

Recent Progress in Ironmaking Technology for CO₂ Mitigation at JFE Steel[†]

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Abstract:

In order to contribute of the prevention of global warming, the reduction of CO₂ from the steel works becomes a major issue imposed on the steel industry. A variety of new CO₂ reduction technologies have been developed in the past decade in the ironmaking area of JFE Steel. This paper focuses on some of the technologies developed in JFE Steel, such as natural gas injection technology into the sintering bed and burden distribution control of blast furnace with coke mixed charging. The outline of the technology and the effects of application are described.

1. Introduction

Carbon dioxide exhausted by the iron and steel industry accounts for approximately 15% of the total generated in Japan¹⁾, and emissions from the ironmaking process occupy about 70% of that amount. Accordingly, the ironmaking process should play an important role in CO₂ mitigation.

The ironmaking process centers on the blast furnace, which produces molten pig iron, and also includes the sintering machine, which produces sintered ore as a raw material for the blast furnace, coke ovens that produce coke, which is important as a reducing agent, and hot stoves that generate the high temperature (approx. 1 200°C) hot blast blown into the blast furnace. Large amounts of CO₂ are generated by the reduction reaction ($\text{Fe}_2\text{O}_3 + 3\text{CO} \rightarrow 2\text{Fe} + 3\text{CO}_2$) in the blast furnace and the combustion reaction ($\text{C} + \text{O}_2 \rightarrow \text{CO}_2$, $\text{C}_n\text{H}_m + (n+m/4)\text{O}_2 \rightarrow n\text{CO}_2 + (m/2)\text{H}_2\text{O}$) of carbonaceous materials (coke breeze, etc.) and carbon-containing gases (blast furnace

gas (B gas) and coke oven gas (C gas)) in the sintering machine, coke ovens, and hot stoves.

First, this paper explains the current condition of CO₂ generation in the individual processes of the ironmaking process based on the results of calculations for a model steel works^{2, 3)}. The basic concepts for reducing emissions based on those calculations are also described. Next, the following are taken up as technologies which were developed in JFE Steel based on these concepts and have already been applied practically, and the outline of the technologies and their effects when applied are described. The practical technologies discussed in this paper are a technology for injection of hydrocarbon gas into the sintering bed at the sintering machine⁴⁾, a burden distribution control technology for high coke-ratio mixed coke charging in ore with the aim of achieving a large reduction in the reducing agent rate as a CO₂ mitigation technology for the blast furnace^{5, 6)}, and a city gas injection technology by injecting city gas directly into the blast furnace as a low C, natural gas-based gas⁷⁾. In addition, JFE Steel is also engaged in development of a new blast furnace raw material called Carbon Iron Composite (hereinafter, CIC)⁸⁾ based on a new concept with the aim of realizing a drastic reduction in the reducing agent rate in blast furnace in the future. The technical content and expected results of that technology are also described.

2. Current Condition of CO₂ Emissions in Ironmaking Process³⁾

For a quantitative evaluation of the amounts of CO₂ generated in the individual processes of the ironmaking

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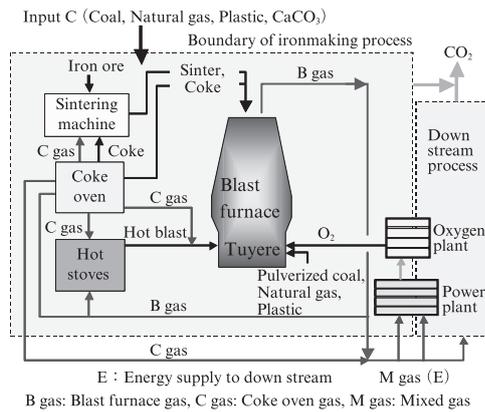


