

# Innovative Technologies to Mitigate CO<sub>2</sub> Emissions during Ironmaking

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## Abstract

*The predominant quantity of CO<sub>2</sub> emissions from the steel industry is generated by iron-making. This study introduces three technologies successfully developed for lowering CO<sub>2</sub> emissions by achieving the high-efficiency blast furnace process, enhancing the reactivity of sintered ores, surmounting the limitations due to the reduction equilibrium, and increasing the productivity of coke ovens. The reducing agent rate of the blast furnace was lowered by decreasing the SiO<sub>2</sub> content of sintered ores and by decreasing the temperature of the thermal reserve zone in the blast furnace using the newly developed agglomerate, RCA. Energy savings in cokemaking were achieved by the implementation of the SCOPE21 process. These technologies decreased annual CO<sub>2</sub> emissions significantly. Further research and development is required, aiming at the complete decarbonization of the steel industry.*

## 1. Introduction

In order to reduce global warming, it is necessary to reduce CO<sub>2</sub> emissions. In 2016, the Japanese Cabinet approved the “Global Warming Prevention Plan”, which aims to reduce 80% of greenhouse gas emissions by 2050. The Japan Iron and Steel Federation (JISF) has also formulated a vision of measures to achieve “zero carbon steel” by 2100, and is strengthening its efforts.<sup>1)</sup> The reason for this is that Japan emits 3.5% of the world’s CO<sub>2</sub>, 15% of which is from the steel industry.<sup>2)</sup> In particular, the ironmaking process, which uses coal as the raw material, accounts for the majority of the emissions, and there are high expectations for CO<sub>2</sub> reduction. The Japanese steel industry has been developing energy-saving technologies since the first oil crisis, and its energy efficiency is among the highest in the world. Contribution to the CO<sub>2</sub> emissions reduction on a global scale requires further development of innovative technologies.

There are three main ways to reduce CO<sub>2</sub> emissions in the iron-making process: low-carbon blast furnace (improvement of reaction efficiency, avoidance of reduction equilibrium constraints, reduction of reduced oxygen, H<sub>2</sub> gas injection, top gas circulation, oxygen blast furnace), shift to CDA (Carbon Direct Avoidance) process (hydrogen reduction such as shaft furnace and fluidized bed), and utilization of CCU (use of biomass and waste plastic).<sup>3)</sup> Furthermore, improved efficiencies of existing processes also contribute to CO<sub>2</sub>

reduction through energy conservation.

In this paper, the COURSE50 project, which is equivalent to H<sub>2</sub>-based gas injection, and the use of waste plastics, which is equivalent to CCU, is discussed in separate papers, and the major CO<sub>2</sub> reduction cases of Nippon Steel Corporation to date are examined, focusing on the improvement of blast furnace reaction efficiency using highly reactive sintered ore, the avoidance of reduction equilibrium constraints, and the improvement of coke productivity.

## 2. Improvement of Blast Furnace Reaction Efficiency by Highly Reactive Sintered Ore (Development of Low-slag Sintered Ore)

The energy saving technology in the sintering process can be broadly classified into the reduction of fossil fuel consumption in the sintering process and the reduction of the reducing agent rate through the reduction of permeability resistance in the blast furnace by reducing the slag content of the sintered ore. In this paper, we focus on the fact that the softening-melting property of sintered ore is related to the permeability of the blast furnace cohesive zone, and introduce a case study of improving the softening-melting property of sintered ore by reducing the amount of SiO<sub>2</sub> in the sintered ore. This paper also describes the verification of the reduction of furnace permeability resistance by an experimental blast furnace (EBF) and the evaluation of the actual furnace.

