New Evaluation and Blending Technique of Coking Coal Based on Surface Tension and Permeation Distance Measurement

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Abstract:

JFE Steel has developed a new coal evaluation and blending technique named "Advanced MOF diagram system". The new system is composed of conventional MOF diagram, which is a coal evaluation and blending technique based on coal rank (R̄o) and maximum fluidity (MF), and novel evaluation indices, "surface tension" and "permeation distance". While the MF is well known as the most important caking property, the evaluation is sometimes ineffective especially in the low and high fluidity range. To improve the evaluation of the thermoplasticity, two measurement approaches were devised: "surface tension" which influences adhesion strength among coal grains and "permeation distance" which affects the pore-wall formation. The new approaches enable us to evaluate the thermoplasticity and coking property of the low and high MF coals with high sensitivity. Thanks to this development, JFE Steel has successfully achieved efficient coal utilization and improvements in control of coke quality using the new technique.

1. Introduction

In recent years, the importance of rational selection and blending of the coal brands used in cokemaking has increased not only because of the sharp rise of coal price, but also the higher coke strength required in blast furnace. Conventionally, the MOF diagram system[1] has been used widely in control of coke strength. In this system, the mean maximum reflectance of vitrinite (R̄o, JIS M 8816) is used as an index of the coal rank, and Gieseler maximum fluidity (MF, JIS M 8801) is used as an index of the caking property. The optimum range of MF is 200 to 1 000 ddpm. Since coke strength tends to decrease when MF is outside this range, the development of techniques which make it possible to control coke strength to a high level when using low MF coal (MF ≤ 200 ddpm) and high MF coal (MF ≥ 1 000 ddpm) had been desired.

Because low MF coal is characterized by inadequate thermoplasticity, it is thought that fusion bonding decreases and adhesive bonding becomes the controlling factor for the bonding state between coal grains during carbonization[2]. However, the effect of adhesive bonding between coal grains has been rarely considered in conventional approaches. In addition, the degree of the decrease in coke strength also varies depending on the "combination" of coal brands that comprise a coal blend[3] (this is known as the compatibility effect), but the cause of this phenomenon had not been sufficiently clarified.

The authors considered the adhesiveness of coal to be an important factor in coke strength, particularly in the low MF region. We also conjectured that the above-mentioned compatibility effect, in which the degree of the decrease in coke strength differs depending on the combination of coal brands, originates from differences in adhesion strength between coal grains. Since it is generally known that surface tension affects adhesion strength in adhesive bonding between various types of materials, we developed a technique for controlling coke strength based on the surface tension of

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heat-treated coal feedstock[4].

On the other hand, even with high MF coal, it is difficult to control coke strength stably to a high level. Because high MF coal curls up on the stirring rod of the Gieseler plastometer during measurement, which sometimes causes the stirring rod to run idle[5], the reliability of the MF determined by this measurement method is regarded as questionable. Thus, in control of the coke strength of coal blends containing high MF coal, there are considered to be cases where MF is not an effective index of the caking property.

In response to this problem, we developed a permeation distance measurement method which can accurately evaluate the thermoplasticity of coals, including high MF types, and a control technique in coke strength based on permeation distance[6].

This report summarizes the knowledge obtained through research and development in connection with surface tension and permeation distance, and introduces a newly-developed evaluation and blending technique for coking coal, which is named "Advanced MOF diagram system."

2. Measurement of Surface Tension

2.1 Experimental

In view of the difficulty of in situ measurement of the surface tension of softening coals during heating, the surface tension of semi-coke obtained by heat treatment of coal at 500°C was measured by the film floatation method[7]. Air-dried coal was crushed to under 200 μm, charged into a crucible at a bulk density of 800 kg-dry/m^3, and heated to 500°C at 3°C/min, followed by rapidly cooled with liquid nitrogen and collected as semi-coke sample. The sample was then crushed to under 150 μm and sprinkled on the surface of each ethanol aqueous solution with ethanol concentration of 0 to 75 wt%. The surface tension of the aqueous solution is varied due to the differences in the concentration. Because the sample either floats or sinks depending on the magnitude relationship of the surface tension of the ethanol solution and the sample, only the floating semi-coke grains were collected. Then their weight was measured after drying.

Figure 1 shows an example of the frequency distribution of the measured surface tension. The weighted average value γ of this distribution was used as the representative value of the surface tension of each coal. The measured values of γ for various coals were roughly in the range of 37 to 45 mN/m.

