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Influence of tarry material deposition on low-strength cokes or pyrolyzed chars of low rank coals on the strength



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ABSTRACT

In this fundamental laboratory scale investigation, carbon/carbon (C/C) composites were prepared from a mixture of pyrolyzed-chars of low-rank coals (lignite and sub-bituminous coals) or low-strength cokes (drum index: $D_{16}^{150} = 52$ and 73) and coke-oven gas (COG) tar to investigate the possibility of producing high-strength coke for blast furnace use from low-grade coals or cokes. In addition, the optimum conditions for composite preparation and the influence of the tarry material on the strength of the prepared C/C composite was examined. The C/C composites were prepared as follows: the tar was first added over particles of coke or char at room temperature, subsequently, the mixture was pyrolyzed at a predetermined temperature (500-900 °C) in an inert atmosphere. The indirect tensile strength of the C/C composite prepared at a pyrolysis temperature of 500 °C increased with increasing weight ratio of tar to cokes (3.5-4.5 MPa) or chars (2.0-3.0 MPa) up to a ratio of 2.0; at this point, the indirect tensile strength reached approximately 6.0 MPa. The indirect tensile strength of the C/C composites prepared with the weight ratio of tar to cokes/chars of 2.0 also increased with increasing pyrolysis temperature, reaching approximately 7.0-7.5 MPa at 800-900 °C, which was comparable to the ca. 7.0 MPa strength of the high-grade cokes ($DI_6^{150} = 87$) used in commercial blast furnaces. Based on pore size distribution measurements using the mercury intrusion porosimetry, N₂ adsorption, and polarized optical microscope methods, it was likely that the increase in the indirect tensile strength of the prepared C/C composite occurred because of the vapor infiltration of tar-derived carbonaceous materials into the micro-, meso- and macro-pores in the low-grade cokes and pyrolyzed chars during the pyrolysis of the mixtures.

1. Introduction

Coke for blast furnaces is conventionally manufactured by the carbonization of blended coal (caking coal and non- or slightly-caking coal), but such caking coal has a confirmed reserve amount of ca. 10% that of coal as a whole. Therefore, future increases in the price and decreases in the prevalence of caking coal are predicted. For these reasons, the development of a new cokemaking method that can reduce the amount of caking coal required the growth of the use of low-grade coals (lignite, sub-bituminous coal, non- or slightly-coking coal, etc.) is necessary. In addition, it is also very important that the CO_2 emissions from the iron-making process using metallurgical coke is significantly reduced. Therefore, high-strength coke is required, assuming blast furnace permeability, for stable operation and the reduction in the

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Table 1

Analyses of samples used in this study.

Sample	Code	Elemental analysis wt%-daf					Proximate analysis, wt%-dry			${\rm DI_{6}^{150d}}$
		С	Н	Ν	S	O ^a	Ash	$\rm VM^b$	FC ^{a,c}	
Lignite	ML	69.6	9.3	0.9	0.1	20.1	2.9	49.8	47.3	-
Lignite char	ML char	97.0	0.7	0.7	0.1	1.5	4.8	-	-	-
Sub-bituminous coal	AD	75.8	7.9	1.0	0.1	15.2	0.3	50.1	49.6	-
Sub-bituminous char	AD char	96.0	0.9	1.3	0.1	1.7	1.0	-	-	-
Extremely low grade coke	DI50 coke	97.6	0.2	1.2	0.3	0.7	6.8	0.7	93.3	51.6
Low grade coke	DI70 coke	95.1	0.4	1.2	0.4	2.9	12.7	1.4	85.9	72.8
High grade coke	DI90 coke	97.3	0.2	1.1	0.4	1.0	11.7	0.6	87.7	87.1
COG tar	tar	94.3	3.9	1.0	0.5	0.3	< 0.1	67.0	33.0	-

^a Estimated by difference.

^b Volatile matter.

^c Fixed carbon.

^d Drum Index.

reducing agent ratio [1,2]. The drum index (DI), coke-strength after reaction (CSR), and coke-reaction index (CRI) are the main indicators of coke-quality for blast furnaces [3–5]. For coke-reactivity, to date, low-reactivity coke has been in demand because the CSR increases with decreasing CRI, as shown by the Nippon Steel Corporation (NSC) method [4,6]. However, recent studies have showed that the use of high-reactivity coke can reduce the thermal-reserve-zone temperature of the blast furnace, and it is thus possible to improve the efficiency of ore reduction and optimize the reducing-agent ratio, resulting in a reduction in CO_2 emissions [7,8] Therefore, it is necessary to develop a high-strength and high-reactive coke to drastically reduce CO_2 emissions in the iron-making process; to date, several works concerning this have been reported [8–14].

