     | 1.60      | 0.70                | 20.1     | 9.2            | 17.6           | 3.0                           |
|          | CoalL1     | 1.14      | 1.77                | 35.0     | 9.2            | 24.2           | 4.9                           |
|          | CoalM1     | 1.00      | 2.20                | 33.3     | 11.6           | 27.7           | 4.6                           |
|          | CoalN1     | 0.67      | 1.00                | 23.0     | 10.7           | 36.6           | 0.0                           |
|          | CoalO1     | 1.38      | 2.49                | 29.4     | 10.9           | 20.9           | 8.7                           |
|          | CoalC2     | 0.87      | 3.63                | 17.2     | 7.6            | 34.5           | 18.7                          |
|          | CoalH2     | 0.93      | 2.82                | 27.7     | 9.0            | 28.6           | 11.7                          |
|          | CoalP2     | 1.00      | 2.29                | 38.6     | 9.6            | 26.3           | 2.2                           |
|          | CoalJ2     | 1.29      | 1.11                | 33.6     | 7.6            | 20.1           | 0.9                           |
|          | CoalQ1     | 1.51      | 1.85                | 29.1     | 10.2           | 19.1           | 7.0                           |

*1 Mean maximum reflectance of vitrinite in oil  *2 Maximum Fluidity  *3 Total Inert  *4 Volatile Matter
of the coal blends used in the test. In Test 2, Coal E1 was selected as the long permeation distance coal. The particle size levels of the whole coal blend were set to 100 wt% of 3.0, 6.0 mm or less, while the blending ratio of Coal E1 was varied from 0 wt% (Blends 1, 2) to 10 wt% (Blends 3, 4). The weighted average $R_O$ and log MF of the coal blend were controlled so as to be constant. The prepared coal blends were carbonized under the conditions listed in Table 3. After cooling in a nitrogen atmosphere, the drum strength of each obtained coke was tested in accordance with JIS K 2151.17)

In Test 3, the particle size of only high MF coal with different maximum permeation distances which were contained in coal blend was changed to investigated the influence on coke strength. In this test, as the high MF coal becomes finer, it is possible that Effects 1 and 2 described above may occur when the high MF coal is a long permeation distance coal, and Effects 3 and 4 are likely when the high MF coal is a non-long permeation distance coal. Table 4 shows the properties of the coals used in the test and their blending ratios in the coal blends. In Test 3, Coal A2 was selected as the long permeation distance and high MF coal, while B2 was chosen as the non-long permeation distance coal. The particle size of the whole coal blend was kept constant. For this objective, the size of the long permeation distance coal was reduced in the case of coarser particle sizes of the non-long permeation distance coal, whose properties were relatively close to the weighted averaged properties of the coal blend. Therefore, if the particle size of the long permeation distance coal is reduced, there is a possibility that adverse Effects 3, 4, and 5 as well as Effects 1 and 2 described above may occur. Conversely, if the particle size of the long permeation distance coal is coarsened, while the adverse Effects 1 and 2 can occur, Effects 3, 4 and 5 can also appear. In addition, based on the theory of homogenization proposed by Suginobe et al.,7) the homogenization effect of size refinement of coal having properties relatively close to the weighted averaged properties of a coal blend is comparatively small. In other words, Effect 5 is presumed to be relatively small because the variation of homogeneity with the change in particle size should be comparatively small. Table 5 shows the coals used in the test, their blending ratios and the property values of the coal blends. In Test 4, Coal C2 was selected as the long permeation distance coal and blended at 10 wt% in each coal blend. The coal brands and the blending ratios of all coal blends (Blends 11–13) used in this test were the same. Blend 11 was the Base condition. All coal brands contained in Blend 11 were prepared to 70 wt% of 3 mm or less. In Blends 12 and 13, the particle size of the long permeation distance Coal C2 was varied 8–10 respectively have the same compositions. The particle sizes of Coal A2 and B2 were adjusted to 100 wt% of 1.0, 3.0 and 6.0 mm or less, and the other coals were adjusted to 100 wt% of 3.0 mm or less. The coal blends were carbonized under the carbonization conditions shown in Table 3. After quenching and cooling, the strength of the obtained coke was measured.

In Test 4, the effect of the particle size of long permeation distance coal on coke strength was investigated while the particle size of the whole coal blend was kept constant.

