Preparation of High Strength Coke from Indonesian Lignite by Deeply Pulverized

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Abstract: Coke production from lignite was studied as a continuation of research showing the effectiveness of a sequence of hot briquettes and carbonization on preparations high strength coke from Indonesian lignite. The hot briquetting and carbonization cokes from three Indonesian lignites with a deeply pulverized. Ball milling carried out for 10 hours and 20 hours to obtain a small particle size. Raw lignite hot briquettes at mechanical pressure and temperature of 128 MPa and 200 °C, respectively, allowed cokes to be produced with a tensile strength (TS) of 7-11 MPa. Pretreatment, 10 hour and 20 hour ball milling, respectively increasing TS from the resulting coke to 15-23 and 19-36 MPa. Moreover, it is proved in this study that the structure on the coke fracture has a smooth texture and solid with less pore. There is a strong correlation between TS and coke density of individual lignites.

1. Introduction
Metallurgical coke production has been relying on so-called ‘coking’ or ‘caking’ coals that undergo softening or even melting in bulk upon heating, and then resolidify forming a mechanically strong lump of coke. The total reserve of such coals is only 10% of the total coal reserve, and there is risks in its supply and also price in the future. On the other hand, lignite, a typical type of low rank coal, has hardly been employed as a feedstock of metallurgical coke mainly due to its no caking and highly porous natures. The artificial process is one promising way of producing cokes.

A recent study [1] claimed preparation of coke from a type of lignite, which had tensile strength much higher than those of commercial metallurgical cokes. The key process was briquetting at temperature around 200 °C (hot briquetting; HB) before carbonization. Require making coke from lignite coal should be compacted firstly [2-6]. It is believed that the hot briquetting induces plasticization of the lignite matrix, thereby causing particles’ bonding and densification of briquette as the coke precursor. It is important to understand the relationship between decreased particle size and coke strength [7]. Coal milling is one of the efficient methods to increase the strength of coke.

The primary purpose of this study was to examine the applicability of the HB method to preparation of high strength coke from some Indonesian lignites. The second purpose was to promote
the advantages of the HB by physically modifying a property of the lignites crucial to the strength of resulting coke.

2. Experimental methods

Three different types of lignites (KAL, PKN and PSA) were individually pulverized to sizes smaller than 106 µm and then dried to reduce moisture contents to less than 10 wt%. Table 1 presents the properties ultimate and proximate analyses of the raw coals used in this study. Details of the procedure are reported elsewhere [1 - 2]. In briefly, each pulverized lignite was subjected to HB at 200 °C under mechanical pressure of 128 MPa. The resulting briquette in form of disk (diameter; ca. 14 mm, thickness; ca. 5 mm) was converted to coke by heating to 1000°C at a rate of 5 °C/min in atmospheric flow of high purity nitrogen. Properties of coke such as bulk/true densities, tensile strength (TS), pore size distribution, specific surface area were measured by general methods.

The above-mentioned pulverized lignites were further pulverized by a general ball milling (BM) for 10 or 20 h so that particle sizes were reduced to < ca. 10 µm or < ca. 5 µm, respectively. For approximately 10 grams of coal and zirconia balls (ZrO₂) were added to the 250 ml bottles and were milled for 10 hours-20 hours at the rotation of 70 rpm. The SEM observation showed after deep pulverization reduce the particle size of lignite. Such ‘deeply pulverized’ lignites were converted into cokes in the same manner as above.

3. Results and Discussion

The above-mentioned pulverized lignites were further pulverized by a general ball milling (BM) for 10 or 20 h so that particle sizes were reduced to < ca. 10 µm or < ca. 5 µm, respectively. Such ‘deeply pulverized’ lignites were converted into cokes in the same manner as above. The change in raw size is shown in Fig. 1. The particles size was decreased after ball milling, there is no particle size above 10 µm.

| Sample | Ultimate (wt%)\textsuperscript{c,h} | Proximate (wt %)\textsuperscript{c} |
|--------|----------------------------------|----------------------------------|
|        | PSA | PKN | KAL | Moisture | VM | Ash | FC | HHV (MJ/kg) | LHV (MJ/kg) |
| C      | 59.6 | 63.4 | 62.4 | 6.25     | 46.5 | 6.74 | 40.5 | 28.6         | 27.2         |
| H      | 4.74 | 4.3  | 4.4  | 47.5     | 45.8 | 6.88 | 39.8 | 28.8         | 27.5         |
| N      | 0.94 | 0.91 | 0.71 | 8.62     |      |      |      | 28.1         |      |
| O+S\textsuperscript{d} | 34.7 | 31.4 | 32.5 | 39.7     |      |      |      | 26.8         |      |

