Effect of Coal Briquette Size on Coke Quality and Coal Bulk Density in Coke Oven

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1. Introduction

One of the important technical subjects in blast furnace cokemaking is deterioration of hard coking coal quality. In order to cope with this, various technologies to utilize semi-soft coking coals by increasing coal charge bulk density in slot-type coke ovens have been developed or commercialized such as dry coal charging process (CMC (Coal Moisture Control)) and DAPS (Dry-cleaned and Agglomerated Precompaction System)), briquette blending carbonization process and stamp charging process.

After the successful commercialization of briquette blending carbonization process in Japan, the development of a formed coke process (FCP) was carried out as a national project in 1980s. In the former process, briquettes are mixed with powder coal and charged into a conventional slot-type coke oven chamber, while in the latter process (FCP) briquettes are carbonized continuously in a vertical shaft furnace. A combined effect of the size and type of coal/binder feedstock on coke strength has been reported by several authors. However, the briquette blending carbonization process had been gradually replaced by dry coal charging process and FCP was not commercialized, coal briquetting technology was less attractive in the field of cokemaking technology.

Now coal briquetting has been considered important as a useful technology for recycling low-value and carbon-containing wastes and for increasing the amount of semi-soft coking coals in the blends. Diez et al. investigated the partial briquetting of carbon-containing wastes from steelmaking such as coal-tar sludges, deposits and oily wastes for metallurgical coke production and showed that the coke quality parameters did not show any significant deterioration when an optimum condition was chosen.

Briquettes from coke breeze and anthracite and brown coal were studied from the view point of metallurgical coke production. Montiano et al. prepared briquettes using sawdust, a non-coking coal and the binder (coal tar and coal-tar sludge) and studied the effect of coal briquette size on coke quality by using a larger briquette (23 g) and a smaller one (4–6 g). They mixed the briquettes with blended coal by 5–15% and showed that the small briquette decreases coke cold strength (DI15015) less, increases coke reactivity (CRI) more and decreases coke strength after reaction (CSR) more than the large one.

It is considered that the briquette size affects both coal charge bulk density and coke quality; however, there have been no reports on the effect of coal briquette size on coke quality by using briquettes made of normal blended coals for blast furnace coke making (hard coking coals and semi-soft coking coals). Therefore, we studied the effect of coal briquette size on coke quality and bulk density in coke oven by using briquettes made of normal blended coals.

2. Experiment

2.1. Coal Sample

Four coals as shown in Table 1 were used in the experiment and four blended coals were prepared as shown in Table 2. Coals A, B and C are hard coking coals and Coal D is a semi-soft coking coal with low total dilatation of 28%. Blend 4 is a blended coal used in a commercial cokemaking plant. Coal tar pitch (abbreviated as pitch) was used as binder for briquetting. The softening point of the pitch measured by the ring and ball method was 25°C and the insoluble fraction in quinoline and toluene was 8% and 17% respectively.

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2.2. Powder and Briquette

Since coke strength is dependent on specific dilatation volume (abbreviated as SV) which is similar to total dilatation (TD) measured with dilatometry, it is interesting to change the SV of coal briquette and powder coal by changing constituting coal blends, coal grind and addition ratio of pitch in order to have deep insight into the effect of briquette size on coke quality.

In order to carbonize the mixture of powder coal and coal briquette, powder coal and pre-briquette coal were prepared as shown in Table 3. Pre-briquette coal means the coal sample to be supplied to a briquetting machine to form briquettes. In test 1, powder coal was prepared by crushing blend 1 to −3 mm 85%. Powder coal means the crushed coal sample used for carbonization test as it is. Pre-briquette coal was prepared by crushing blend 1 to −1.5 mm and mixing that with pitch by 8%. Coal (with the moisture of about 3%) and pitch was mixed with a high-speed agitating mixer at 70–80°C, where steam was injected into the mixer during mixing. In test 2, powder coal was prepared by crushing blend 2 to −3 mm 85%. Pre-briquette coal was prepared by crushing blend 4 to −3 mm 85% and mixing −0.5 mm of crushed blend 4 with pitch by 12%. In test 3, powder coal was prepared by crushing blend 3 to −3 mm 85%. Pre-briquette coal was prepared by crushing blend 3 to −1.5 mm and mixing that with pitch by 8%. In test 4, blend 3 was crushed to −3 mm 85% and +0.3 mm of crushed blend 3 was used as powder coal. Pre-briquette coal was prepared by mixing −0.3 mm of crushed blend 3 with pitch by 10%

