



## Full Length Article

# Investigation into the effects of ash-free coal binder and torrefied biomass addition on coke strength and reactivity



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## ABSTRACT

The effects of adding ash-free coal (AFC) as a binder on the physical and chemical properties of coke were investigated to understand the mechanism of coke strength increase and to measure CO conversion reactivity. Two AFC samples (AFC\_P and AFC\_E) were first extracted from Indonesian low-rank coal. Torrefied biomass was also produced from woody biomass for use as an additive to AFC samples. Coke samples were then prepared by adding 3% AFC and 3% AFC + torrefied fuel. The coke strength was investigated by performing indirect tensile tests and <sup>13</sup>C NMR analyses. Coke reactivity was examined using thermogravimetric analysis and a coke reactivity test apparatus. The use of AFC as a binder increased the tensile strength of coke between 39% and 48%. Specifically, coke tensile strength was 2.32 MPa with AFC\_P and 2.16 MPa with AFC\_P + torrefied fuel; and 2.17 MPa with AFC\_E and 2.10 MPa with AFC\_E + torrefied fuel. The reactivity of the coke containing AFC + torrefied fuel was higher than that of the coke containing AFC alone. The coke produced with AFC\_E + torrefied fuel had the highest reaction rate and average CO emission of 257,543 ppm.

## 1. Introduction

Coal is an underground resource that is globally distributed; sub-bituminous coal and brown coal, which are relatively low-grade coals, account for approximately half of all reserves [1,2]. Exhaust gas emissions from direct coal use, such as SO<sub>x</sub> and NO<sub>x</sub>, particulate matter, toxic metal emissions, and greenhouse gas emissions, have long been at the forefront of environmental issues. Furthermore, emissions have worsened owing to the increased use of low-grade coals. Various studies [3,4] have been conducted to address this problem, including a proposal to upgrade low-grade coals via ash removal. One example is a production method for ash-free coal (AFC) [5] that extracts the organic components in coal using a solvent, which leads to thermal pyrolysis as well as changes in the physical structure of the coal. In Japan, AFC is manufactured under the name Hyper-Coal (HPC), and the extracted AFC [6] contains less than 200 ppm of ash, indicating a large increase in the potential heat output compared with raw coal (7800 kcal/kg or more). Owing to this feature, AFC could be used in chemical looping combustion (CLC), catalytic gasification, direct carbon fuel cells (DCFC), and as a blending agent in coke production [2]. AFC exhibits a

swelling behavior during pyrolysis that is found in high-grade coking coal; however, the parent low-grade coal does not show any thermal plasticity characteristic. It is well known that the coke-making process [7] in the coke oven involves softening, swelling, and resolidification, forming a porous and hard coke structure. In this regard, AFC may be used as a substitute and binder [8] to replace a certain amount of coking coal in coke production.

In a blast furnace, coke plays three major roles: as a fuel (heat source), a chemical reducing agent, and an indispensable permeable support. Therefore, the coke strength and coke reactivity are generally regarded as the coke evaluation criteria for modern blast furnaces. However, there is a trade-off relationship between coke strength and coke reactivity [9] as a high coke strength implies a low coke reactivity. To solve this problem, many studies have been conducted on the characteristics of coke to increase both the coke strength and coke reactivity. Sand et al. [10] reported the enhancement of coke mechanical strength of coking coal blends containing high-volatile coals, which have poor caking properties, by densifying the coal prior to carbonization using the stamp-charging method. Nomura et al. [11] has reported the use of semi-soft coking coal using petroleum-derived binder,

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whose interaction with the coal leads to the enhancement of caking property and coke strength. A study by Takanohashi et al. [12] investigated the strength and thermoplasticity of coke by adding a maximum of 35 wt% HPC, based on the higher thermoplasticity of AFC and HPC compared with coking coal, and demonstrated successful improvements in the tensile strength of coke. Based on these findings, the mixing of AFCs extracted from low-grade coal with coking coal during coke production could be a valuable research topic in terms of cost efficiency. Previous studies [13] have reported that average low-grade coal contains 1–2 aromatic rings, and the Indonesian low-grade coal used to produce AFC has a similar number of aromatic rings. A study in Japan [14] evaluated the effect on coke strength of the chemical structure of HPCs produced from various grades of coal, and reported that the effects depended on the number of aromatic rings. Therefore, this study compared the addition of AFC produced in Korea with HPC produced in Japan during coke production, in terms of improvements in coke strength.

Recently, to increase the coke reactivity, biomass was added to industrial coking coal to produce bio-coke. Since biomass is regarded as a carbon neutral fuel, this can be considered a method to reduce carbon dioxide emission. Montiano et al. [15] reported the effect of the addition of waste sawdust on the coke quality and showed that the coke reactivity improved and coke strength decreased with increase in biomass addition. With increased biomass addition, the coke porosity increased but the bulk density decreased. The low bulk density owing to the hygroscopic nature and large volumes of biomass can be improved through densification by the torrefaction process [16]. Torrefaction refers to a thermal pretreatment aimed at upgrading solid biomass fuel by carbonizing biomass under inert gas conditions at 200–300 °C. Matsumura et al. [17] investigated metallurgical coke prepared with carbonized woody biomass. The bulk density of woody biomass increased after the carbonization process, and the hot compaction of biomass at 200–350 °C under 80–100 MPa could inhibit the deterioration of coke strength and increased the addition rate of biomass to the coal blend.