\textsuperscript{a}dry basis. \textsuperscript{b}ash free basis. \textsuperscript{c}Air dried basis \textsuperscript{d}by difference
Table 2 shows the TS’s of the cokes prepared by the sequence of the HB and carbonization. The TS’s are in a range of 5.4–11.8 MPa, and equivalent to or clearly higher than that of commercially available cokes, i.e., 5–6 MPa. The HB-carbonization sequence thus converted the lignites successfully to high strength cokes without any additives such as binders. The ‘non-caking’ properties of the lignites were rather favored in the carbonization of briquettes because of no swelling (formation of no large pores) upon heating.

However, the TS’s of the cokes were lower than that reported by Mori et al. [1] who employed a Victorian lignite (Australia) and produced cokes with TS even greater than 30 MPa. It was suspected that such lower TS’s of the present cokes were at least partly attributed to lower thermomechanically-induced plasticity. This property is determined by chemical, exactly saying, macromolecular structure, and it was therefore difficult to modify by an economically acceptable process.

Use of the deeply pulverized lignites caused great increase in TS of the corresponding cokes, as seen in Table 3. Longer BM time, in other words, smaller size of primary particles, greatly contributed to increase in TS. Although not shown in the table, the BM also slightly increased the bulk density of coke. It is known that the porosity of porous solid is a negative factor for its mechanical strength. The mechanism of the BM induced density increase in the briquette/coke density (i.e., decrease in the porosity) was not clear, but it was estimated that smaller-sized particles allowed more significant deformation of the packed bed of the lignite and enhanced densification. It was more importantly found that the BM greatly reduced the pore size in the coke matrix, as presented by Fig. 2 and Fig. 3. It was thus believed that the BM avoided greater pores at which stress concentration occurred, thereby increasing TS.

The significant effect of the coal particle size on TS is evidenced reasonably in Fig. 3, that shows the effectiveness of the BM on the macropore-size reduction and resultant elimination of stress concentration points.

| Feedstock | KAL | PKN | PSA | Ref. |
|-----------|-----|-----|-----|------|
| TS, MPa   | 11.8| 5.4 | 6.8 | 5–6  |

*Table 2. TS of coke prepared by sequence of HB and carbonization
Ref.: typical TS of commercially available metallurgical cokes.*
Table 3. Effect of time for BM before briquetting on TS of coke from HB-carbonization.

| BM time, h | KAL  | PKN  | PSA  |
|------------|------|------|------|
| 0 (< 106 µm) | See Table 2. |
| 10 (< 10 µm) | 23.1 | 7.4  | 14.7 |
| 20 (< 5 µm) | 36.2 | 14.9 | 19.5 |

Parenthesized number: range of primary particle size.

Figure 2. Scanning electron micrographs of fractured and polished surfaces of cokes from (left) pulverize and (right) further 10-h ball-milled KAL.

Figure 3. Effect of the particle size of KAL (before HB) on the size distribution of macro-pores of coke.
Figure 4. Relationship between bulk density and TS of coke: (a) KAL (b) PKN (c) PSA

Tensile strength is a plot one more its bulk density in Fig. 4. All lignite TS of coke were strong related to bulk density. Linear relationships between TS and bulk density shown in Fig. 4 in broad agreement with the coke prepared from Indonesia lignite. The bulk density and porosity with systematic variations obtained change tensile strength [5], there is a frequency of connected nonadhesion boundaries between particle was pivotal for the tensile strength. Its also seen that the 20 h (<5 µm) was given an optimum range of condition for the bulk density.

4. Conclusion
This study has demonstrated applicability of the HB-carbonization sequence to preparation of high strength coke from Indonesian lignites and also effectiveness of the BM on increase in TS to even > 30 MPa. Deeply pulverization treatment has positive effects on increase TS. The maximum TS is of KAL BM20 h coke by three times higher TS than raw 106 µm. The value of coke bulk density is linear to TS, due to a decrease in densities during carbonization release volatiles matter to make solid porous and also depolymerization of coke.

Acknowledgment
Author would like acknowledgment to the member of Hayashi and Kudo Laboratory to acknowledge assistance or encouragement, Azka Mori and Ashik who has taken the time and taught using several equipments and also make discussion with me
Reference

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