Then pre-briquette coal was briquetted using a roll press briquetting machine. The size of the briquette was 64 mm × 42 mm × 32 mm and its volume was 56 cm³. In order to prepare briquette with different sizes, briquettes with 56 cm³ were crushed and sieved with 0.6 mm, 2 mm, 6 mm, 15 mm and 40 mm sieves. All these samples are called briquettes in this study even though they are as small as 0.6 to 6 mm. The briquettes used in the experiment are shown in Table 3. In test 2, briquettes having different volumes (4.7 cm³

### Table 1. Characteristics of the coals used in the experiment.

<table>
<thead>
<tr>
<th>Coal</th>
<th>Proximate Analysis</th>
<th>Ruhr Dilatometry</th>
<th>Gieseler Plastometry</th>
<th>Petrographic Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>VM (% db)</td>
<td>Ash (% db)</td>
<td>Total Dilatation (%)</td>
<td>Log₁₀ (Max. Fluidity/ddpm)</td>
</tr>
<tr>
<td>A</td>
<td>21.3</td>
<td>9.4</td>
<td>22</td>
<td>1.38</td>
</tr>
<tr>
<td>B</td>
<td>24.4</td>
<td>8.9</td>
<td>134</td>
<td>3.00</td>
</tr>
<tr>
<td>C</td>
<td>36.8</td>
<td>9.4</td>
<td>233</td>
<td>4.39</td>
</tr>
<tr>
<td>D</td>
<td>36.6</td>
<td>9.3</td>
<td>28</td>
<td>2.32</td>
</tr>
</tbody>
</table>

### Table 2. Characteristics of the blended coals.

<table>
<thead>
<tr>
<th>Blend</th>
<th>Coal Proximate Analysis</th>
<th>Ruhr Dilatometry</th>
<th>Total Dilatation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blend 1</td>
<td>80%</td>
<td>20%</td>
<td>26.8</td>
</tr>
<tr>
<td>Blend 2</td>
<td>30%</td>
<td>30%</td>
<td>28.4</td>
</tr>
<tr>
<td>Blend 3</td>
<td>30%</td>
<td>30%</td>
<td>28.4</td>
</tr>
</tbody>
</table>

### Table 3. Test conditions (powder coal and briquette).

<table>
<thead>
<tr>
<th>Test 1</th>
<th>Test 2</th>
<th>Test 3</th>
<th>Test 4</th>
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<tbody>
<tr>
<td>Powder</td>
<td>Coal blend</td>
<td>Blend 1</td>
<td>Blend 2</td>
</tr>
<tr>
<td></td>
<td>Coal grind</td>
<td>−3 mm 85%</td>
<td>−3 mm 85%</td>
</tr>
<tr>
<td></td>
<td>Specific dilatation volume (cm³/g)</td>
<td>1.88</td>
<td>2.24</td>
</tr>
<tr>
<td>Briquette</td>
<td>Coal blend</td>
<td>Blend 1</td>
<td>Blend 4</td>
</tr>
<tr>
<td></td>
<td>Coal grind</td>
<td>−1.5 mm</td>
<td>−0.5 mm</td>
</tr>
<tr>
<td></td>
<td>Pitch</td>
<td>8%</td>
<td>12%</td>
</tr>
<tr>
<td></td>
<td>Specific dilatation volume (cm³/g)</td>
<td>2.62</td>
<td>1.94</td>
</tr>
<tr>
<td></td>
<td>Briquette size</td>
<td>0.6–2 mm*</td>
<td>0.6–2 mm*</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2–6 mm*</td>
<td>2–6 mm*</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6–15 mm*</td>
<td>6–15 mm*</td>
</tr>
<tr>
<td></td>
<td></td>
<td>15–40 mm*</td>
<td>4.7 cm³</td>
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</tbody>
</table>

* prepared from 56 cm³ briquette
and 15 cm$^3$) were prepared by changing the cup size of the briquetting machine. The apparent density of the briquette determined by immersing briquettes in water was 1.18 g/cm$^3$ on average. Figure 1 shows examples of the briquettes.

SV (cm$^3$/g) is characterized by measuring the volume after dilatation per unit coal sample mass. In the experiment, 2.5 g of coal sample was charged as powder (without pulverizing coal sample and preparing pencil sample) in a dilatometer retort tube at the height of 60 mm and heated up to 550°C at the heating rate of 3°C/min as in dilatometry. The maximum height after dilatation was measured and SV was obtained. SV for powder coal and pre-briquette coal is shown in Table 3.