Next, in order to evaluate the effect of the surface tension difference ($\Delta\gamma$) of different types of coals on the interfacial structure between the coals, two-layered
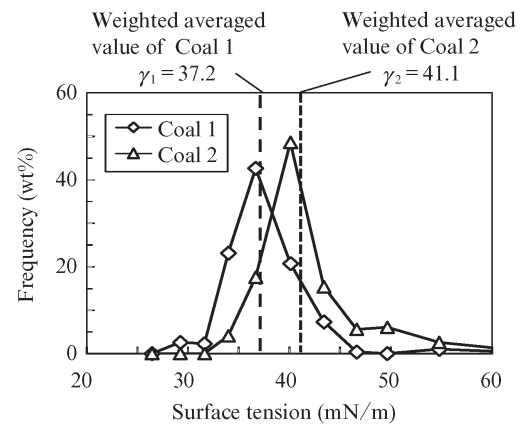

Fig. 1　An example of measured surface tension distribution

Table 1　Properties of coal used in two-layered tablet sample

Coal	$\overline{R}o$ (%)	log MF (log ddpm)	γ (mN/m)	$\Delta\gamma$ to coal R1 (mN/m)
R1	0.73	2.37	40.9	(Reference)
A1	1.28	1.23	40.8	0.1 (Small)
A2	1.23	0.6	38.4	2.5 (Large)

tablet samples with different coals arranged in the upper and lower layers were prepared and carbonized, and their interfacial structures were observed. **Table 1** shows the properties of the coals used. First, combinations of coals with small $\Delta\gamma$ (R1 and A1) and large $\Delta\gamma$ (R1 and A2) were selected. After air-drying, the respective coals were then crushed to under 200 μm. The mold was filled with the lower-layer coal (A1 or A2), followed by the upper-layer coal (R1), and a load was applied to produce a two-layer tablet sample (φ 6.7 mm×H 2.3 mm). The tablet sample and coke breeze as an oxidation suppressant were placed in the carbonization vessel (W 200 mm×L 200 mm×H 70 mm) and carbonized by heating to 1 000°C at 3°C/min. After cooling, the vertical cross section of the obtained coke was observed with a microscope.

The relationship between the surface tension difference ($\Delta\gamma$) and adhesion strength of the coke originating from the binary coal blends was also investigated. First, single coal samples air-dried and crushed to under 75 μm and two types of binary coal blends, consisting of a combination of coals (R2 and A3) having low $\Delta\gamma$ and a combination (R2 and A4) with large $\Delta\gamma$, were prepared. These binary blend samples were mixed at a weight ratio of 50 : 50 after crushing the coals to under 75 μm. The properties of the coals used and the blending conditions of the binary coal blends are shown in **Table 2** and **Table 3**, respectively. The coal samples were molded into tablets (φ 6.7 mm×H 2.3 mm). 10 tablet samples were prepared for all the blending conditions, and arranged in the carbonization vessel with coke

Table 2 Properties of coal that used in binary coal blends

Coal	$\overline{R}o$ (%)	log MF (log ddpm)	γ (mN/m)
R2	1.62	1.28	37.8
A3	1.07	2.09	38.9
A4	1.00	2.43	39.6

Table 3 Blending condition of binary coal blends

Sample No.	Combination of coals		$\overline{R}o$ (%)	log MF (log ddpm)	$\Delta\gamma$ (mN/m)
1	R2	A3	1.35	1.68	1.1
2	R2	A4	1.31	1.86	1.8

Table 4 Properties of used coal and blending condition of multi-coal blends

Coal	$\overline{R}o$ (%)	log MF (log ddpm)	γ (mN/m)	Note	Coal	Blending ratio (wt%)		
						BLEND 1	BLEND 2	BLEND 3
H1	0.65	4.11	41.4	Higher γ	H1	3	18	22
B1	0.85	3.13	40.9	Middle γ	B1	20	17	
B2	0.75	1.82	40.6		B2	12		2
B3	1.29	1.04	40.6		B3	25	15	15
B4	0.98	2.88	40.1		B4			5
B5	1.07	3.18	40.1		B5	40		
L1	1.1	2.88	39.5	Lower γ	L1		10	
L2	1.1	2.03	39.4		L2		40	40
L3	1.06	3.21	38.9		L3			16
					$\overline{R}o$ (%)	1.01	1.01	1.01
					log MF (log ddpm)	2.35	2.35	2.35
					$\sigma\gamma$ (mN/m)	0.35	0.82	0.91