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2.1 Design for production of low slag sintered ore  
2.1.1 Sample and evaluation method

The effects of sinter composition (CaO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, MgO, and FeO) on high-temperature properties were investigated. The sintered ore was prepared in the laboratory using a sintering test apparatus with a diameter of 300 mm and a height of 500 mm. The components of the sintered ore were CaO from limestone, SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> from iron ore, MgO from dolomite, and FeO from coke breeze, and the amounts of these components were adjusted. The high-temperature ventilation resistance index of the sintered ore is calculated from the pressure drop measured by the softening-melting test (reduction test during elevating temperature under load) shown in Fig. 1, and is evaluated by the KS value.<sup>4)</sup>

2.1.2 Results and discussion

As shown in Figs. 2 and 3, decreasing SiO<sub>2</sub> (CaO/SiO<sub>2</sub> 1.8 constant, therefore CaO also decreased) and increasing MgO were effective in lowering the KS value.<sup>4)</sup> The influence of Al<sub>2</sub>O<sub>3</sub> and FeO was negligible. In order to reduce the KS value, it is necessary to suppress the formation of the liquid phase of slag in the reduction process of sintered ore. The slag composition of the sintered ore ranges in the FeO-saturated region in the CaO-SiO<sub>2</sub>-FeO phase diagram.<sup>4)</sup> Therefore, the decrease of CaO and SiO<sub>2</sub> is effective in reducing the amount of liquid phase, which is consistent with the results in Fig. 2.

On the other hand, the improvement of reducibility leads to the decrease of unreduced FeO, and the composition of sintered ore shifts to the calcium silicate saturation region in the phase diagram. In this region, liquid phase formation can be suppressed because there is no liquid phase at 1200°C. It was also found that the formation of calcium silicate (CaO-SiO<sub>2</sub>-FeO), which is an irreducible mineral, could be suppressed by using dolomite (CaO-MgO) as the MgO source instead of SiO<sub>2</sub>-bearing serpentine. We consider that the suppression of the formation of this irreducible mineral promoted the reduction and suppressed the formation of the CaO-SiO<sub>2</sub>-FeO liquid phase.

2.2 Blast furnace evaluation of low slag sintered ore

2.2.1 Test conditions

The reduction of SiO<sub>2</sub> and CaO in the sintered ore and the increase of MgO by dolomite cause the KS value to decrease. The improvement of the permeability in the blast furnace (hereafter referred to as KR) by decreasing the KS value was evaluated in the EBF. Figure 4 shows the main specifications of the EBF.<sup>5)</sup> The inner diameter of the furnace at the tuyere level is 0.9 m, and it has three tuyeres. The amount of blast furnace slag was changed by two types

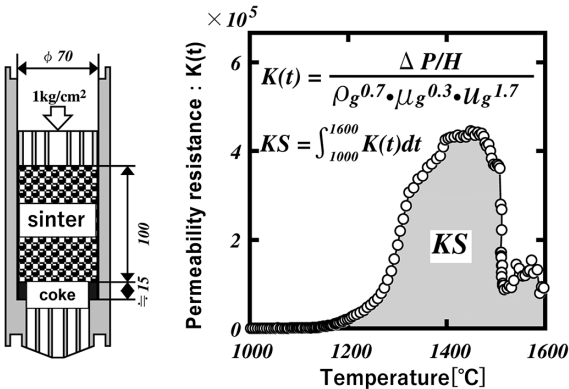


Fig. 1 Schematic diagram of experimental device for sinter softening property test

of sintered ores (SiO<sub>2</sub> 5.0%, KS×10<sup>5</sup> 1500, and SiO<sub>2</sub> 3.9%, KS×10<sup>5</sup> 648) prepared in an actual sintering machine and by the blending amount of flux.