Fig. 1 Schematic diagram of material flow in ironmaking process and boundary of ironmaking

process, the boundary conditions (boundary) differentiating the ironmaking process and the downstream processes and the material flow in the ironmaking process were set as shown in **Fig. 1**. Externally-supplied coal comprises coking coal which is charged to the coke ovens, where it is carbonized, and steam coal and natural gas which are injected directly into the blast furnace. Coke produced by the coke ovens is screened, after that coke breeze is used as a bonding agent for sinter, and lump coke is charged to the blast furnace. The coke oven gas (C gas) and blast furnace gas (B gas) generated by the coke ovens and blast furnace are used as fuels in various ironmaking processes (sintering, coke ovens, hot stove, power plant), or are injected into the blast furnace and consumed as reducing agents, after which the surplus gas is supplied to downstream processes (steelmaking, rolling processes, etc.) as mixed gas (M gas). The mixed gas which is supplied to the power plant is treated by separating the amount supplied to the ironmaking process and that supplied to downstream processes. The amount supplied to the ironmaking process is assumed to be only the amount which is necessary and sufficient for generating the electric power and oxygen required in the ironmaking process.

Figure 2 shows the results of a calculation of the C

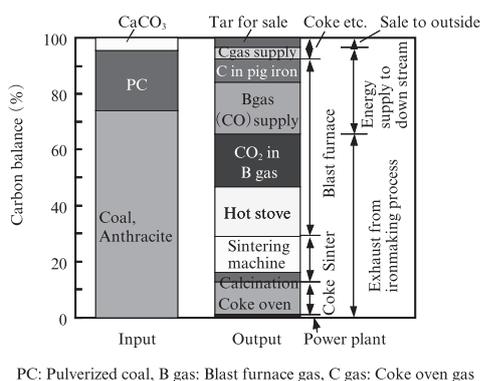


Fig. 2 Carbon (C) balance in the model steel works

balance in a model steel works using a heat and mass balance model²⁾ which considers the above-mentioned boundary conditions and material flow. The basic conditions of the model works are reducing agent ratio (RAR) = 495 kg/t, pulverized coal ratio (PCR) = 150 kg/t, and coke ratio (CR) = 345 kg/t. Almost carbon input to the steel works (hereinafter, Input C) is coal-derived, the remainder being carbon in the limestone (CaCO₃) that is introduced in the calcinating process to obtain burnt lime (CaO), which is necessary in sintering. On the other hand, carbon discharged from the ironmaking process (hereinafter, Output C) includes approximately 65% from blast furnace-related equipment (C originating from CO₂ generated by reduction of iron ore and fuel combustion in hot stoves), and approximately 15% each from the coke ovens and sintering machine (fuel combustion for carbonization of coke, combustion of bonding agents in the sintering machine, etc.). The remainder is the balance, which is sold outside and is subtracted from CO₂ generation.

The aforementioned surplus M gas which is supplied to downstream processes is effectively utilized as a fuel for heating furnaces, or for generation of electric power. The C in the molten pig iron is also utilized as a fuel in the same manner after converted to converter gas (LD gas). Accordingly, because the CO in B gas, hydrocarbons in C gas, and C in molten pig iron are all finally converted to CO₂, the total C contained in these gases is equivalent to the CO₂ emission from the downstream process. As shown in Fig. 2, quantitatively, this is approximately 30% of the total. The remainder is exhausted directly from the ironmaking process and is a balance of approximately 70%.

3. Approach to CO₂ Mitigation in Ironmaking Process

From Fig. 2, the fundamental approach to CO₂ mitigation is reduction of the necessary Input C in the ironmaking process by the cumulative effects of the following and similar countermeasures:

- (1) Reduction of the necessary hot blast by reduction of RAR in the blast furnace, and reduction of hot stove energy consumption.
- (2) Reduction of the coke oven working ratio by reduction of the blast furnace coke rate, and accompanying this, reduction of total carbonization heat.
- (3) Reduction of CO₂ in the combustion off-gas in sintering by reducing the bonding agent ratio.

Chapters 4–7 present outlines of each of the above-mentioned processes and the contribution to CO₂ mitigation in that order.