### Table 3. Carbonization test conditions (Test 2–4).

| Test 2, 3 | Test 4 |
|----------|--------|
| Moisture content (wt%) | 8 | 8 |
| Bulk density (kg-dry/m³) | 750 | 775 |
| Dimensions of test coke oven (mm) | W270×H220×L263 | W273×H300×L260 |
| Wall temperature (°C) | 1 050 | 1 150 |
| Coking time (min) | 360 | 360 |

### Table 4. Coal blending and size conditions (Test 3).

| Coal brand | Blend 5 | Blend 6 | Blend 7 | Blend 8 | Blend 9 | Blend 10 |
|------------|--------|--------|--------|--------|--------|---------|
| CoalA2: −6 mm | 20.0 | 20.0 | 20.0 | 0.0 | 0.0 | 0.0 |
| CoalB2: −6 mm | 0.0 | 0.0 | 0.0 | 20.0 | 20.0 | 20.0 |
| CoalD3: −3 mm | 20.0 | 20.0 | 20.0 | 19.0 | 19.0 | 19.0 |
| CoalJ2: −3 mm | 7.0 | 7.0 | 7.0 | 11.0 | 11.0 | 11.0 |
| CoalK1: −3 mm | 0.0 | 0.0 | 0.0 | 4.0 | 4.0 | 4.0 |
| CoalL1: −3 mm | 11.0 | 11.0 | 11.0 | 11.0 | 11.0 | 11.0 |
| CoalM1: −3 mm | 20.0 | 20.0 | 20.0 | 13.0 | 13.0 | 13.0 |
| CoalN1: −3 mm | 13.0 | 13.0 | 13.0 | 14.0 | 14.0 | 14.0 |
| CoalO1: −3 mm | 9.0 | 9.0 | 9.0 | 8.0 | 8.0 | 8.0 |
| $R_o$ (%) | 0.99 | 0.99 | 0.99 | 0.99 | 0.99 | 0.99 |
| logMF (log/ddpm) | 2.2 | 2.2 | 2.2 | 2.2 | 2.2 | 2.2 |

### Table 5.

| Coal size (wt%) | CoalA2: −6 mm | CoalA2: −3 mm | CoalA2: −1 mm | CoalB2: −6 mm | CoalB2: −3 mm | CoalB2: −1 mm |
|----------------|---------------|---------------|---------------|---------------|---------------|---------------|
| Others: −3 mm 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% |
|Others: −3 mm 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% |
| Others: −3 mm 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% | 100 wt% |
from 45 to 90 wt% of 3 mm or less, while the size of the non-long permeation distance Coal H2, whose properties were relatively close to the weighted average properties of the coal blend, were changed from 67–74 wt% of 3 mm or less and the size of other coal brands were set to 70 wt% of 3 mm or less. As a result of this preparation, the overall particle size of Blends 11–13 became 70 wt% of 3 mm or less, which is almost equal to that of the Base condition. The prepared coal blends were carbonized under the carbonization conditions in Table 3, and after quenching and cooling, the strength of the coke was evaluated.

The pore structure of the coke produced in Test 4 was quantified by using an image processing software (Nippon Roper: Image-Pro Plus version 7.0) according to the following procedure. First, two lumps of coke were sampled for each level. Next, the lumps were embedded in resin and polished, and 60 images were taken with an optical microscope (Olympus: BX51M) at a magnification of 50 times. All photographed images (2,000 × 1,500 μm) were binarized on the basis of predetermined threshold values of RGB and classified into the coke matrix and pores. The pore-wall thickness (i.e., thickness of coke matrix) and the diameter and roundness of the pore portion were then determined by the following image processing methods. As in previous papers,3,18 the pore-wall thickness is measured according to the following procedure: Parallel measurement lines (width: 5 μm) were placed on the photographed image at intervals of 100 μm, and all of the coke matrix located on the lines was enclosed with a the circumscribed square so that the sides of the square were parallel and perpendicular to the measurement lines. The length of the side of the square parallel to the measurement line was measured for all squares, and the average value of the lengths measured in the whole analysis region was calculated as the pore-wall thickness. For each pore, the average value of the pore diameter obtained by connecting two points on the outer periphery of the pore and measuring the diameter passing through the center of gravity at intervals of 2° was taken as the measured value of the pore diameter. The roundness of the pore is expressed by the following Eq. (1):

\[ R = \frac{4\pi S}{Pe} \]