While SV for pre-briquette coal is larger than that for powder coal in test 1, it is the other way around in test 2. By comparing these two tests, the effect of the difference in SV between powder coal and pre-briquette coal was investigated. In tests 3 and 4, SV for powder coal and pre-briquette coal are as low as 1.51–1.52 and 1.71–1.84 respectively because blend 3 contains 40% of semi-soft coking coal. These tests are expected to reveal the effect of the briquette size on coke quality in using large amount of semi-soft coking coals.

2.3. Carbonization and Coke Quality Test

Powder coal and briquettes were mixed with the ratio of 70:30 and charged into the electrically heated test coke oven (420 mm in width, 610 mm in length, 400 mm in height) at the coal charge bulk density of 850 dry-kg/m$^3$. After carbonized for 18.5 hours under heating conditions equivalent to the flue temperature of 1250°C in an actual coke oven, the coke was quenched in nitrogen atmosphere and coke quality was measured as follows. Firstly, coke was dropped from 2 m height 3 times for stabilization, and JIS drum index of coke (DI$_{150}^{6–15}$: percentage of coke mass retained on 15 mm sieve to the original mass of the coke sample after 150 revolutions in the JIS drum tester) was measured following the standard JIS K2151. Also measured were CRI (Coke Reactivity Index: percentage of reacted coke mass to the mass of coke sample after 200 g of 20 mm coke samples were reacted with CO$_2$ at 1 100°C for 2 hours) and CSR (Coke Strength after Reaction: percentage of coke mass retained on 9.5 mm sieve to the mass of reacted coke sample after rotated 600 revolutions in the I-type drum tester).

2.4. Coal Charge Bulk Density Measurement

In order to investigate the effect of briquette size on coal charge bulk density, coal charge bulk density was measured for test 4 (mixture of powder coal (blend 3) and 4 different sizes of briquettes (0.6–2 mm, 6–15 mm, 15–40 mm and 56 cm$^3$) as a representative. 34 kg of the mixture of powder coal and briquettes was dropped from 2 m height into a cube box 305 mm on a side (inner volume 0.0283 m$^3$), which was placed on a weight scale. After the sample above the top surface was removed, the coal mass was measured without shaking the box and the coal charge bulk density was obtained by the coal mass in the box divided by the box volume.

3. Results and Discussion

3.1. Effect of Briquette Size on Coke Strength

The relationship between briquette size and coke strength are shown in Fig. 2. The briquettes size is expressed either as the average of the upper sieve size and the lower one or as equivalent sphere volume diameter D (mm), which is calculated by $2\times(3\sqrt[3]{V/4\pi})$×10, where V (cm$^3$) is the volume of the briquette.

DI$_{6–15}^{50}$ in Fig. 2(a) is the index which stands for surface breakage strength of coke. DI$_{6–15}^{50}$ is obtained by subtracting the percentage of −6 mm coke fines generated in drum test from 100. DI$_{6–15}^{50}$ is the index which stands for volume breakage strength of coke and is the percentage of 6–15 mm coke fines in drum test.22) The relationship between DI$_{6–15}^{50}$ and DI$_{6–15}^{150}$ is expressed as follows: DI$_{6–15}^{50}$ = DI$_{6–15}^{150}$ - DI$_{6–15}^{150}$.

Figure 2(b) shows that the relationship between briquette size and DI$_{6–15}^{150}$ varies dependent on the test condition. In test 1, DI$_{6–15}^{150}$ is obtained at the briquette size of 10 mm, in test 2 DI$_{6–15}^{150}$ increases with increasing the briquette size and in tests 3 and 4 DI$_{6–15}^{150}$ shows a minimum at the briquette size of 10 mm.

The reason for this can be explained by a model considering both effects of briquette swelling and pitch dispersion shown in Fig. 3. It is known that the larger the coal particle size is, the more it swells.23) This is considered because the larger the coal particle size is, the longer the length of the path for the gas generating in thermoplastic coal particles to escape becomes. This increases the amount of gas trapped in the molten coal particles, which helps to enhance swelling. In the case of briquettes, it is considered similarly that the larger the briquette size is, the more it swells. The swelling of briquettes compresses the powder coal surrounding the briquette in a thermoplastic state, which improves coke strength.17) The surface condition of crushed and sieved briquettes is different from that of “mother” 56 cm$^3$ briquette.

Fig. 1. Photographs of briquettes.
The packing density of coal particles near the surface of crushed and sieved briquettes may be slightly lower than that of 56 cm³ briquette since the latter briquette is considered to break at weaker (in other words, less dense) parts. The effect of the difference in density is considered small when it comes to discussing the swelling of briquette, however, it is worth investigating the difference in density.