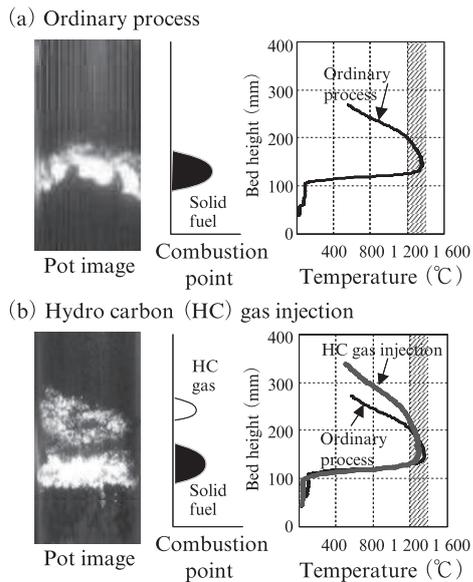


Fig. 3 Schematic diagram of heat pattern and ignition behavior with hydrocarbon gas injection technology

4. Injection of Hydrocarbon Gas into Sintering Bed⁴⁾

4.1 Background of Research

Conventionally, in order to improve sinter strength in the sintering process, bonding agents such as coke breeze, anthracite, etc. are usually increased. However, if increased blending of bonding agent is excessive, the maximum temperature in the sinter bed arises and the calcium ferrite decomposes into hematite and slag. As a result, sinter strength and product yield will be decreased.

4.2 Outline of Developed Technology

In the sintering technology using hydrocarbon gas, hydrocarbon gas is added from the sintering bed surface for heat compensation in the upper layer of the bed, where the sinter strength is low due to heat shortage, at the point when sintering has progressed to this part of the bed. As a result, it is considered possible to produce sinter with higher strength and reducibility by retaining calcium ferrite, which has high strength and reducibility, while promoting liquid phase sintering by maintaining the high temperature region in the sintering bed at 1 200–1 400°C for a longer time.

Figure 3 shows a schematic diagram of the heat pattern in the sintering technology using hydrocarbon gas injection. According to the experimental results, the hydrocarbon gas added from the surface of the sintering bed initiates combustion at around 700°C before reaching the position of bonding agent combustion, that is, above the position of coke combustion. Therefore, the heat pattern changes so that the high temperature part expands upward, and it is possible to prolong the hold-

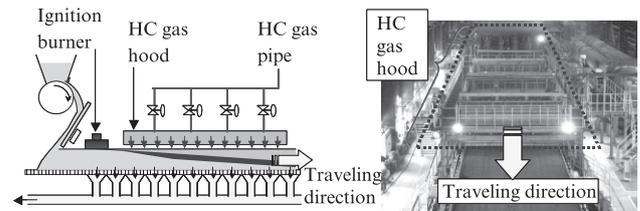


Fig. 4 Hydrocarbon (HC) gas injection equipment at No. 1 sinter plant in East Japan Works (Keihin)

ing time at 1 200–1 400°C.

4.3 Results of Application in Actual Sintering Machine

Figure 4 shows an outline of the sintering equipment using hydrocarbon gas injection. In the actual machine, city gas was used as the hydrocarbon gas considering the facts that (1) is not toxic, (2) has a high lower explosion limit concentration, (3) can be supplied to the plant at high pressure, and other advantages. The typical composition is CH₄: 89%, C₂H₆: 5%, and C₃H₈: 6%. In order to provide heat compensation to the upper layer of the sintering bed, multiple city gas addition hoods were installed above the sintering machine adjacent to the ignition burner over a range of 1/3 of the length of the sintering machine, and the specified amount of city gas is added/mixed with the air sucked into the machine. The results of an actual operational test confirmed that shatter strength improves by approximately 1%, the reducibility index, RI, improves by approximately 3%, and unit consumption of the bonding agent decreases by 3 kg/t. From the viewpoint of enthalpy balance, the bonding agent could be reduced by an amount equivalent to about 4 times the added hydrocarbon gas. Since use of this sinter improved reducibility, a reducing agent ratio reduction effect of approximately 3 kg/t could be obtained at the blast furnace.

5. Development of Technology for High Mixed Coke Charging into Blast Furnace Ore Layer⁵⁾

5.1 Background of Research

Low reducing agent ratio (RAR) operation at the blast furnace is important for CO₂ mitigation in the ironmaking process. However, various problems arise in low RAR operation, including delayed reduction of the burden (iron ore), decrease in permeability due to enlargement of the cohesive zone, and deterioration of the heat balance. Improvement measures include improvement of burden material properties, improvement of reducibility by burden distribution control, improvement of the heat balance by control of the peripheral gas flow. However, JFE Steel focused on high mixed coke charging to the

ore layer. As mixed coke charging to the ore layer not only suppresses ore shrinkage in the cohesive zone, but also serves to protect the coke layer because the mixed coke is preferentially subject to the solution loss reaction, then disintegration of coke is suppressed and the void fraction of the cohesive zone is expected to be maintained. As a result, gas permeability is improved, with the potential for reduction of RAR, increased productivity, and increased use of low quality burden materials.