where \( R \) is roundness (−), \( S \) is pore area (mm²), and \( Pe \) is pore peripheral length (mm). The degree of roundness ranges from 0 to 1, and as roundness approaches 1, the shape approaches a true circle. It is known that pores with low roundness cause a decrease in coke strength because stress concentrates on sharp parts of the pores and those parts become starting points for fracture when an external force is applied.7,19,20 Kubota et al. conducted a quantitative investigation of the relationship between pore data and DI150, which is an index of coke strength mainly due to surface fracture, and found that there was a good correlation between DI150 and the total area of pores with low roundness. Moreover, it is appropriate to extract pores with a roundness of 0.2 or less as connected pore defects in coke.19 Therefore, in this paper, connecting pores are also defined as low roundness pores having a roundness of 0.2 or less. The area occupied by low roundness pores in the whole analysis region was measured, and the relationship with the coke strength index DI150 was examined.

### 4. Results and Discussion

As a result of Test 1, Fig. 3 shows the relationship between the particle size of coal and the maximum permeation distance. The maximum permeation distance of all coals showed a tendency to decrease with size refinement. This result was consistent with the prediction from the previous paper.17 Thus, it was shown that control of the maximum permeation distance is possible by changing the particle size of coal. Regarding Effect ①, it was suggested that size refinement may shorten the permeation distance of the long permeation distance coal, and a mechanism in which the generation of defects derived from long permeation distance coal is suppressed may occur.

Figure 4 shows the relationship between the particle size of the whole coal blend and the drum strength of the coke obtained in the carbonization test in Test 2. The relationship in this figure is shown by classifying the blending ratio of the long permeation distance Coal E1. When compared at the same particle size levels, the coke strengths of the coal blends containing 10 wt% of long permeation distance Coal E1 were confirmed to be lower than those of the blends not containing Coal E1, in spite of the fact

| Table 5. Coal blending and size conditions (Test 4). |
|-----------------|-----------------|-----------------|-----------------|
| Coal brand      | Blend 11        | Blend 12        | Blend 13        |
| CoalH2          | 10.0            | 10.0            | 10.0            |
| CoalP2          | 60.0            | 60.0            | 60.0            |
| CoalP2          | 10.0            | 10.0            | 10.0            |
| CoalJ3          | 10.0            | 10.0            | 10.0            |
| CoalQ1          | 10.0            | 10.0            | 10.0            |
| Ρ₀ (%)          | 1.02            | 1.02            | 1.02            |
| logMF (log/ddpm)| 2.6             | 2.6             | 2.6             |

| Coal size (wt%)| Coal blend: | CoalH2: | CoalH2: |
|----------------|-------------|---------|---------|
| −3 mm 70 wt%   | −3 mm 90 wt%| −3 mm 67 wt%| −3 mm 74 wt%| Others: −3 mm 70 wt%| Others: −3 mm 70 wt%|

Fig. 3. Influence of grain size on maximum permeation distance (Test 1). (Online version in color.)
that the $R_0$ and log MF of all the coal blends were almost equal. When compared under the same blending conditions (i.e., Blends 1 and 2, Blends 3 and 4), coke strength was higher when the particle size of the coal blend was finer. The improvement in strength due to size refinement was larger in Blends 3 and 4, in which the long permeation distance Coal E1 was blended, than in Blends 1 and 2, which did not contain Coal E1. As described above, it was presumed that Blends 1 and 2 have Effects ④ and ⑤ of non-long permeation distance coal by size refinement, while Blends 3 and 4 can have Effects ①, ② and ③ on the long permeation distance coal in addition to Effects ④, ⑤. Thus, it was suggested that the effect of size refinement of the coal blend on coke strength changes depending on the presence or absence of long permeation distance coal in the coal blend, and the effect of Effects ① + ② + ③ may be relatively larger than that of Effect ④ + ⑤.