Nomura et al. studied the effect of the size of plastic on coke quality. They mixed various plastics having 4 or 5 different sizes with blended coals by 2% and showed that DI_{150} shows a minimum at a certain plastic size and that the minimum size is dependent on the type of plastics, 10 mm for polyethylene (PE) and 3 mm for polystyrene (PS).

As in the case of plastic addition, when the blending ratio of the briquette is constant, the total surface area of the briquette decreases in inverse proportion to the briquette size. This implies that the area impressed by compression decreases with increasing briquette size. It is possible that the latter effect is relatively small and that an increase in the briquette size has a positive improving effect on coke cold strength in this study. The extent to which briquette swells depends on the coal blend composition. It is considered that an improving effect by pitch dispersion is larger for high swelling briquette (C1 in Fig. 3) than that for low swelling briquette (C2 in Fig. 3).

On the other hand, considering that the briquette contains 8–12% of pitch, the smaller the briquette size is, the more uniformly the pitch is dispersed. It is considered that dispersed pitch helps to improve coke quality. Considering the apparent density of briquette (1 180 kg/m³) and the bulk density of the mixture of powder and briquette (850 kg/m³), the charge coal bulk density of powder part is estimated as low as 710 kg/m³. The effect of pitch dispersion on coke quality depends on the composition of powder coal. It is considered that an improving effect by pitch dispersion is larger for high swelling powder coal (D2 in Fig. 3) than for high swelling powder coal (D1 in Fig. 3). This is because the increasing effect of dilatation on coke strength becomes plateau as the dilatation of the coal blend increases.

In test 1, SV of pre-briquette coal is 2.62, which is higher than that of powder coal, 1.88. In this test, DI_{150} shows a maximum at the briquette size of 10 mm owing to the sum of both compression effect (C1 in Fig. 3) and dispersion effect (D2 in Fig. 3). There is little effect of briquette size on DI_{150} resulting in DI_{150} showing a maximum at the briquette size of 10 mm.

In test 2, SV of powder coal is 2.24, which is higher than that of pre-briquette coal, 1.94. In this test, monotonic increase in DI_{150} with increasing the briquette size can be explained by the model of small dispersion effect (D1 in Fig. 3) and large compression effect (C1 in Fig. 3). There is little effect of briquette size on DI_{150} resulting in DI_{150} increasing with increasing the briquette size.

In tests 3 and 4, SV of powder coal and pre-briquette coal is smaller than that in tests 1 and 2. The effect of the briquette size is considered the sum of compression effect (C2 in Fig. 3) and dispersion effect (D2 in Fig. 3). In the region where the briquette size is large, both compression effect and dispersion effect are so small that the effect of briquette size on DI_{150} is small. On the other hand, DI_{6–15} shows a maximum at the briquette size of 10 mm. This is similar to the result that DI_{6–15} shows a maximum at the polyethylene (PE) size of 10 mm. SV of powder coal and pre-briquette coal is small in tests 3 and 4, then the adhesion between briquette and powder coal becomes weak as shown in Fig. 3. As a result of this, cracks with the size of 6–15 mm are likely to generate more in coke and the percentage of 6–15 mm coke fines increases as a result of volume breakage of coke. This explains a maximum of DI_{6–15} and a minimum of DI_{150} at the briquette size of 10 mm. There is a possibility that a certain size of coal briquettes may lead to an increase in volume breakage coke fines.

In this study, the mixture of powder coal and briquettes was carbonized in the test coke oven at the same coal charge bulk density (850 dry-kg/m³) for all the tests. In the case of top charge in a commercial cokemaking process, it is necessary to consider the effect of particle size distribution.
of ‘powder’ because bulk density depends on particle size.

In the study of Montiano et al., the weight of the briquette 23 g and 4–6 g corresponds to volume of 20.0–26.3 cm$^3$ and 4.4–5.7 cm$^3$ (based on apparent density of 1.149–0.873 g/cm$^3$) and to equivalent sphere volume diameter of 33.7–36.9 mm and 20.3–22.2 mm respectively. In the case of briquette blending ratio of 15%, D1 for smaller briquette is larger than that for larger briquette, which corresponds to test 1 of this study. It is considered that the briquette used in the previous study has little caking property because it consists of non-caking coals and biomass and that there is no compression effect (C2 in Fig. 3). Their results can be explained well by considering that only pitch dispersion effect (D2 or D1 in Fig. 3) appears.