5.2 Outline of Developed Technology

The effects of the mixed coke ratio and sinter ratio on the reduction degree of sinter at 1 200°C and maximum pressure drop were investigated by a softening and reduction experiment under load. The results are shown in Fig. 5. At all sinter ratios, maximum pressure drop tends to decrease as the mixed coke ratio increases. However, while this effect is remarkable up to a mixing ratio of 12.5 mass%, it tends to become saturated at higher mixing ratios. Regarding the reduction degree, in spite of the large variations in the data, the reduction degree increases with the mixing ratio up to a mixed coke ratio of 10 mass%. From the above, it can be understood that promotion of the reduction degree of sinter and a large improvement in pressure drop can be obtained up to a mixed coke ratio of approximately 10 mass%.

5.3 Results of Application to Commercial Blast Furnace

For application to a commercial blast furnace, the burden distribution pattern for obtaining a uniform

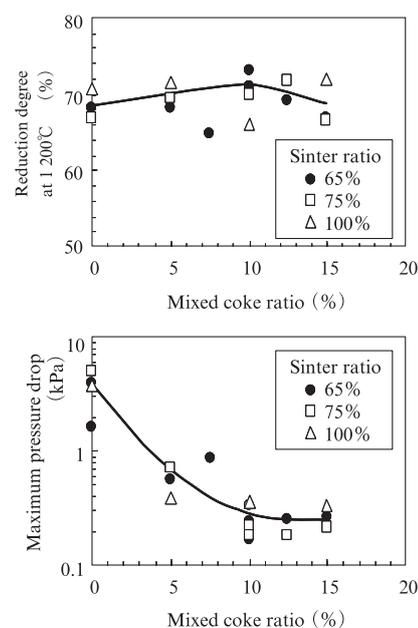


Fig. 5 Effects of mixed coke ratio and sinter ratio on the reduction degree at 1 200°C, and on high temperature gas permeability of burden material

mixed coke ratio distribution in the radial direction was studied with a numerical model and in cold model experiments. Based on the results, at the commercial blast furnace, a policy was adopted in which the coke and ore were divided into two batches each, and lump coke was mixed in the first ore batch and small to medium lump coke was mixed in the second ore batch. As the method of charging lump coke in the first ore batch, the 3-parallel bunker system at No. 6 Blast Furnace at East Japan Works (Chiba), JFE Steel was used. The target was to secure uniform charging including the

Table 1 Comparison of the operational data for conventional charging (period A) and coke mixed charging

		Conventional charging (Apr. 6–May 8, 2004) Period A	Coke-mixed charging (May 10–June 21) Period B	After No. 5 Blast Furnace blown off (July 1–11) Period C
Operation	Productivity (t/m ³ ·d)	2.06	2.18	2.39
	Reducing agent ratio (kg/t)	517.8	495.8	495.7
	Coke ratio (kg/t)	412.4	396.1	395.1
	η_{CO} (= CO ₂ /(CO+CO ₂)) (%)	47.3	49.9	49.8
	Shaft efficiency (%)	87.5	91.0	90.4
	Gas permeability index	0.993	0.927	0.913
	Stack heat loss (GJ/h)	40.6	31.8	29.5
Burden	Agglomerate ratio (%)	72.9	74.1	74.7
	No. 4 Sinter ratio (%)	49.4	45.4	39.5
	Mixed-coke ratio (kg/t)	60.0	109.0	116.3
	Tumbler index of coke 400–6 (%)	84.4	85.3	84.9
Tapping	Si Content in metal (%)	0.288	0.305	0.268
	(MnO)/[Mn]	1.31	1.13	1.17
	Tapping (d ⁻¹)	7.53	7.43	7.45

radial direction, while preventing separation/segregation of coke when simultaneously discharging sinter and lump coke from each top bunker. For this, reverse tilting charging was performed, as it is difficult to charge burden materials from the furnace center to the periphery. The aimed coke mixing ratio was 3.75 mass% in both of the batches, for a total of 7.5 mass% (mixed coke ratio: 120 kg/t).