(a) Sample No. 1 ($\Delta\gamma = 0.1$)

Coal R1 (γ: 40.9) Interface
Coal A1 (γ: 40.8)
500 µm

(b) Sample No. 2 ($\Delta\gamma = 2.5$)

Coal R1 (γ: 40.9) Interface
Coal A2 (γ: 38.4)
500 µm
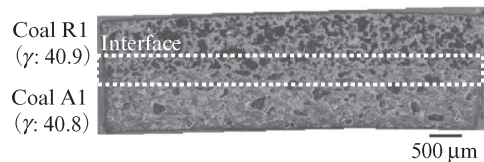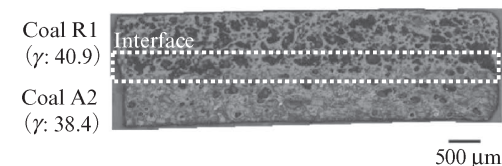
Fig. 2 Interfacial structure of two-layered tablet samples

breeze and heated to 1 000°C at 3°C/min. The compressive strength of the recovered samples was measured by compressing the tablet samples in the sample height direction with a universal tester (Shimadzu Corporation, AG-I 50 kN) at a pressing speed of 1 mm/min. This test was conducted 10 times at each level, and the average value was calculated.

Next, a carbonization test and drum strength index test were carried out to clarify the relationship between the surface tension and coke strength of multi-coal blends which are able to be used commercially in coke plants. The properties and blending conditions of the coal samples are shown in **Table 4**. The coal samples were crushed to under 3 mm and adjusted to a moisture content of 8 wt%. The standard deviation $\sigma\gamma$ of the surface tension of the coal blends was adjusted by changing the composition of the blends from middle γ coals (coals B1 to B5) to either a high γ coal (coal H1) or low γ coal (coals L1 to L3). It is thought that this operation changes the ratio of the interface with different $\Delta\gamma$. The conventional indices of the coal blend (weighted average $\overline{R}o$, logMF) were constant. The prepared coal blends were filled in a carbonization vessel (W 273 mm×L 260 mm×H 300 mm) at a bulk density of 750 kg-dry/m^3, and were then carbonized in an electric furnace controlled to a furnace wall temperature of 1 100°C. After cooling, the drum strength index (DI$^{150}_{15}$, JIS K 2151) of the obtained coke was measured.

2.2 Results and Discussion

Figure 2 shows the microstructure of the interface between two coal layers of the carbonized samples with a surface tension difference ($\Delta\gamma$) of 0.1 mN/m (Sample
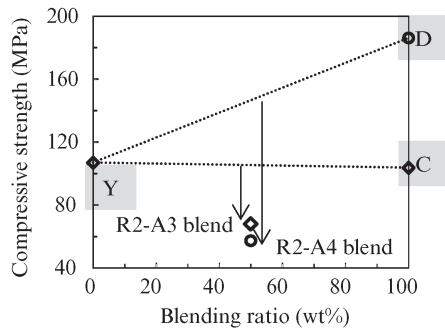
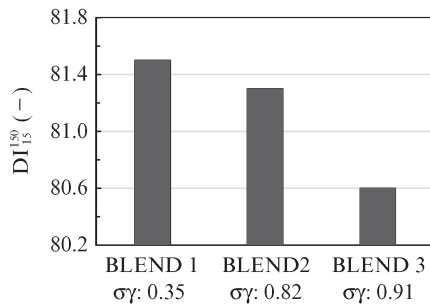Fig. 3   Compressive strength of coke from binary coal mixtures

Fig. 4   Effect of standard deviation of surface tension ($\sigma\gamma$) on coke strength

Fig. 5   Schematic diagram of measurement apparatus for permeation distance

No. 1) and $\Delta\gamma$ of 2.5 mN/m (Sample No. 2). In Sample No. 1, a strongly adhered structure with good wetting at the interface was observed. In contrast, Sample No. 2 displayed the formation of large pores, which were considered to be caused by poor wetting in the vicinity of the interface. From these results, it is considered that adhesion failure is induced by large $\Delta\gamma$ of the interface between dissimilar coals.