Figure 5 shows the relationship between the particle size of high MF and long permeation distance coal (Coal A2) or high MF and non-long permeation distance coal (Coal B2) and drum strength for the coke obtained in the carbonization test in Test 3. Under the same particle size conditions of high MF coal (Coal A2 and B2), Blends 5–7 containing long permeation distance coal (Coal A2) had lower strength than Blends 8–10 containing non-long permeation distance coal (Coal B2). In addition, as the particle size of the high MF coal was decreased, the difference in strength between the coal blend including Coal A2 and the coal blend including Coal B2 became smaller. Although each of the Blends 5–7 or each of the Blends 8–10 had the same blending composition, strength improved as the coal size of Coal A2 or B2 was reduced. Furthermore, the improvement width of Blends 5–7 blended with long permeation distance coal A2 was larger than that of Blends 8–10 blended with non-long permeation distance coal B2. As described above, it was considered that Blends 5–7 containing long permeation distance coal A2 had Effects ① and ② due to the size refinement of high MF coal, whereas Effects ④ and ⑤ can occur in Blends 8–10 containing non-long permeation distance coal B2. Therefore, it was suggested that Effect ① + ② of the size refinement of the long permeation distance coal is larger than Effect ④ + ⑤ of the size reduction of the non-long permeation distance coal (Effect ① + ② > Effect ④ + ⑤).

Next, we attempted to confirm each effect in Test 3. Based on the concept of homogenization by crushing proposed by Suginobe et al., 3) the impact of homogenization by crushing becomes greater with a coal brand whose $R_0$ and MF have larger differences from the weighted average $R_0$ and MF of the coal blend. The properties of all Blends 5–10 used in this test were all $R_0 = 0.99\%$, log MF = 2.2 (ddpm/log). The properties of the long permeation distance coal (Coal A2), whose particle size was changed, were $R_0 = 0.80\%$, log MF = 3.17 (ddpm/log), while the properties of the non-long permeation distance coal (Coal B2) were $R_0 = 0.66\%$, log MF = 3.55 (ddpm/log). That is, since the non-long permeation distance Coal B2 has a larger difference from the weighted average properties of the coal blend than the long permeation distance Coal A2, the effect of homogenization of Coal B2 due to size refinement presumably is greater than that of Coal A2 (Effect ② < Effect ④). Interestingly, the actual magnitude relationship was Effect ① + ② > Effect ④ + ⑤. Based on these findings, it was suggested that not only the effect of homogenization of long permeation distance coal (Effect ②) but also the effect of suppression of defect generation of long permeation distance coal (Effect ④) occur when long permeation distance coal is refined. Furthermore, the comparison of the magnitudes of Effect ① and Effect ② is considered. Among the strength improvement effects of size refinement of the non-long permeation distance coal (Coal B2) in Blends 8–10, the maximum magnitude of Effect ⑤ associated with the homogenization can be considered as the gradient of the regression line of the plot of Blends 8–10 shown in Fig. 5. This means the maximum magnitude of Effect ⑤ can be obtained if Effect ① equals 0. As described above, in the case of this test including Blends 5–7, because the magnitude relationship was Effect ② < Effect ④, the maximum magnitude of Effect ② in relation to homogenization can be considered as the gradient of the regression line of the plot of Blends 8–10. That is, when the particle size of the long permeation distance coal (Coal A2) was changed from the coarsest Blend 5 to the finest Blend 7, the change on the straight line whose gradient corresponds to the regression line of the plot of Blends 8–10 (see the dash line in Fig. 5) showed the maximum impact of Effect ② of Coal A2. Conversely, it was considered that the difference between the whole strength improvement effect accompanying size
refinement of the long permeation distance coal (Coal A2) and the dash line showed the minimum impact of the defect suppression effect (Effect ①) of Coal A2. Therefore, based on the comparison of the presumed minimum Effect ① and the maximum Effect ②, it was understood that Effect ① is larger.  

Figure 6 shows the relationship between the particle size of the long permeation distance coal (Coal C2) contained in Blends 11–13 and the drum strength obtained in Test 4. Although the blending composition and the particle size of the whole coal blend were the same, it was confirmed that strength improved as the size of Coal C2 was reduced. More specifically, focusing on the change in Blend 13 → Blend 11 → Blend 12, it was presumed that Effect ① + ② was obtained by size refinement of the long permeation distance coal (Coal C2), while coarsening of the non-long permeation distance coal (Coal H2) resulted in occurrence of the opposite Effect ③ + ④ + ⑤ at the same time. However, strength was actually improved. Thus, it was suggested that the strength improvement effect of Effect ① + ② was greater than the opposite Effect ③ + ④ + ⑤. In other words, as shown in Fig. 1(c), Effect ③ (effect of suppressing defect formation by shortening the permeation distance of long permeation distance coal by decreasing the size of the coal particles around the long permeation distance coal) can be limited compared to Effect ① + ②.  