Table 1 shows the results of operation during high mixed coke charging at No. 6 Blast Furnace, East Japan Works (Chiba) prior to blowing-out No. 5 Blast Furnace at the same works. In comparison with conventional operation (Period A), during high mixed coke charging (Period B), an RAR decrease of 22 kg/t was achieved as a result of improved gas utilization efficiency (shaft efficiency), improved gas permeability, and reduced heat loss. During Period C, when increased blast furnace productivity was required after No. 5 Blast Furnace was stopped, it was possible to continue low RAR operation, even under the condition of a reduced No. 4 sinter ratio, thereby confirming the effect of high mixed coke charging.

6. Technology for Utilization of Injected Hydrogen-based Reducing Agents into Blast Furnace⁷⁾

6.1 Background of Research

In addition to techniques for RAR reduction at the blast furnace by improvement of reduction efficiency, reduction of heat loss, and use of metallic iron, injection of reducing agents with a high hydrogen content, for example, waste plastics (C_nH_m) or natural gas (main component: CH₄) is considered effective. In order to utilize these high-hydrogen reducing agents effectively in the blast furnace, simultaneous injection with pulverized coal (PC), which is already injected through the tuyeres in normal operation, is considered. However, little quantitative information was available concerning the effect of hydrogen injection on pulverized coal combustion, or the effect of hydrogen on the reduction behavior of the burden material in the blast furnace.

6.2 Outline of Developed Technology

In order to clarify the effect of a gaseous reducing agent (CH₄) on the combustibility of solid reducing agents (pulverized coal, waste plastics), combustion experiments were carried out using a hot model. The particle diameter of the pulverized coal used in the experiments was $-74\ \mu\text{m}$, 80%, and that of the waste plastics was approximately 4 mm. The experiments were conducted at a constant blast temperature of 1 200°C, and oxygen enrichment was set in the blast so as to

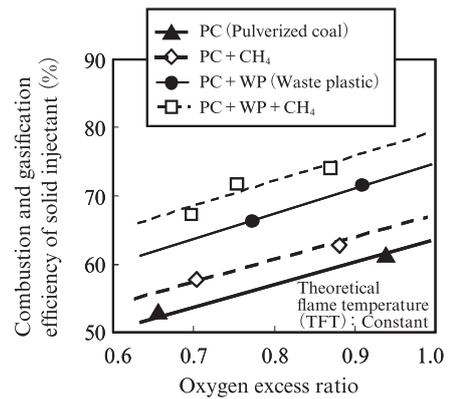


Fig. 6 Effect of methane gas injection on combustion and gasification efficiency of solid injectants

maintain a constant theoretical flame temperature (TFT) at the tuyere nose. As injection conditions, two cases were examined, one using only a solid reducing agent, and the other with simultaneous injection of the solid reducing agent with CH₄. The relationship between the excess oxygen ratio (ratio of oxygen in the blast and the amount of oxygen necessary for perfect combustion of 100% of the injected reducing agents) and the combustion and gasification efficiency of the solid reducing agents was investigated. Combustion and gasification efficiency is obtained experimentally from the carbon (C) balance in the hot model and is a total value for the solid reducing agents.

Figure 6 shows the relationship between the excess oxygen ratio and combustion and gasification efficiency of solid injectants. In comparison with single combustion of the solid injectants, combustion and gasification efficiency of solid injectant in case of simultaneous injection of CH₄ and solid injectant increased by approximately 4% in the case of pulverized coal and by approximately 5% in the case of simultaneous injection of pulverized coal and waste plastics. These increases are thought to be due to acceleration of the series of combustion gas and gasification reactions, namely, ignition and combustion of CH₄, which is a gaseous reducing agent with a high combustibility, immediately after injection, increased atmospheric temperature, heating up and volatilization of pulverized coal and plastics, combustion of the volatile fraction, and combustion of the solid fraction. Since the amount of generated unburned char decreased as a result of this improvement in combustion and gasification efficiency, the phenomenon of accumulation of these substances in the deadman and consequent deterioration of permeability in the lower part of the blast furnace, is also expected to decrease.