The compressive strength of the coke derived from the single coals and binary coal blends is shown in **Fig. 3**. The weighted average values were calculated from the strength of the single coals (Y, C and D in the figure; compressive strength of coke derived from R2, A3 and A4 in that order) and the blending ratio. The actual strength of the coke derived from the coal blends was lower than the weighted average value. In addition, in the case of coal blend with large $\Delta\gamma$ (combination of coals R2 and A4), the measured value of coke strength decreased remarkably in comparison with the weighted average value. This result is thought to be caused by breakage in the area where adhesion failure generated at the interface between the coals with large $\Delta\gamma$.

Figure 4 shows the relationship between the standard deviation $\sigma\gamma$ of the surface tension of the multi-coal blends and the drum strength index. The drum strength index decreased as $\sigma\gamma$ increased. As $\sigma\gamma$ increases, the number of interfaces with comparatively
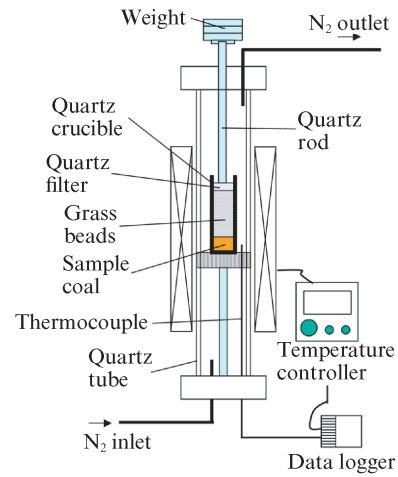
large values of $\Delta\gamma$ also increases, suggesting that the drum strength index decreases due to an increase in defects that become sites of adhesion failure.

The results described above clarified the fact that the surface tension of semi-coke is a key factor that affects the adhesion property and drum strength index of coke. Furthermore, although the compatibility effect due to the "combination" of coals used in coal blends was difficult to detect by the conventional method, this experiment also demonstrated that the compatibility effect can be evaluated by $\sigma\gamma$.

3. Measurement of Permeation Distance

3.1 Experimental

A schematic diagram of the apparatus developed for measurement of the permeation distance is shown in **Fig. 5**. This apparatus simulates the phenomenon of permeation of thermally plastic coal grains in a coke oven into the surrounding voids during softening and melting of the coal. A layer of glass beads simulating the permeation region was arranged above the sample coal, and pressure was applied from above with a weight, assuming the condition of constraint that exists in a coke oven. The sample coal was then heated, resulting in a thermally plastic condition, and the coal permeated into the glass bead layer above it. The permeation distance was measured as an evaluation index. For details of this measurement method were reported in a previous paper by the authors[6].

Figure 6 shows the relationship between the permeation distance and log MF for various types of coal. Although the permeation distance shows a rough positive correlation with log MF, large deviations were observed when using high MF coals. Based on these
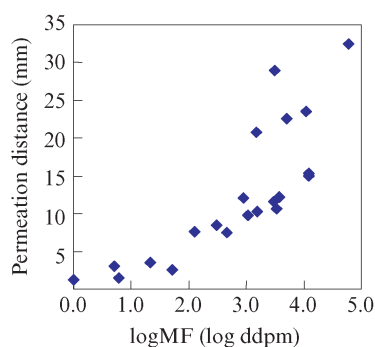
Fig. 6 Comparison between permeation distance and MF

Table 5 Blending condition of coal blends

	$\overline{Ro}$ (%)	log MF (log ddpm)	High MF coal conditions		
			Brand	Blending ratio (wt%)	Permeation distance (mm)
Blend 1	0.99	2.22	Coal A	20	12.2
Blend 2	0.99	2.17	Coal B	20	20.8
Blend 3	0.99	2.14	Coal C	20	22.6
Blend 4	0.99	2.20	Coal A Coal B	10 10	16.5

Table 6 Carbonization test condition

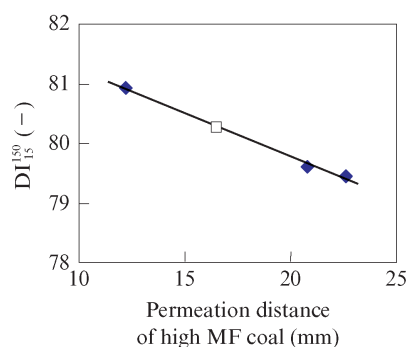| Coal size (wt%)	− 3 mm: 100
Moisture content (wt%)	8
Bulk density (kg-dry/m^3)	750
Dimensions (mm)	W 250 × H 600 × L 413
Wall temperature (°C)	1 050
Coking time (min)	500