2.2.2 Test results

As shown in Fig. 5, the reduction of the KS value of sintered ore and the reduction of the blast furnace slag content were effective in improving the KR of the blast furnace cohesive zone and dropping zone.<sup>5)</sup> The former effect was particularly significant. Furthermore, the results of the EBF operation using high and low slag sintered ore showed that the use of low slag sintered ore (RI 72.3%, KS×10<sup>5</sup> 1104) had not only the effect of decreasing KR, but also the effect of decreasing the reducing agent rate compared to the use of high slag sintered ore (RI 65.3%, KS×10<sup>5</sup> 1508).<sup>6)</sup>

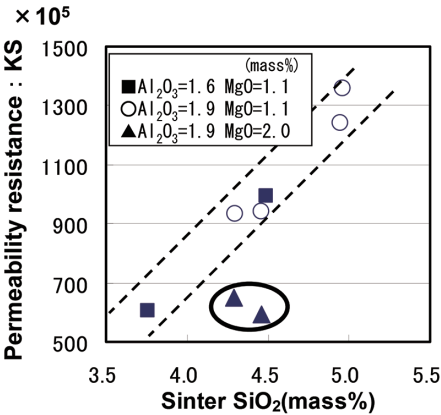


Fig. 2 Effects of SiO<sub>2</sub>, MgO, and Al<sub>2</sub>O<sub>3</sub> concentration in sinter on permeability resistance (KS)

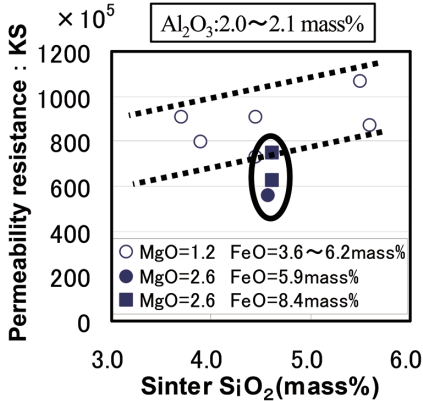


Fig. 3 Effects of SiO<sub>2</sub>, MgO, and FeO concentration in sinter on permeability resistance (KS)

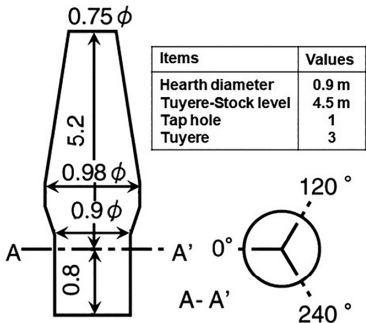


Fig. 4 Configuration of experimental blast furnace

### 2.2.3 Verification in commercial blast furnace

Ventilation analysis of the sintered ore and coke layers<sup>7)</sup> was conducted to verify the consistency with the KR of the commercial furnace. As a result, as shown in Fig. 6, the improvement of KR by lowering the KS value was confirmed in the commercial blast furnace, and the measured value and calculated value were in good agreement.<sup>9)</sup> The reducing agent rate in the furnace was reduced by 3 kg/t-HM due to the effect of KR improvement.<sup>8)</sup> This is equivalent to a CO<sub>2</sub> reduction of 200 000 tons/year if this is applied to all of the Steel Works of Nippon Steel. After this evaluation, all the plants promoted the reduction of slag in the sintered ore, and continued to achieve less than 5% SiO<sub>2</sub> in the sintered ore for more than 10 years. In particular, East Nippon Works (Kashima) produced an extremely low slag sintered ore with SiO<sub>2</sub> of 4.34% in 2005.

## 3. Avoidance of Reduction Equilibrium Constraints (Reduction of Reducing Agent Rate of Blast Furnace by Carbon-containing Agglomerate)

### 3.1 Principle of reduction of reducing agent rate and reaction evaluation

Reduction of the reducing agent rate of the blast furnace is effective as a means of CO<sub>2</sub> reduction in the ironmaking process. In re-