Next, the effect of the hydrogen concentration in the reducing gas on the reduction behavior of sinter was investigated by softening and reduction experiment under load test. **Figure 7** shows the effect of hydrogen

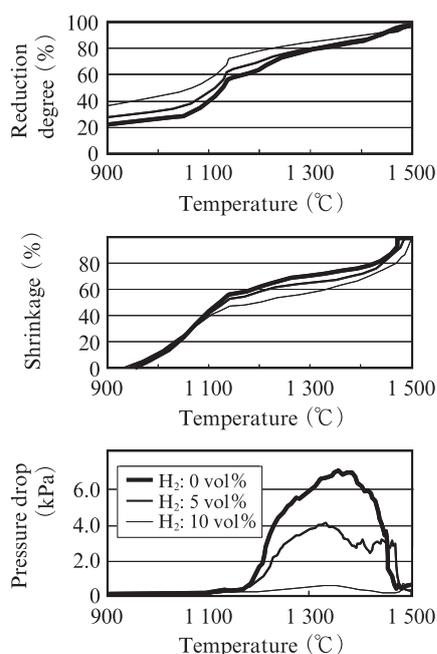


Fig. 7 Effect of gas composition on reduction degree, shrinkage, and pressure drop

on the reduction degree and shrinkage of sinter and pressure drop when the hydrogen level was changed to 3 levels (0, 5, 10 vol%). An increase in the reduction degree, a decrease in shrinkage, and a remarkable decrease in pressure drop were observed at the same temperature as the hydrogen concentration increased. This is considered to be caused by an increase in the reduction rate of the sinter by hydrogen and a decrease in the amount of FeO that remained unreduced until reaching the softening and melting temperature zone. In other words, these changes are considered to occur because the amount of slag holdup containing FeO in the cohesive layer decreased, and as a result, voids were secured in the cohesive layer.

6.3 Results of Application to Actual Blast Furnace

City gas injection was begun at East Japan Works (Keihin) in December 2004 using a natural gas-based

Table 2 Comparison of operating results before and after natural gas (NG) injection

		Before NG injection	After NG injection
NG rate	(kg/T)	0	30
Productivity	(T/m ³ ·d)	2.34	2.52
O ₂ Enrichment	(%)	4.7	5.6
Permeability index at upper part		0.23	0.23
Permeability index at lower part		1.22	1.11
CO ₂ Emission		100	97.7

gas with an adjusted composition. **Table 2** shows a comparison of the operating results before and after the start of city gas injection. The city gas injection rate was set at 30 kg/t, and O₂ enrichment was adjusted so as to maintain a constant TFT. Although permeability in the upper part of the furnace was on the same order as before injection, it can be understood that permeability improved greatly in the lower part of the blast furnace. This was attributed to the above-mentioned acceleration of reduction, securing of voids in the cohesive layer, etc. By utilizing this permeability improvement, a productivity increase of approximately 10% was achieved. The CO₂ mitigation effect under a constant production rate was estimated at approximately 2.3%.

7. Development of New Blast Furnace Raw Material (Carbon Iron Composite)⁷⁾

7.1 Background of Research

In order to achieve a drastic reduction in RAR of the blast furnace, a new blast furnace raw material called Carbon Iron Composite (hereinafter, CIC) was developed. CIC is a kind of formed coke containing metallic iron. Due to the catalytic effect of iron particles, it has remarkably high reactivity with CO₂ gas in comparison with conventional coke. The aim is to reduce the temperature of the thermal reserve zone and thereby decrease RAR by using this high reactivity CIC in the blast furnace.

7.2 Outline of Developed Technology

Photo 1 shows the appearance of CIC produced in the laboratory. As raw materials, non- or slightly-caking coal and iron ore were blended at a ratio of 7 : 3, formed into briquettes, and carbonized. The iron contained in CIC is virtually all reduced to metallic iron in the carbonization process.