Fig. 7 Effect of permeation distance of high MF coal on coke strength (Blend 1-4)
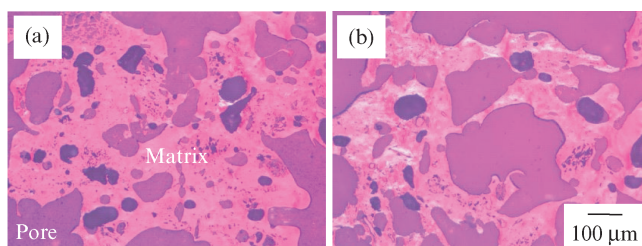

Fig. 8 Polarizing microscope image of coke structure ((a) Blend 1, (b) Blend 2)

results, it is cleared that the permeation distance is a unique parameter that can detect deviations of high MF coal, which is not possible by the conventional evaluation method.

Next, the effects of the permeation distance of high MF coal on the coke strength and structure were investigated. The blending conditions of the coal blends are shown in **Table 5**. In Blends 1 to 3, high MF coals (Coals A, B, C) having different permeation distances were blended with other coal so that the ratio of the high MF coal in the coal blend was 20 wt%. Blend 4 contained 10 wt% each of Coal A and Coal B. The prepared coal blends were carbonized in an electric furnace under the conditions shown in **Table 6**, and the resultant coke was cooled in nitrogen and recovered. A drum strength index test was conducted using the recovered coke. In addition, samples of the coke prepared from Blends 1 to 3 were embedded in resin and polished, and micrographs of the polished surface were

taken with an optical microscope at a magnification of 100 times. In order to quantify the pore-wall structure of the coke, the thickness of the pore-walls in the micrographs was measured using an image analysis software.

3.2 Results and Discussion

Figure 7 shows the relationship between the permeation distance of the high MF coal contained in the coal blends and coke strength for Blends 1 to 4. As the permeation distance of the high MF coal increased, the coke strength after carbonization decreased. **Figure 8** shows polarizing microscopic images of Blend 1 and Blend 2. In these observation results, the pore-wall thickness of the coke obtained from Blend 2, which contained the coal with a long permeation distance Coal B (Fig. 8 (b)) was thinner than that of the coke obtained from the short permeation distance Coal A (Fig. 8 (a)). **Figure 9** shows the relationship between the

permeation distance of the high MF coal and the mean pore-wall thickness. Long permeation distance coal results in a thin pore-wall thickness, suggesting that the coke structure is weak.

Figure 10 shows the differences in the formed pore-wall structure and the estimated formation behavior due to differences in the permeation distance. As shown in Fig. 10 (b), it is thought that the long permeation distance coal permeates in voids and gaps between the filled grains during heating, a thin pore-wall is formed when permeation is complete. Moreover, since large pores remain where grains had originally existed, these pores behave as defects and coke strength decreases. On the other hand, as shown in Fig. 10 (a), the short permeation distance coal displays less permeation in voids and gaps than the long permeation distance coal. As a result, a comparatively thick pore-wall is formed and the formation of large pores is suppressed. According to past studies investigating the relationship between coke strength and the coke pore structure, coarse pores with a mm-scale diameter especially behave as defects and reduce coke strength [8–10]. In addition, it has also been reported that coke strength decreases as the pore-

wall thickness becomes thinner [11]. Based on these facts, it is inferred that the permeation distance affects coke strength as a result of the pores formed after permeation and the difference in the pore-wall structure.

As described above, this study showed that the permeation distance is a useful index for detecting not only differences in the thermoplasticity of high MF coal but differences in the effect on coke strength, which cannot be evaluated by conventional MF.

4. Advanced MOF Diagram System

The authors integrated the knowledge gained from our research and development on surface tension and permeation distance measurement in the conventional MOF diagram system, and newly proposed the "advanced MOF diagram system" shown in **Fig. 11**. The MOF diagram system is a technique in which coke strength is controlled based on the weighted average $\overline{Ro}$ and logMF of coal blends. However, in addition to these average properties of the coal blend, the advanced MOF diagram system considers the effects of the properties of the individual coal brands on coke strength. The concept of the proposed system was arranged as follows.