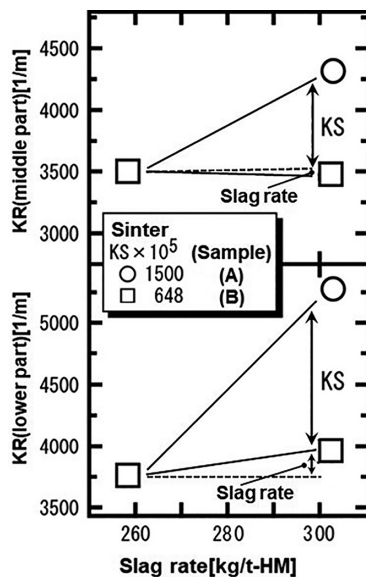


Fig. 5 Comparison of effect of KS on KR with that of slag rate

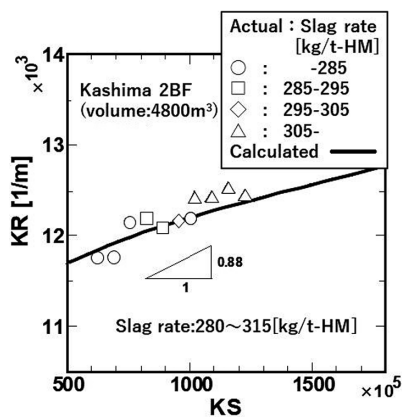


Fig. 6 Effect of KS on gas flow resistance in commercial blast furnace

cent years, a variety of reactive agglomerates have been proposed to control the reduction equilibrium condition (point W on the RIST diagram), which is one of the limiting conditions for the reaction efficiency of the blast furnace. One of the means to control the reduction equilibrium condition in the blast furnace is to lower the thermal reserve zone temperature through improvement of carbon reactivity, and various raw materials for the blast furnace have been proposed so far (Fig. 7).<sup>9)</sup> One of them is Composite Agglomerate (CA), which is composed of fine carbonaceous materials and fine iron oxide, and the high reactivity of CA has been understood for a long time. Nippon Steel has discovered the excellent effect of reduction of the reducing agent rate in a composite agglomerate called the Reactive Coke Agglomerate (RCA),<sup>10)</sup> which has a higher carbon content and more enhanced functionality than conventional agglomerates, and has commercialized RCA.<sup>11)</sup> RCA has high carbon reactivity and reducibility due to the coupling phenomenon between the gas reduction reaction of iron oxide (Equation (1)) and the gasification reaction of carbon (Equation (2)).



Figure 8 shows the results of the test using the Blast Furnace Inner-reaction Simulator (BIS furnace), which can reproduce the counter-current reaction in the blast furnace shaft.<sup>9)</sup> The temperature of the thermal reserve zone was measured using various agglomerates (CA, ferro-coke, and nut coke), and the influence of the distance “L<sub>OC</sub>”, the distance between the iron oxide and carbon in the agglomerates was observed: the shorter the L<sub>OC</sub>, the lower the temperature of the thermal reserve zone. It was shown that the proximity of the two materials by mixing and compacting fine carbonaceous

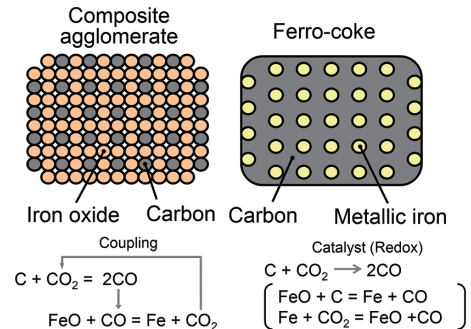


Fig. 7 Innovative agglomerates for surmounting operational limit due to reduction equilibrium of blast furnace

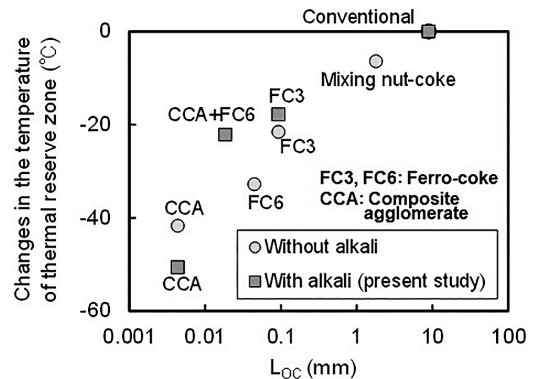


Fig. 8 Relationship between changes in temperature of TRZ (results of BIS test) and L<sub>OC</sub> (average distance between iron oxide and carbon)

materials and fine iron oxide, as in the case of CA, contributes to the above coupling phenomenon and leads to a significant decrease in the temperature of the thermal reserve zone.