This study analyzes the physical properties of AFC produced from low-grade coal, and investigates coke mixed with AFC and torrefied biomass (hereinafter referred to as torrefied fuel) using the indirect tensile test (ITT) and a chemical analysis based on NMR spectroscopy, to clarify the strength improvement mechanism for coke. It also provides an indicator that can be compared with the strength of coke produced by adding HPC in Japan. The reactivity, an important factor in evaluating coke, was also investigated using thermogravimetric analysis (TGA) and coke reactivity test apparatus (CRTA). The evaluation of the reactivity of coke with added AFC provides little data in the present study. For this reason, we also examined the phenomenal aspects and principles in terms of reactivity. Furthermore, since the torrefied biomass originated from biomass fuel, the REC (Renewable Energy Certificate) weight value [18] can be applied to help reduce carbon dioxide in the steelworks.

## 2. Materials and methods

### 2.1. Material analysis

Approximately 5 g of each sample was analyzed using a thermogravimetric analyzer (TGA 701, LECO Co., St. Joseph, MI, USA) in American standards for testing of materials (ASTM) proximate analysis standard, while an ultimate analysis was obtained using a commercial device (Leco-TruSpec Micro CHNS, LECO Co., St. Joseph, MI, USA). The free-swelling index (FSI) was determined according to the ASTM D 720 method using an automated crucible heating device with the repeat experiments, and an oxide analysis was analyzed by X-ray fluorescence (XRF) spectrometer (S8 TIGER, Bruker, Karlsruhe, Germany) and conducted according to the ASTM D 4326 method.

#### 2.1.1. Raw material analysis

This study used AFC extracted from KCH, a sub-bituminous coal from among the Indonesian low-grade coals, using the solvent extraction method [5,6]. AFC was produced by the solvent extraction process at 0.1 ton/day using experimental apparatus at the Korea Institute of Energy Research (KIER). Non-polar 1-methylnaphthalene (1-MN) was used as the organic solvent for the solvent extraction of coal, at a reactor pressure of approximately 2.5 MPa and at 350 °C. Under these conditions, extraction yields an extract solution in which the organic component of coal is dissolved in an organic solvent. Solid-state AFC was obtained from this extraction solution via two different processes. In the first method, the extraction solution was mixed with hexane at room temperature to precipitate the organic component dissolved in the organic solvent, and the mixed solution was dried at 110 °C to obtain solid-state AFC. The sample obtained via the first method will be referred to as AFC\_P. In second method, the solid-state AFC was obtained by evaporating the organic solvent by heating the extraction solution in a vacuum oven at 250 °C. The sample obtained through this method [19] will be referred to as AFC\_E.

During the torrefaction process, nitrogen is used as the carrier gas in most experimental apparatuses to produce non-oxidizing conditions. The raw biomass fuel is characterized by a high moisture content, low calorie content, and low density owing to its hygroscopic nature and the large volumes involved. The characteristics of these biomass fuels are improved by the torrefaction process [16]. The torrefied fuel in this study was produced using a wood pellet, and the carbonization temperature was in the region of 250 °C. Compared to the raw wood pellet, the torrefied fuel has a reduced moisture content, and the amount of fixed carbon content versus reduced volatile matter was significantly increased.

#### 2.1.2. Semi-automated reflectance measurements

The coal petrographic equipment used in this study consisted of a reflected light microscope (Zeiss, Axioscope-A1), equipped with a spectrophotometer and Coalpro-III software, to measure light reflectance from the samples. The unit was equipped with two light sources: white light from a halogen lamp (100 W), used as the light source for reflectance measurements, and a UV light source from a 110-W halogen lamp, used as the light source for creating fluorescence to determine maceral reflectance (which is specific for certain macerals and minerals, such as liptinite, clay minerals, etc., with known fluorescent characteristics). Vitrinite reflectance represents the maturation of coal; which is related to the coal grade or rank, and also to other properties, such as the volatile matter and fixed carbon contents, the calorific value, etc. Strong correlations can arise between coal rank and coal utilization in a given area (e.g., combustion, gasification, upgrading coal, liquefaction, etc.).

## 2.2. Coal sample preparation

### 2.2.1. Coking coal and additive preparation

Coking Coal A, a hard-coking coal, was used as the standard coal for coke production. The coking coal A sample was ground by using a vibratory disc mill (RS 200, Retsch GmbH, Haan, Germany) and separated by using a sieve shaker (AS 200, Retsch GmbH, Haan, Germany) into three particle distributions: < 0.15 mm, 0.15–0.38 mm, and 0.38–1.00 mm. The torrefied fuel was produced from domestic wood pellets processed for combustion fuel. To prepare the torrefied fuel, samples were placed in a crucible made of quartz, heated at 23 °C/min under a nitrogen atmosphere, maintained at a temperature of 230–250 °C for 30 min, cooled to 100 °C at 22 °C/min, and maintained at this temperature for 5 min. The samples for the experiment were classified into reference coke, coke using AFC as a binder (3% AFC), and coke with torrefied fuel added to 3% AFC (3% AFC + Torrefied Fuel). Table 1 shows the mixing ratios by mass and particle sizes for each sample.

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