In general, it is well known that the reactivity and strengths of coke or carbon materials are influenced by the pore structure, carbon structure, and catalytic activity of the minerals, and it is important to clarify the effects of these factors on the reactivity and strength of the carbon material. So far, it has been reported that the coke strength and gasification reactivity can be increased and decreased by coating with pyrolytic-C and polyvinyl-alcohol [15-21]. Although high-strength coke production from lignite or sub-bituminous coal is important for achieving the maximum usage of low-quality coal, it has not been investigated by pyrolytic-C or coating methods. The inherently-present metals in low-rank coals are well known to have high catalytic gasification effects. Therefore, if coke could be produced from the above coals, the coke would be both high-strength and highly-reactive. In addition, coke produced from non- or slightly-caking coal is generally low strength. If it is possible to increase the strength of low-grade coke, it would be possible to create high-strength and low-reactivity coke, which could be used in processes requiring reducing-agents, such as coke oven gases (COGs) or H₂, in a blast furnace. Our research group has so far reported the preparation of iron oxides/carbon composites from limonite, cold-bonded pellets, and pre-reduced sintered-ores; in these materials, the mesopores were completely filled by carbonaceous materials derived from COG tar by impregnation or vapor deposition methods. We have found that the crushing strengths of the prepared composites are greater than those of the original samples [22-26]. Based on our findings, we hypothesized that high-strength coke could be produced if carbonaceous material derived from coal tar could be used to fill the pores of chars prepared from low-rank coals or lowgrade coke.

From the viewpoint of reducing CO_2 emissions, the development of effective renewable energy resources such as woody biomass is being conducted. On the other hand, coal-tar is mainly used as a raw material for chemical products and pitch. If the development of these products from inexpensive biomass-tar is possible, coal-tar will become surplus to requirement. Thus, it is possible that the production of high-strength coke from low-grade coal or coke and tar utilization will become

economically viable. Therefore, it may be important to investigate highstrength coke production methods for integrated steelworks using coal tar from coke ovens and low-grade coal or coke. In addition, the findings using coal-tar may allow the use of other carbonaceous materials having thermoplasticity (for example, vacuum-residual-oil, hyper-coal, or new thermoplastic materials). When using coal-tar, the handling of harmful, carcinogenic substances is a problem. However, in general, methods of handling tar have already been established in integrated steelworks or chemical products manufacturing industry. Therefore, the installation of the tar treatment, which is already utilized in integrated steelworks, using the method proposed in this work will solve the above-problems in the production process of high-strength coke from coal-tar and low-grade coal or coke.

In this fundamental research, therefore, we first prepared carbon/ carbon composites in which COG tar-derived carbonaceous materials were used to fill the pores in pyrolyzed chars of lignite, sub-bituminous coal and low strength coke. The indirect tensile strengths of the composites were measured to demonstrate the production of high strength coke from low rank carbonaceous materials. In addition, the influences of the carbon structure and pore structure on the indirect tensile strength of the prepared composites were investigated in detail.

2. Experimental

2.1. Samples

Indonesian lignite (ML), sub-bituminous coal (AD), three cokes having different drum indexes (DI₆¹⁵⁰), and tar derived from a commercial cokemaking process were used in the present study. In the preparation of pyrolyzed-char of low-rank coals, ML or AD particles with a size fraction of 4.7-6.0 mm were pyrolyzed using a flow-type fixed-bed quartz-made reactor in helium at 10 °C/min up to 900 °C. The chars prepared from ML and AD are denoted ML char and AD char, respectively, and these comprised particles of 4.0-4.7 mm in size. Table 1 also lists the analysis of the samples used. The three-kinds of cokes and COG tar were received from an iron-making company in Japan. The two cokes used were produced from the carbonization of blended coal (caking coal and non- or slightly caking coal) in a several kilogram-scale furnace at the above-mentioned company. In the present study, the as-received cokes were crushed and sieved to size-fraction of particles was 4.0-4.7 mm. The DI₆¹⁵⁰ values of the as-received cokes ranged from 52 to 87. The cokes used in this study were denoted as DI50 coke, DI70 coke, and DI90 coke based on the DI_6^{150} values. The dry and ash-free (daf) carbon contents of the ML or AD pyrolyzed-chars, cokes, and tar were 96-97, 95-98 and 94 wt%, respectively.

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