Figure 9 shows a comparison of the reaction behavior of various agglomerates with different gas conditions ( $\text{CO}_2/(\text{CO}+\text{CO}_2)$ ) when heated to  $1200^\circ\text{C}$ .<sup>9)</sup> The carbon consumption rate of all agglomerates was higher than that of normal coke and increased with the increase of  $\text{CO}_2/(\text{CO}+\text{CO}_2)$ , but RCA had the largest increase in the carbon consumption rate (Fig. 9(a)). The reduction rate of ferrocoke and RCA remained above the 90% reduction rate, although it slightly decreased during the high  $\text{CO}_2/(\text{CO}+\text{CO}_2)$  condition, and the agglomerates were not oxidized under this heating condition, even though FeO was stable in the heating condition (Fig. 9(b)).

Figure 10 shows a comparison of the reduction progress between RCA and sintered ore.<sup>12)</sup> The reduction of RCA proceeded from a lower temperature than that of sintered ore, and it showed high reducibility. From the calculation results of the mathematical model of the blast furnace using the reduction rate parameters obtained in this experiment, the relationship of the reducing agent rate decrease of 0.36 kg-C/tHM per 1 kg-C/tHM of RCA-derived carbon, which was obtained in the RCA blast furnace test, was validated. A reducing agent rate decrease of up to 4 kg-C/tHM<sup>10)</sup> was also validated.

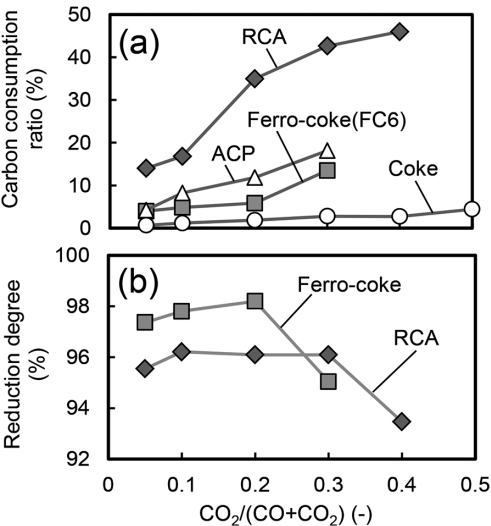


Fig. 9 Changes in CCR (a) and reduction degree (b) of agglomerates after reaction until  $1200^\circ\text{C}$  with respect to inlet-gas composition

3.2 Implementation of carbon-containing agglomerate RCA

RCA was implemented in Kyushu Works (Oita) in November 2011. The production flow of RCA is shown in Fig. 11.<sup>11)</sup> The raw materials are coarsely crushed, mixed with cement, and granulated in a pelletizer. The raw pellets are subjected to primary curing using steam and then transported to the yard for secondary curing. The amount of RCA produced is about 900 t/d, and it is used in a large blast furnace (furnace volume:  $5775\text{ m}^3$ , two units) at a maximum rate of 40 kg/tHM. Table 1 shows the changes in the operational parameters before and after the use of RCA.<sup>11)</sup> The use of RCA resulted in a reduction in the reducing agent rate that was almost the same

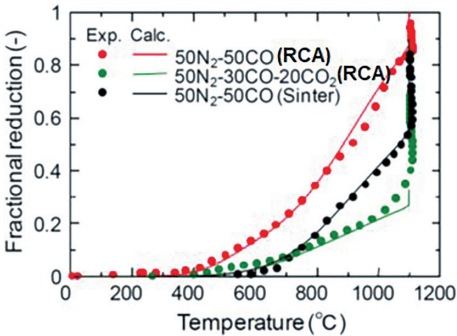


Fig. 10 Comparison of fractional reduction curve of RCA with that of sinter

Table 1 Operational change of Oita No.1 blast furnace between before and after RCA use

