Development of Carbon Iron Composite Process [†]

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Abstract:

In recent years, the development of innovative energy saving technologies for preventing global warming has become necessary. Means of realizing innovative energy saving in the steel manufacturing process and radical low reduction agent ratio (RAR) operation in the blast furnace include improvement of the heat balance and active control of the reduction equilibrium (temperature of thermal reserve zone). As a means of realizing low RAR operation, this paper describes the development of an innovative raw material "Carbon Iron Composite" for satisfying both high coke reactivity and iron ore reduction, focusing on the temperature of the thermal reserve zone. Carbon Iron Composite is produced by mixing coal and iron ore (pellet feed) and densification by hot briquetting, followed by carbonization of the briquetted materials. The results of a fundamental investigation of the production process for Carbon Iron Composite as a new blast furnace burden material obtained by carbonization of the densified briquettes clarified the following: (1) It was possible to form briquettes as raw material for Carbon Iron Composite by hot briquetting without using a binder. (2) The possibility of producing Carbon Iron Composite with a compression strength of more than 2 000 N using a mixture of 40% iron ore and 60% coal was confirmed. The reduction ratio of the product exceeded 70%. (3) Carbon Iron Composite has a coke reactivity index (CRI) of more than 50% and displayed a typical surface reaction. Melting and separation phenomena were not observed after heat treatment at 1 700°C.

1. Introduction

The Japanese steel industry accounts for 14% of domestic CO₂ emissions. Because the ironmaking process (sintering, cokemaking, blast furnace) generates an overwhelmingly high percentage of these emissions, at 70%, the industry has historically made positive efforts to develop environment-friendly energy saving technologies. However, against the background of the recent rapid rise in resource prices, it is imperative that the steel industry develop an ironmaking process which can respond flexibly to resource conditions, making it possible to absorb fluctuations in raw material and fuel prices. Moreover, around 2010, the existing ironmaking plants in Japan will reach the end of their useful life, and replacement or modernization of large-scale ironmaking equipment such as coke ovens will be required. To ensure the superior international competitiveness of the Japanese steel industry over the long term, it will be necessary to accelerate the response described above. In particular, the creation of an innovative ironmaking technology which satisfies all the requirements of energy saving, environmental protection (prevention of global warming), high productivity, flexible use of raw materials (reform of low grade resources), and labor saving is strongly desired.

Where basic and generic technologies are concerned, the European steel industry has launched the ULCOS (Ultra Low CO_2 Steelmaking) Project with the aim of reducing CO_2 , and vigorous research and development are now in progress at the related companies. In Japan, joint projects involving steel makers which investigated original Japanese energy saving/ CO_2 reduction processes include the national project "Energy Half Consumption

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Senior Researcher Deputy Manager, Ironmaking Res. Dept., Steel Res. Lab., JFE Steel Project" (1999–2003) sponsored by MEXT (Ministry of Education, Culture, Sports, Science and Technology) and the "Blast Furnace Limit Phenomena Research Committee" (FY2003–2005) in the Iron and Steel Institute of Japan. Several seminal technologies were developed in these projects, but at present, none has reached the stage of process implementation.

In the blast furnace process, technologies for improving reaction efficiency in the blast furnace make it possible to reduce the reduction agent ratio (RAR), and thus are extremely important in that a CO_2 emission reduction effect can be expected. Among these, the method of reducing the temperature in the thermal reserve zone in the blast furnace by using high reactivity coke¹) is considered particularly promising.

Therefore, the aims of the present research are to realize energy saving (for reduction of CO₂ emissions) by low RAR operation of the blast furnace and strengthen resource flexibility by high rate use of inferior raw materials such as steam coal and low-grade iron ore, utilizing the seminal technologies mentioned above. This paper describes the results of a basic study on a Carbon Iron Composite production process, in which coke briquettes containing a partially-reduced powdery iron source are produced by mixing and hot briquetting coal and fine iron ore raw materials to form coal/iron ore composite briquettes, which are then carbonized to produce a composite of coke and partially-reduced iron. The new process does not depend on the existing cokemaking or sintering processes, and thus also offers a potential solution to the above-mentioned problem of aging production equipment.

2. Experimental Method

2.1 Hot Briquetting Tests

Hot briquetting experiments were performed using a hot briquetting machine equipped with a moving bedtype coal heating device and a hot briquetting unit, as shown in **Fig. 1**. As main specifications of the briquetting machine, maximum pressure was 40 t, the maximum rotation speed of the rolls was 10 rpm, the maximum rotation speed of the screw feeder used to charge the raw materials was 140 rpm, and the roll diameter and roll width were 400 mm and 120 mm, respectively. Two cup shapes were used, a pillow type and a smaller ovoid type (resembling the common "bean charcoal" briquettes used in Japan). The cup volume of the pillow type was 50 cm³ ($60 \times 46 \times 14$ mm), and that of the ovoid type was 18 cm³ ($39 \times 39 \times 19$ mm).