### 3.2. Effect of Briquette Size on the Mean Size of Coke, CRI and CSR

The relationship between briquette size and mean size of coke is shown in Fig. 4. The mean size of coke is the average size of +25 mm coke after 30 revolutions in the JIS drum tester. It is known that fissures in coke are one of the important factors determining the size of coke and that the more the amount of fissures is, the smaller the mean size of coke becomes. The mean size of coke decreases with

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**Fig. 3.** Mechanism to explain the effect of briquette size on coke strength.

**Fig. 4.** Relationship between briquette size and mean size of coke.
increasing the briquettes size. It is considered that fissures are likely to be generated in a high density region of larger briquettes; however, clearly further study is necessary on the cause of this and on the fissuring pattern in coke.

The relationship between briquette size and CRI and CSR is shown in Figs. 5 and 6. Except for test 4, CRI changes little with the briquette size. In test 4, CRI of the smallest briquette is the lowest. It is known that an addition of pitch lowers CRI.\(^{25}\) It is considered that pitch dispersion lowered CRI in this case. This effect appears only in test 4 because CRI in test 4 (over 25) is higher than that in the other tests (15–22 of CRI) and CRI is easily to be lowered by pitch dispersion.

It is known that in the case of the addition of biomass (wood, charcoal), the distribution of mineral species, especially calcium, in the coke matrix and surface affects coke reactivity with CO\(_2\).\(^{26-29}\) The addition of fine-sized charcoal increases CRI more than coarse-size charcoal, because the finely dispersed calcium may act as a catalyst agent. However, in cokes produced by the addition of coarse-size charcoal, calcium would be dispersed among discrete areas and would experience less catalytic activity. According to this fact, it is suggested that the smallest size of ground briquettes will behave in the same way.

CSR tends to increase with increasing the briquette size except for test 4. CSR is determined by the combination of \(D_{150}\) and CRI. Although the relationship between briquette size and \(D_{150}\) is classified into three patterns (monotonic increase, maximum and minimum), the relationship between briquette size and CSR are not. The highest CSR for the smallest briquette in test 4 corresponds to the lowest CRI for the same briquette.

In the study of Montiano et al.,\(^{16}\) CRI is higher and CSR is lower for smaller briquette than those for larger briquette. In their study the dispersion effect of non-caking coals and biomass which increases CRI and decreases CSR is considered larger than the dispersion effect of pitch which decreases CRI and increase CSR. The reason why the lowest CRI and the highest CSR for the smallest briquette in test 4 is unclear. The sample coke size of CRI and CSR test is around 20 mm and this size of coke may contain a larger amount of coke texture derived from small briquette, which is considered to have low reactivity due to high pitch content. Further study is clearly necessary such as investigating the coke texture of CRI and CSR test sample in test 4.

3.3. Effect of Briquette Size on Coal Charge Bulk Density

The relationship between briquette size and coal charge bulk density is shown in Fig. 7. Coal charge bulk density increases with increasing the briquette size. It is known that when the void fraction in a two component random mixture
of spheres decreases with increasing the ratio of the size of larger component to that of smaller component.\(^\text{30}\) The average size of powder coal is about 1–2 mm and the mixture of coal briquette and powder coal can be considered as a two component. This explains why the void fraction decreases (the bulk density increases) with increasing the size of coal briquette. A change in briquette size from 0.6–2.0 mm to 50 mm increases the bulk density by 110 kg/m\(^3\), which is a great increase. An increase in bulk density results in improving coke quality and productivity, however, it also has negative effect on coke oven operation such as increasing coking pressure (leading to coke oven wall damage) and worsening coke pushing (heavy push and sticker).\(^\text{17}\)

Increase in briquette size may result in enhancing segregation of coal briquettes,\(^\text{5}\) which may cause uneven coke quality and unstable coke pushing. It is of importance to choose suitable size of briquettes in terms of coke quality, production and coke oven operation.

4. Conclusions

The effect of coal briquette size on coke quality and bulk density in coke oven was investigated by using briquettes made of normal blended coals for blast furnace coke making (hard coking coals and semi-soft coking coals). DI\(^{150}_{15}\) is dependent on the briquette size and its dependency is categorized in three types; firstly DI\(^{150}_{15}\) shows a maximum at the briquette size of 10 mm, secondly it increases with increasing the briquette size and thirdly it shows a minimum at the briquette size of 10 mm. Using briquettes with low caking property may lead to an increase in volume breakage coke fines (DI\(^{150}_{6-15}\)) and a decrease in improving effect of DI\(^{150}_{15}\) by briquette blending. The mean size of coke decreases and coal charge bulk density increases with increasing the briquette size. Briquette size is important from the viewpoint of coke quality, productivity and coke oven operation.

REFERENCES