		Without RCA	With RCA	Difference
Production	t/d	13 554	13 815	+261
Reducing agent rate *		490.2	487.6	-2.6
Coke rate	kg/tHM	338.7	324.7	-14.0
PCR	kg/tHM	151.5	162.8	+11.3
Ore composition				
Sinter	%	82.8	76.5	-6.3
Pellet	%	1.7	4.9	+3.2
RCA	%	0	2.1	+2.1
Hot metal temperature	$^\circ\text{C}$	1 530	1 537	+7
Horizontal shaft probe data				
Temperature	$^\circ\text{C}$	687	672	-15
$\text{CO}_2/(\text{CO}+\text{CO}_2)\cdot 100$	%	37.1	38.8	1.7

\*RAR includes carbon in RCA.

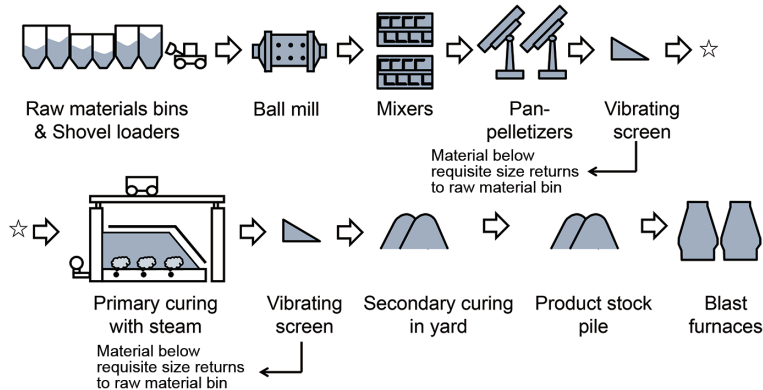


Fig. 11 Production flow sheet of RCA implemented in Kyushu Works (Oita)



as that of the previous findings<sup>10, 12)</sup>. From the mid-shaft probe data, the gas reduction efficiency increased with the use of RCA for the same temperature level, suggesting that the coupling reaction (carbon proximity effect) improved the reduction efficiency at the shaft. Furthermore, the rapid curing method using steam curing was also implemented, and a strength of 100 daN/p was obtained in a curing period of 4 days, contributing to the stabilization of the logistics and quality of the RCA.

The idea of lowering the thermal reserve zone temperature to avoid the reduction equilibrium constraint has been around since the invention of the RIST diagram. However, it had not been put into practical use because of the difficulty in optimizing the conditions considering the reaction rate. Nippon Steel has succeeded in developing a novel agglomerate that can reduce the reducing agent rate in the blast furnace by using a tool that can precisely reproduce the reduction conditions in the shaft, optimizing the conditions of the reduction process, designing novel agglomerate, and optimizing the actual production method. This method is currently being applied to six blast furnaces, including large ones in three works, and is contributing to a total reduction of 100 000 tons/year of CO<sub>2</sub> through the reduction of the reducing agent rate of the blast furnace.

## 4. Coke Productivity Improvement (Development of Innovative New Coke Oven SCOPE21)

### 4.1 Overview of the SCOPE21 process

The Super Coke Oven for Productivity and Environmental enhancement toward the 21st century (SCOPE21) was developed as a next-generation innovative process based on a slot type coke oven, before the time of new coke oven construction. It was developed from 1994 to 2003 as a national project with the participation of companies in the Japan Iron and Steel Federation.<sup>13)</sup> Based on the results, the new coke ovens were installed in Kyushu Works (Oita) and Nagoya Works. The development of this process was conducted with four goals in mind: effective utilization of coal resources by greatly increasing the ratio of low-grade coal (non- or slightly-coking coal), significant improvement of coke oven productivity, and improvement of energy conservation and environmental responsiveness.