In evaluation of hot briquetting, briquetting yield and the strength of the briquette product were considered. The conceivable control variables for these evaluation items include the briquetting pressure, briquetting tem-



Fig. 1 Schematic illustration of hot briquetting machine

perature, roll cup volume, roll speed, and the rotation of the screw feeder used to charge the raw materials. In this paper, the effect of the roll cup volume on briquetting yield and briquette strength was investigated.

The briquetting conditions are shown in **Table 1**. The briquetting pressure (linear load) was held constant at 3.5 t/cm. The briquetting temperature was assumed to be the temperature of the briquetting sample material in the hopper above the screw feeder, and was in the range of $110-130^{\circ}$ C.

The method used in the briquetting experiments is described below. Coal and iron ore adjusted to the specified size and mixture were introduced into the raw material hopper of the hot briquetting machine shown in Fig. 1, and were heated to the specified temperature while passing through a moving bed-type coal heating device equipped with a single fin screw. The heated raw material mix was then charged to the screw feeder hopper installed above the briquetting rolls, and hot briquetting was performed at the specified roll speed and screw feeder speed. Approximately 20 kg of a mixed sample of coal and iron ore were used in each briquetting experiment. Briquettes were sampled from a sampling box under the rolls.

The industrial analysis value, mean maximum reflectance, and fluidity of the coal used are shown in **Table 2**.

Table 1 Briquetting condition

Briquetting pressure	3.5 t/cm
Rotation number of roll	3 rpm
Rotation number of S/F	35 rpm
Briquetting temperature	110–130°C
Cup volume	18, 50 cm ³
Brending ratio Coal/ore	60%/40%
Total mass of raw materials	20 kg

Iron ore: Pellet feed Coal: Slightly caking coal rpm: Revolutions per minute

Mean maximum reflectance was measured in accordance with JIS M 8816. As fluidity, maximum fluidity was measured with a Gieseler plastometer in accordance with JIS M 8801. The particle size of the coal was adjusted to 100% -3 mm by pulverizing. The iron ore used was pellet feed, which is a type of fine iron ore. The particle size measured with a laser diffraction/scattering particle size distribution analyzer showed that $-74 \,\mu m$ comprised 81mass%. Table 3 shows the results of a chemical analysis of the fine iron ore.

2.2 Carbonization Tests

Carbon Iron Composite was produced by carbonizing the mixed samples of coal and fine iron ore using an electric heating-type coal carbonizing furnace²⁾. The properties of the mixed samples are shown in Table 4. In the experiments, the carbonizing drum (360 \times 270 \times 400 mm) was packed with a bed of coke breeze so as to avoid contact between the briquettes. The samples were then introduced into the carbonizing furnace, and carbonization was performed for 6 h under conditions in which the temperature in the coal reached 850°C at a furnace wall temperature of 1 100°C. After carbonization, the Carbon Iron Composite was dry quenched in a nitrogen gas flow.

2.3 Evaluation of Briquettes and **Carbon Iron Composite**

The briquetting yield and compression strength of the sampled briquettes were measured. Briquetting yield was measured as briquetting yield I, which shows the mass% of +1 mm in the total briquette product sampled, and briquetting yield II, which shows the mass% of

VM	Ash	Ro	MF		
(Dry basis, S	%)	(%)	(logddpm)		
35.3	8.8	0.70	1.5		
VM: Volatile matter	Ro: Refle	ectance of coal			

Table 2 Coal properties

MF: Maximum fluidity ddmp: Dial division per minute

Table 3 Chemical composition of the iron ore

							(mass%)
T-Fe	FeO	SiO	Al ₂ O ₃	CaO	MgO	Mn	TiO ₂
67.50	0.21	1.31	0.73	0.01	0.01	0.11	0.07

Table 4	Raw	material	properties
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_							(m	ass%)
Mixing ratio			Composition					
	Iron ore	Coal	T-Fe	Fe_2O_3	FeO	Ash	С	Н
	40.0	60.0	27.1	38.5	0.2	5.2	45.6	2.9
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Iron ore: Pellet feed Coal: Slightly caking coal

67.

the original form in the total briquettes not containing cracks. Measurement of compression strength was performed using an autograph. In evaluation of the Carbon Iron Composite, compression strength was measured in the same manner as with the briquettes, and a high temperature reaction test and high temperature heat treatment test were performed as high temperature characteristics tests. The high temperature reaction test was performed at a reaction temperature of 1 100°C using a CO₂ reaction device³⁾ by passing 100% CO₂ gas and measuring the reaction time when the reaction degree reached 20%. The high temperature heat treatment test was performed by reacting samples for 1 h in a 100% argon atmosphere at a heat treatment temperature of 1 700°C using a high temperature hot drum test device⁴).

3. Experimental Results and Discussion

3.1 Yield and Compression Strength in Hot Briquetting Test

Photo 1 shows a photograph of the external appearance of the briquettes and an optical microscope photograph of the cross section of a briquette. The briquetting temperature of 110-130°C is far lower than the temperature at which coal softens and begins to melt, and the original shape has been retained in the briquettes. Although different from the microstructures of briquettes produced at temperatures near the softening temperature/melting point of coal⁵), the coal has undergone densification in a condition in which the fine iron ore has infiltrated into the crevices between the coal particles. As a result, it was found that briquettes can be produced by densification without using a binder.