Figure 8 shows the results of a comparison of the reactivity of CIC and conventional coke with CO₂ gas under the conditions assumed in the blast furnace. It can be understood that CIC reacted with CO₂ gas from a temperature approximately 150°C lower than the conventional coke. Accordingly, there is a possibility that

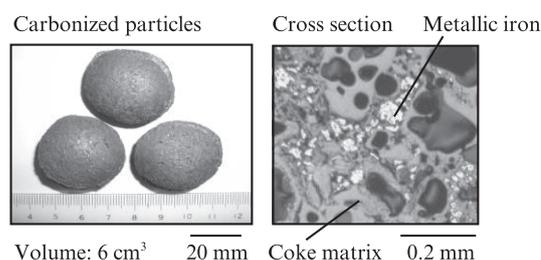


Photo 1 Appearance of Carbon Iron Composite

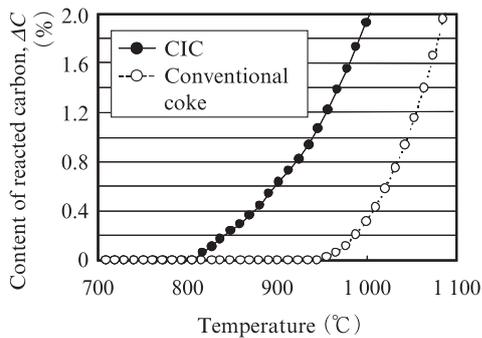


Fig. 8 Comparison of reaction initiating temperature between Carbon Iron Composite (CIC) and conventional coke

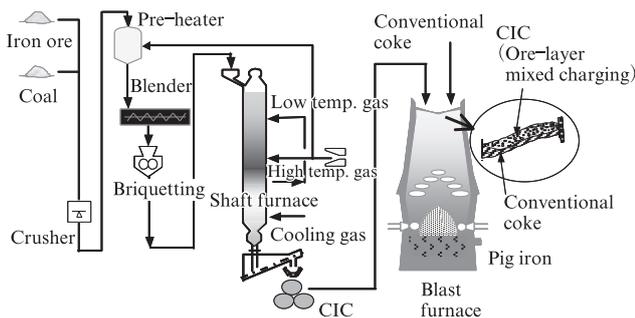


Fig. 9 Process flow for the Carbon Iron Composite (CIC) production

the temperature of the thermal reserve zone in the blast furnace can be reduced by using CIC as a partial substitute for conventional coke as blast furnace feed. According to a calculation by a numerical model of the blast furnace, an RAR reduction of approximately 30 kg/t in the blast furnace can be expected by using this CIC (100 kg/t).

Figure 9 shows the process flow of this technology. Carbon Iron Composite (CIC) is produced by crushing, blending, and briquetting inexpensive iron ore and non- or slightly-caking coal, followed by heating and carbonization in the shaft furnace. Although the strength of CIC is an important property for blast furnace feed, there is a possibility that CIC with strength on the same level as conventional coke can be produced, even from low quality raw materials, by the densification effect of briquetting and comparatively high accuracy temperature control by the shaft furnace. The assumed method of using CIC in the blast furnace is mixing in the ore

layer. A concept of functional differentiation is supposed, in which CIC is used to control the temperature of the thermal reserve zone and conventional coke maintain gas/liquid permeability. The ore reduction reaction is promoted by CIC, by using the higher reactivity of CIC and the fact that solution loss reaction of CIC begins from a lower temperature. Accordingly, a CIC use ratio of 80–100 kg/t, which corresponds to the solution loss reaction in the blast furnace, is considered appropriate.

8. Conclusion

In this paper, CO₂ mitigation processes developed at JFE Steel were introduced. Although CO₂ mitigation cannot be achieved in a short period of time, reductions can be achieved gradually by an accumulation of individual technologies developed over a 10 year span, as illustrated in this paper.

From the short- to mid-term perspective, it may be necessary to reduce CO₂ by horizontal application of technologies described here, which have been applied to actual equipment. From the long-term perspective, it is hoped that new blast furnace raw material production processes, represented by the development of CIC, will be completed, and this will lead to a drastic reduction in the reducing agent rate in the blast furnace and a large reduction in CO₂ emissions from the steel works.

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