Figure 12 shows schematic diagram of the SCOPE21 process flow installed at Nippon Steel,<sup>14)</sup> and Table 2 shows the specifications of the main equipment. Compared with the conventional coke-making process, the SCOPE21 process is characterized by the advanced pre-processing of coal. The coal used for coke production is first dried and heated rapidly in the pretreatment process. Similar to the conventional cokemaking process, the coal is blended and crushed, then dried and preheated in a fluidized bed dryer-classifier, and the raw coal is classified into coarse-grained coal and fine coal.

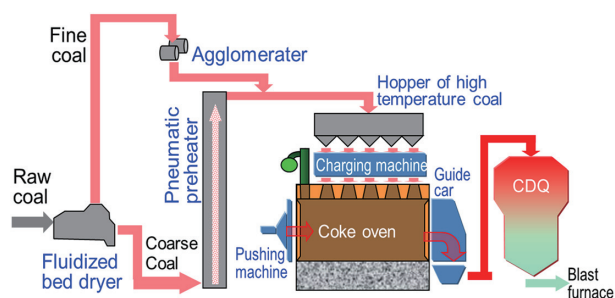


Fig. 12 Schematic diagram of SCOPE21 process flow in Kyushu Works (Oita) and Nagoya Works

The classified coarse coal is rapidly heated to about 330–380°C in a pneumatic preheater. The classified fine coal is agglomerated in heat, mixed with the rapidly heated coarse coal, and charged into the coke oven at high temperature for carbonization. The coke discharged from the coke oven is sent to the blast furnace after heat recovery and cooling in the CDQ.

During the pretreatment process, especially the rapid heating process, the coal is modified and its coking property is improved. As a result, coke with sufficient strength can be produced even if the ratio of non- or slightly-coking coal, which is usually unsuitable as a raw material for the conventional cokemaking process, is greatly increased.

In addition, by drying and preheating the coal and charging it into the coke oven at a high temperature, the processing time in the coke oven, i.e., the coking time, which requires a long time in the conventional process, can be significantly shortened. This greatly improves the productivity of the coke oven and leads to energy conservation.

The coke oven is a batch-type facility, and the coal charged into the coking chamber is heated by indirect heating from the adjacent combustion chamber where fuel gas is burned for carbonization. Due to the low thermal conductivity of coal, poor heating efficiency has been a problem. Figure 13 shows schematically the heating conditions in the coke oven of the conventional process and the SCOPE21 process. In the conventional process, the charge coal contains nearly 10% moisture, and it takes about 8 to 10 of the 18 hours of coking time just to evaporate the moisture out of the coal. In the SCOPE21 process, coal is rapidly dried and heated in the coal pretreatment process before charging the coal into the coke oven, thus shortening the coking time in the coke oven.

In the SCOPE21 process, coal particles are directly heated by gas in the fluidized bed dryer-classifier and pneumatic preheater, which enables efficient and short heating. In the fluidized bed dryer-classifier, it has been shown that coal (including moisture) can be heated up to 300°C in 100s to several 100s,<sup>15)</sup> depending on the operating conditions. The heating rate ranges from 30 to 100°C/min in the region above 200°C.

Table 2 Specifications of SCOPE21 process in Kyushu Works (Oita) and Nagoya Works

Fluidized bed dryer	161 t/h
Pneumatic preheater	106 t/h
Agglomerator	36 t/h × 2
Number of ovens	64
Dimensions of ovens	W 0.45 × H 6.7 × L 16.6 m
CDQ	123 t/h