Figures 2 and 3 show the relationship between the cup volume when briquetting a mixture of 60 mass% coal and 40 mass% fine iron ore, and the resulting briquetting yield and compression strength, respectively. As the cup volume decreases, both briquetting yield and compression strength increase. The assumed explanation for this is as follows: Because the amount of the raw material mixture fed to the briquetting rolls is the same, transmission of briquetting pressure to the entire



 $100 \,\mu m$

Photo 1 Appearance and sectional photograph of hot briquette

Photo 2



Fig. 2 Effect of briquetting cup volume on briquetting yield



Fig. 3 Effect of briquetting cup volume on compression strength of briquette

briquette improves as the cup size of the briquetting rolls decreases and the thickness in the depth direction becomes thinner. As a result, briquetting is improved and the apparent density of the briquettes increases. From this result, it was understood that apparent density is one of important factors for increasing the strength of the briquettes.

3.2 Properties of Carbon Iron Composite

3.2.1 External appearance and compression strength

Photo 2 shows photographs of briquettes before and after carbonization, cross-sectional photographs of the same briquettes, and a polarization microscope image of the microstructure of the Carbon Iron Composite. The carbonization yield and reduction rate were 69% and 76%, respectively. No cracking accompanying heat treatment was observed. This is estimated to be because initiation of cracking was suppressed by the use of coal with a low fluidity and coefficient of thermal expansion. As the coke in contact with the iron ore in the Carbon Iron Composite was consumed and reduced, a condition in which the surrounding coke microstructure became porous was observed. However, the compression strength of the Carbon Iron Composite was more than



After carbonization

bonization

Appearance of hot briquette and Polarization microscopic photograph of hot briquette after car-



Fig.4 Properties of carbon iron composite

4 000 N, showing a higher value than that of coke oven coke (Fig. 4). Although low fluidity coal was used, it is estimated that the adhesion strength of the coal particles was further strengthened by the synergistic effect of the appearance of thermoplastization and the reduction of the distance between particles by densification.

3.2.2 High temperature reaction properties of carbon iron composite

Photo 3 shows sectional views of Carbon Iron Composite before reaction and after reaction for 1 h in the CO₂ reaction device, together with polarization microscope images of the same briquettes. At a reaction temperature of 1 100°C, the reaction mode is on the boundary between a condition in which the reaction is rate-governing and one in which intragranular diffusion is rate-governing. Therefore, in addition to the porous structure of the coke, it is considered that reactivity at 1 100°C also depends on the reactivity of the coke microstructure. In Carbon Iron Composite, the reaction proceeds from the surface, displaying a typical characteristic of high reactivity coke. Although the reaction rate including the iron fraction was 26.6%, a trial



Photo 3 Comparison of sectional view between the reacted and the non-reacted carbon iron composite



Fig. 5 Comparison of compression strength between the reacted and the non-reacted carbon iron composite



Photo 4 Sectional photograph and microscopic photograph

calculation of coke reactivity index (CRI)³⁾ based on a coke standard gave a result of 53%, which is more than twice as large as that of conventional coke oven coke, while also maintaining a compression strength of more than 2 000 N (**Fig. 5**). Thus, suppression of powdering in the blast furnace can be expected. **Photo 4** shows a photograph of the external appearance of Carbon Iron Composite after heat treatment at 1 700°C, together with a metallographical microscope image. Heat cracking behavior due to thermal expansion of iron was not detected. Cohesion of molten metallic iron with adjoining metallic iron and growth as a large structure were observed.



3.3 Image of Carbon Iron Composite Production Process

An image of the Carbon Iron Composite production process is shown in **Fig. 6**. Iron ore and coal crushed to the specified particle sizes are mixed in the specified proportions, and the resulting raw material mix is densified by hot briquetting without using binder. The densified briquettes are then charged into a direct heatingtype shaft carbonization furnace equipped with high temperature and low temperature tuyeres, and Carbon Iron Composite containing metallic iron is produced. As a high reactivity coke, Carbon Iron Composite is expected to be used as a partial substitute for coke oven coke and will enable low RAR operation of the blast furnace.

4. Conclusions

A fundamental study was carried out on a production process for a new blast furnace raw material, Carbon Iron Composite, which is obtained by densification of a mixture of iron ore and coal by hot briquetting, followed by carbonization of the densified briquettes. As a result, the following points were clarified.

- (1) It was possible to produce briquettes of the raw material for Carbon Iron Composite by hot briquet-ting without using a binder.
- (2) The possibility of producing Carbon Iron Composite having a compression strength exceeding 2 000 N using a mixture of 40% iron ore and 60% coal was confirmed. The reduction rate of the product exceeded 70%.
- (3) Carbon Iron Composite has a CRI of more than 50% and displays a typical surface reaction. No melting and separation phenomena were observed after heat treatment at 1 700°C.

The majority of the results obtained in this research are basic research results. However, based on a detailed

future study for optimization of the selection of raw materials, blending conditions, briquetting conditions, carbonization process, etc., the authors are confident that this process can make an important contribution to renovation of aging coke ovens and low reduction agent rate operation of blast furnaces.

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