Figure 7 shows the results of imaging the pore structures of Blends 12 and 13. In addition, the pore size distributions of Blends 11–13 are shown in Fig. 8, and the relationship of the particle size of Coal C2, the pore-wall thickness of the coke, and the area of low roundness pores is illustrated in Fig. 9. As shown in Fig. 7, it was observed that Blend 12, in which the size of the long permeation distance coal (Coal C2) was refined, had a thicker pore wall and less distorted pores compared to Blend 13, in which Coal C2 was coarser. Moreover, as presented in Figs. 8 and 9, it was confirmed that the pore-wall thickness was increased and the area of pores with a low degree of roundness was also reduced by size reduction of the long permeation distance Coal C2, even though there was no significant difference in the pore size distribution. From the results of the pore structure analysis, it was understood that Effect ① (effect of suppressing the generation of defects derived from the long permeation distance coal) can be one factor in coke strength improvement when the long permeation distance coal (Coal C2) is fine-grained.

![Fig. 6. Relationship between grain size of Coal C2 and coke strength (Test 4, Coal C2: Long permeation distance coal). (Online version in color.)](image-url)

![Fig. 7. Optical microscope images of coke structure (Test 4, (a) Blend 12, (b) Blend 13). (Online version in color.)](image-url)

![Fig. 8. Histograms of mean pore diameter (Test 4). (Online version in color.)](image-url)

![Fig. 9. Effect of grain size of Coal C2 on pore-wall thickness and area of low roundness pores (Test 4, Coal C2: Long permeation distance coal). (Online version in color.)](image-url)
The above experimental results and discussion are summarized below. As shown in Fig. 1(b), it was found that the effect of suppressing the generation of defects derived from long permeation distance coal (Effect ①, which has not been sufficiently confirmed in the past) and the well-known effect of homogenization (Effect ②) occurred when the long permeation distance coal itself was fine-grained in a coal blend containing long permeation distance coal. As a result, it has become clear that the adverse effect of long permeation distance coal on coke strength can be suppressed. In particular, it was suggested that Effect ① was relatively larger than Effect ②. Therefore, in order to maintain coke strength when using long permeation distance coal in a coal blend, refining the size of the long permeation distance coal itself is effective. The same Effects ① and ② can also occur in case of refinement of the whole coal blend containing long permeation distance coal because the size of the long permeation distance coal is refined together with that of the other coals. It is considered that these are the main effects suppressing the adverse effect of long permeation distance coal.

5. Verification at Commercial Plant

5.1. Analysis of Commercial Data

To verify the effect of size refinement in suppressing the reduction of coke strength by long permeation distance coal at a commercial plant scale, the operational data of Fukuyama 3 coke oven (number of chambers: 104, W 0.43 × H 6.5 × L 15.4 m) was analyzed. At that plant, several coals are first blended through a coal-bin-blending system and crushed by crushers to prepare the coal blend. The particle size of the coal blend is also controlled to a target value by using the weight ratio of 3 mm or less of the coal blend (−3 mm (wt%)) as a control index. The data analyzed here were the drum strength of the coke after CDQ treatment from November 2010 to September 2013. The data were narrowed to the range of certain coal properties (R₀: 1.00 to 1.05%, log MF: 2.3 to 2.7 ddpm/log) and carbonization conditions (gross coking time: 18 to 21 h) in order to exclude effects other than the maximum permeation distance and particle size, and the influences of the weighted average maximum permeation distance and particle size of the coal blend on drum strength were analyzed. Coal blend containing higher blending ratio of long permeation distance coal has the longer value of weighted average maximum permeation distance basically. The stratification conditions and basic statistical information of the analysis data are listed in Table 6.

5.2. Results and Discussion

The drum strength classified under the conditions of maximum permeation distance and particle size is shown in Fig. 10. When the coal blend with a longer maximum permeation distance (at least 9.0 mm) and one with a shorter distance (less than 6.5 mm) were compared, the drum strength of the coal blend with the long maximum permeation distance was lower under all particle size

| Weighted averaged maximum permeation distance | Number of data | Mean value of weighted averaged maximum permeation distance (mm) | Mean value of −3 mm (wt%) |
|---------------------------------------------|---------------|---------------------------------------------------------------|-------------------------|
| < 6.5 mm < 75 wt%                            | 45            | 5.8                                                           | 72.5                    |
| < 6.5 mm 75 wt% ≤                            | 26            | 5.7                                                           | 78.0                    |
| 9.0 mm ≤ 75 wt%                              | 19            | 9.5                                                           | 73.1                    |
| 9.0 mm ≤ 75 wt% ≤                           | 34            | 9.8                                                           | 77.7                    |