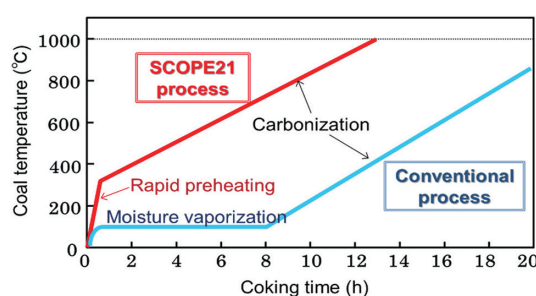


Fig. 13 Comparison of coking time between SCOPE21 and conventional process

In the pneumatic preheater of rapid heating, coarse-grained coal is heated while being accompanied by a large amount of heating gas. It has been confirmed that coal is heated rapidly at a rate of more than 5000°C/min.<sup>16)</sup>

The time required for coke production consists mostly of the coking time required for heating in the coke oven. In the case of dry and preheated high-temperature charge coals of the SCOPE21 process, the coking time in the coke oven is shortened because the evaporation of moisture is not required and the heating temperature range is smaller.

In order to achieve such a rapid coking time, the charge coal in the coke oven must be uniformly heated. In the SCOPE21 coke oven, heating uniformity in the height direction and low NO<sub>x</sub> combustion are achieved by the three-stage combustion system and the arrangements of the flue ports at the bottom of the combustion chamber.<sup>17)</sup>

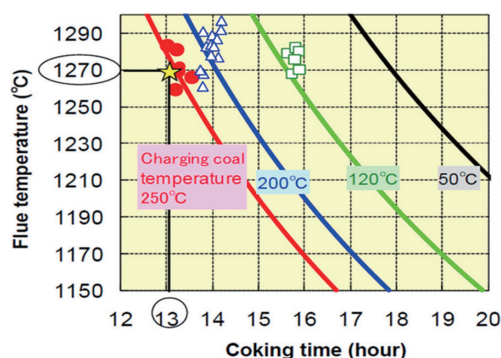
#### 4.2 Actual operation status of SCOPE21 process

The first SCOPE21 process was constructed as the No.5 Coke Oven in Kyushu Works (Oita) in 2008, based on the results of the technological development of the national project. After that, the actual operation results were accumulated, and the second unit was put into operation as the No.5 Coke Oven in Nagoya Works in 2013. Since the start of operation, the working rates of both units have been increased step by step, and the highest working rate of 184.5% was achieved, resulting in a significant improvement in productivity in actual operation.

**Figure 14** shows the actual coking time of the Oita SCOPE21 process.<sup>18)</sup> The oven temperature was set near the maximum temperature, and then the temperature of the charged coal was increased step by step. As the temperature of the charged coal increased, the coking time was shortened, and the target value of 13 h of coking time (=184.5% working rate) was achieved at the oven temperature of 1270°C with the charged coal temperature of 250°C. This result is consistent with the results obtained by the heat transfer model calculation.

Thus, as a result of the improved productivity, energy saving in the cokemaking process has also been achieved. As a result of evaluating the energy reduction for one year of operation in Oita No.5 Coke Oven, the energy reduction used for a coke production rate of 1 million tons/year corresponds to a CO<sub>2</sub> emission reduction of 260000 tons/year.<sup>19)</sup> Nagoya Works has also achieved a maximum reduction of 200000 tons/year of CO<sub>2</sub>.

The SCOPE21 process has been in operation for around 10 years, contributing to the reduction of the energy used in the Nippon Steel's cokemaking process.



**Fig. 14** Correlations between carbonization flue temperature and coal charge temperature

## 5. Conclusion

This paper has outlined three major technological developments in the effort to reduce CO<sub>2</sub> emissions by improving the efficiency of the blast furnace ironmaking process: improving blast furnace reaction efficiency using highly reactive sintered ore, avoiding reduction equilibrium constraints, and improving coke productivity.

In addition, the COURSE50 project and the utilization of waste plastics, which will be discussed in separate papers, have been earnestly pursued in the ironmaking field.

We must accelerate our efforts to achieve zero-carbon steel in the future. Nippon Steel will continue to pursue further innovative research and development toward the decarbonization of the ironmaking process with a sense of mission.

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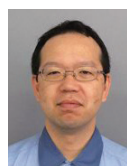
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