(1) Coke strength is affected not only by the maximum fluidity (MF) of the coal, but also by wettability or adhesion strength. The adhesion strength between two coal brands can be evaluated by the difference in the surface tension ($\Delta\gamma$) of the two coal brands after heat treatment. If low MF coal is used in a coal blend, the thermoplasticity of the entire coal blend decreases, and bonding between coal grains is controlled by simple adhesive bonding rather than by fusion bonding. In this case, adhesion strength has a more sensitive effect on coke strength. Accordingly, when using a coal blend containing low MF coal, the decrease in coke strength
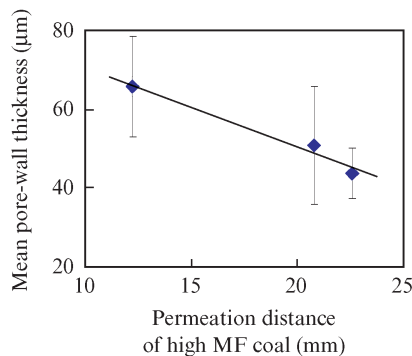

Fig. 9 Effect of permeation distance of high MF coal on mean pore-wall thickness (Blend 1-3)

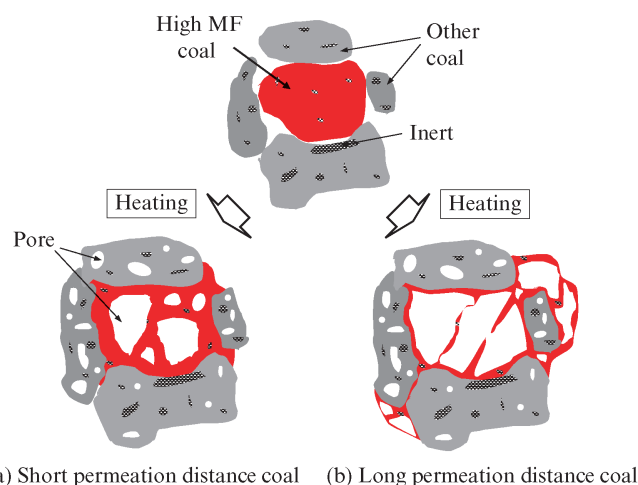(a) Short permeation distance coal   (b) Long permeation distance coal

Fig. 10 Schematic illustration of pore-wall formation behavior of coal with short or long permeation distance
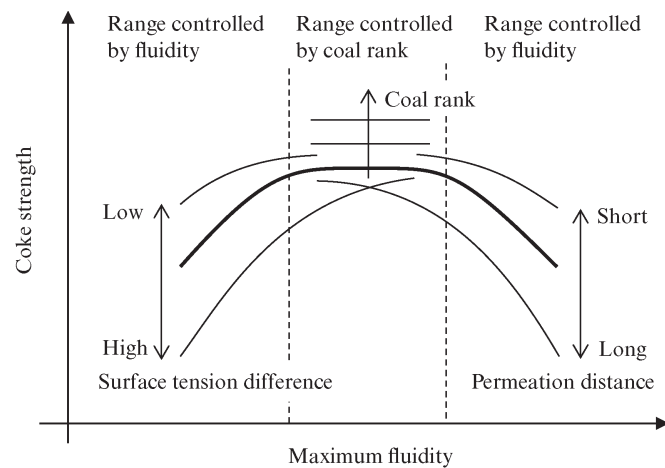
Fig. 11 Conceptual diagram for advanced MOF system, including effects of surface tension and permeation distance

can be minimized by reducing the difference in the surface tension of the constituent coals of the coal blend in order to increase its adhesion strength.

(2) Coke strength is reduced if a large ratio of coal with an excessively long permeation distance is used. Moreover, strength of coke made from a coal blend with a high weighted average MF is sometimes lowered possibly because the coal blend includes a large amount of coal with a long permeation distance, which has generally also high MF. Therefore, the decrease in coke strength can be minimized by reducing the use ratio of long permeation distance coal.

Based on the concept of the advanced MOF diagram system, the method for selecting coal brands for cokemaking can be organized as follows.

(3) When considering the constituent coal brands of a coal blend, coke strength is increased by selecting coals with a small difference in surface tension. In other words, a suitable combination of coals for cokemaking can be found by using surface tension as an index. When selecting a new coal brand for use, it is desirable to determine the average surface tension of the coal blend which is currently being used, and to select a coal that is close to that surface tension.

(4) In case a high MF coal is selected, assuming the other coal qualities are the same, a coal with a relatively short permeation distance should be selected for production of high quality coke.

5. Conclusion

The selection of coal brands for use and the coal blending design have become more rational thanks to the new coal evaluation and blending technique intro-duced in this report, contributing to effective use of coal resources and more advanced control of coke strength.

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