Fig. 10. Relationship of weighted average maximum permeation distance, coal size (−3 mm) and drum index at actual plant. (Online version in color.)
conditions. When the particle size was −3 mm < 75 wt%, the drum strength difference in a t-test using a two-sided 95% confidence interval against two levels was statistically significant. When the relationship between the particle size and drum strength was compared under the condition of the same maximum permeation distance, drum strength was high in the case of −3 mm ≥ 75 wt% and the fine particle side. Under the same maximum permeation distance condition, the difference in drum strength between groups with different particle sizes was statistically significant. In particular, the coke strength of the coal blend having a long maximum permeation distance was significantly higher in the fine grain case than in the coarse grain case. Therefore, the same tendency as in the laboratory test results was observed in actual operation. That is, the analysis of actual operational data confirmed that the impact of the maximum permeation distance on coke strength changed depending on the particle size. Moreover, deterioration of coke strength could be suppressed by using a fine particle blend, even though the coal blend had a long maximum permeation distance. In conclusion, even if drum strength is reduced by blending a long permeation distance coal and the maximum permeation distance of the coal blend becomes longer, coke strength deterioration can be suppressed by reducing the particle size of the whole coal blend.

6. Effective Utilization Methods for Long Permeation Distance Coal

Based on the test results and discussion presented above, practical application methods for particle size control which enable effective utilization of long permeation distance coal were summarized, as shown in Table 7 and the following.

6.1. Method of Overall Crushing

The method of overall crushing to refine the whole coal blend, as in Test 2 and the commercial data analysis, does not require any special equipment. It is sufficient to set the target crushed particle size of the fine particles using the crushing and particle size adjustment equipment line that is normally installed in any coke plant. However, if the amount of fine particles in the coal blend is increased, there are concerns regarding adverse effects such as environmental problems due to an increase in dust generation and an increase in operation trouble due to carbon growth on the oven chamber walls. Hence, overall crushing is an applicable method provided the whole particle size is set in an appropriate range considering the degree of adverse effects.

6.2. Method of Selective Crushing of Long Permeation Distance Coal Itself

The method of selective crushing of the long permeation distance coal itself, shown in Tests 3 and 4, requires special equipment which can crush coal according to the brand when it is applied in a coke plant. However, it is possible to enhance the crushing of only the long permeation distance coal or to mitigate the crushing of other coal brands instead of refining the long permeation distance coal. Therefore, it is possible to minimize the grain size reduction of the whole coal blend and avoid the above-mentioned adverse effects of size refinement. In order to coarsen the grain size of some coal brands, it is important to select the brands properly. In Test 4, instead of size refinement of the long permeation distance coal (Coal C2), non-long permeation distance coal (Coal H2) having relatively medium properties was coarsened to maintain the particle size of the whole coal blend. This action contributed to enhancing coke strength. As described above, it is speculated that the effect of the deterioration of homogeneity can be minimized by reducing pulverization of coal brands whose properties are close to the average of the coal blend. On the other hand, the effect of suppressing defect generation also exists in the size refinement operation. However, there have been limited examples of attempts to systematically arrange the effects by coal brand and to organize the effects in isolation from homogenization effects. Thus, many issues concerning evaluation of these effects remain unsolved. In other words, when considered comprehensively, it is not clear at present how to optimize the particle size of each coal brand comprising a coal blend. Ideally, it is desirable to organize and rank the effects of refining and coarsening of particles on coke strength by coal brand and blending condition. It is also preferable to select a coal brand that results in a finer or coarser particle size. This point is an issue for future study. If it is difficult to introduce brand-specific crushing equipment in a coke plant, a good alternative might be pretreatment to refine the size of long permeation distance coal before delivery to the coke plant, for instance at the coal mine. In this way, the particle size of the long permeation distance coal can be made relatively fine at the coke plant if the particle size of the whole coal blend is managed as usual. As a result, it is conjectured that deterioration of coke strength by long permeation distance coal can be alleviated.

As described above, this study has clarified the fact that the reduction in coke strength associated with the use of long permeation distance coal can be suppressed by size refinement by various particle size adjustment methods, even in the case of long permeation distance coal that causes a reduction in coke strength. These findings are expected to
contribute to effective utilization of long permeation distance coal and production of high strength coke using long permeation distance coal.

7. Conclusions

For the development of effective utilization techniques for coal with a long maximum permeation distance (long permeation distance coal), which causes deterioration of coke strength, the influence of the particle size of long permeation distance coal on maximum permeation distance and coke strength were investigated. As a result, the following findings were obtained.

1) The particle size of coal was the controllable and influencing factor for the maximum permeation distance. It was confirmed that the maximum permeation distance was reduced by decreasing the coal size.

When long permeation distance coal was included in a coal blend, coke strength was improved by refining the whole coal blend including the long permeation distance coal or refining only the long permeation distance coal. This research implied that the negative effect of long permeation distance coal on coke strength was suppressed not only by the homogenization effect due to the uniform dispersion of the coal itself but also by decreasing the generation of defects from the coal.

2) This study demonstrated that coke strength can be maintained while using long permeation distance coal in a commercial plant when the coal blend containing the long permeation distance coal is refined into fine particles.

3) Based on the findings of experiments and analysis of operational data from a commercial plant, methods of coal particle size adjustment were proposed for practical effective utilization of long permeation distance coal.

REFERENCES
1) JIS M 8801: 2008, Coal Testing methods.
2) Y. Miura, T. Okuhara, T. Nishi, T. Yamaguchi and H. Haraguchi: Trans. Iron Steel Inst. Jpn., 21 (1981), 518.
3) Y. Dohi, K. Fukuda, T. Yamanoto, T. Matsui, H. Sumi and I. Shimoyama: ISIJ Int., 54 (2014), 2484.
4) Y. Sunami, S. Ogawa, N. Oshikuri and Y. Okuda: Tetsu-to-Hagané, 66 (1980), 571 (in Japanese).
5) T. Fukuyama, T. Miyazu and S. Kimoto: J. Fuel Soc. Jpn., 51 (1972), 628.
6) R. Loison, P. Foch and A. Boyer: Coke, Butterworths, London, (1989), 384.
7) Y. Saito, S. Matsuo, T. Kanai, A. Toishi, A. Uchida, Y. Yamazaki, Y. Matsushita, H. Aoki, S. Nomura, H. Hayashizaki and S. Miyashita: Tetsu-to-Hagané, 100 (2014), 140 (in Japanese).
8) H. Suginohe and T. Miyagawa: Tetsu-to-Hagané, 68 (1982), 2133 (in Japanese).
9) T. Okuhara: Tetsu-to-Hagané, 73 (1987), 1846 (in Japanese).
10) E. Burstlein: Glückauf, 92 (1956), 606.
11) Y. Miura, T. Yamaguchi, T. Nishi and Y. Yone: J. Fuel Soc. Jpn., 60 (1981), 771.
12) S. Asada, M. Nishimura and T. Nojima: J. Jpn. Inst. Energy, 73 (1994), 1060.
13) Y. Kubota, S. Nomura, T. Arima and K. Kato: Tetsu-to-Hagané, 92 (2006), 833 (in Japanese).
14) N. Schapiro, R. J. Gray and G. R. Ensner: Proc. Blast Furnace, Coke Oven, and Raw Materials Conf., Metallurgical Society of AIME, New York, (1964), 89.
15) JIS M 8816: 1992, Solid mineral fuels—Methods of microscopical measurement for the macerals and reflectance.
16) JIS M 8812: 2006, Coal and coke—Methods for proximate analysis.
17) JIS K 2151: 2004, Coke Testing methods.
18) R. Loison, P. Foch and A. Boyer: Coke, Butterworths, London, (1989), 127.
19) Y. Kubota, S. Nomura, T. Arima and K. Kato: Tetsu-to-Hagané, 96 (2010), 328 (in Japanese).
20) K. Ueoka, T. Ogata, Y. Matsushita, H. Aoki, T. Miura, K. Fukuda and K. Matsuda: Tetsu-to-Hagané, 93 (2007), 728 (in Japanese).
21) K. Kato: Bull. Iron Steel Inst. Jpn., 9 (2004), 810.
22) T. Nakagawa, T. Suzuki, A. Furusawa, Y. Maeno, I. Kornaki and K. Nishikawa: Fuel, 77 (1998), 1141.