

Technical Developments for Saving Natural Resources and Increasing Material Recycling

Tetsuzo HAGA*
Tetsuharu IBARAKI

Kenji KATOH

1. Introduction

High quality iron ore and coal resources for the steel industry are gradually being depleted. Given this problem, Nippon Steel Corporation has developed technologies to effectively utilize low-quality resources. This paper focuses on coal drying methods (DAPS and SCOPE21) that enable the use of large quantities of non- or slightly-caking coals for cokemaking, the selective granulation of limonitic ores, and dust recycling using rotary hearth furnaces (RHF). It also describes the problems encountered during the initial investigation and commercial application of these methods and the measures taken to overcome these problems, as well as outlines some of the future prospects for these technologies.

The breakthroughs resulting from the fundamental research in these technical fields originated from a thorough examination of the basic characteristics of the low-quality resources in question, followed by the development of a process to make use of or control the characteristics in order to reform the resources. This approach very often resulted in better utilization of the low-quality resources. In terms of fundamental process technologies, each advancement began with the systematic classification, granulation and thermal treatment of solid materials of different grain sizes. Stable processes were established by addressing equipment problems that arose during field testing of the developed technologies. Because the details of the developed technologies have already been published, this article attempts to clarify the ideas that led to the breakthroughs from the viewpoint of the engineers responsible for the technical developments.

2. Coal Drying Technologies to Expand Raw Coal Resources for Cokemaking

The types of coal most suitable for producing metallurgical coke are caking (or coking) coals, but more than 70% of the world's coal resources are of low-quality, non- or slightly-caking coals, and caking coal resources are being depleted. In this situation, the development of methods for improving the coke quality that enable greater use of low-quality coals is very important. Various coal pretreatment technologies have been developed in Japan, and unique coal drying methods have been industrially applied.

The first such method was the coal moisture control (CMC) process. Japan imports coal for cokemaking from abroad, and the moisture content of imported coal is approximately 10%. The original

idea of the CMC process was to decrease the coal moisture in order to increase the bulk density of the coal charged into the coke ovens, and thus to improve the coke strength. More specifically, the coal moisture is decreased to 5 to 6% using rotary heaters. The first commercial CMC system entered into operation at Nippon Steel's Oita Works in 1983¹⁾. Theoretically, the bulk density of the charged coal increases, and so does the coke strength, when the coal moisture is decreased to less than 5%; however, at such a low moisture content, fine coal dust is generated during transport from the dryers and handling around the ovens, which adversely affects the stable operation of the coke oven.

In view of the above, measures were sought that would both suppress the dust emission from coal containing less than 5% moisture and also further increase the coke strength. Studies into the mechanisms of dust emission from dry coal made it clear that, when the moisture content was high, dust emission was low because the fine grains adhered to the coarse grains and formed pseudo-grains, with water serving as the binder.

When coal is dried to less than 5% moisture, the pseudo-grains disintegrate, and the fine particles begin to act as individual grains, causing increased dust emissions. It thus became clear that, to suppress coal dust emissions, it was necessary to dry the coal to about 2% moisture so that the pseudo-grains would disintegrate completely; the resulting grains could then be classified by size, and the fine grains could be granulated. Further studies were then conducted on the relationship between the maceral and the grain size of the coal. Of the various types of coal maceral, highly caking vitrinite is soft and easily crushable. Microscopic analysis of the coal revealed that the fine coal grains contained vitrinite in high concentrations. In addition, a study of the caking properties of the granulated fine coal made it clear that, as shown in **Fig. 1**, the caking properties increased significantly when the fine coal was granulated (to increase the bulk density of the charging coal)²⁾.

Based on the results of the above fundamental research, development of an industrial process was initiated, whereby the fine coal was pressed into briquettes in order to increase its caking properties and coke strength, while also suppressing dust emissions. The key to the successful development of this process was a facility for classifying a great amount of coal while drying it. Thus, development of a fluidized-bed type dryer was undertaken, and as a result, the world's first efficient dryer-classifier capable of handling

* General Manager, Dr.Eng., Plant Engineering Div., Plant Engineering and Facility Management Center
20-1, Shintomi, Futtsu, Chiba 293-8511

as much as 6,800 t/d of coal was designed (see Fig. 2).

This process, which was named the Dry-cleaned and Agglomerated Pre-compaction System (DAPS), was put into commercial use at Nippon Steel's Oita Works in 1992^{2,3)}. To produce coke of the same strength, the DAPS allows 30% more use of non- or slightly caking coal than the conventional wet coal process does, and 20% more than the CMC process.

As stated above, the CMC and the DAPS processes were developed with the aim of achieving more effective use of coal resources, and were established as commercially applicable processes through technical developments that enabled improved coke strength through improvement in the characteristics of the coal grains and their handling properties.

Another process for improving the caking properties of coal in order to strengthen the coke was developed from a different point of view. It was known that coal molecules are in an aggregated form. Based on this knowledge, a proposal was advanced to improve the coke strength by quickly heating the coal in order to loosen the aggregated structure, and by so doing, increase the caking properties. Based on this idea, the development of an innovative process began in 1994 as a national project that eventually came to a successful end in 2003. The target process of the project was named the "Super Coke Oven for Productivity and Environmental enhancement toward the 21st century" (SCOPE21)⁴⁾.

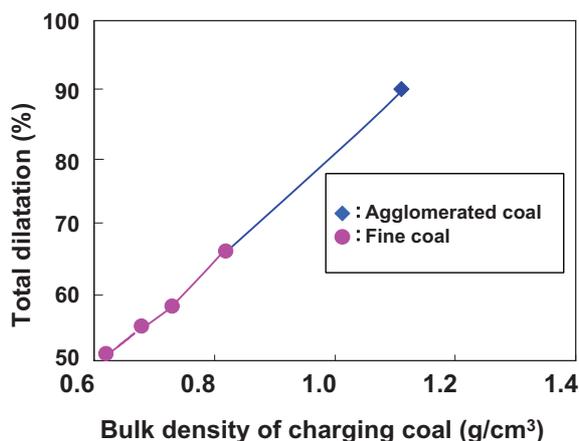


Fig. 1 Relationship between bulk density and total dilatation coefficient of fine coal

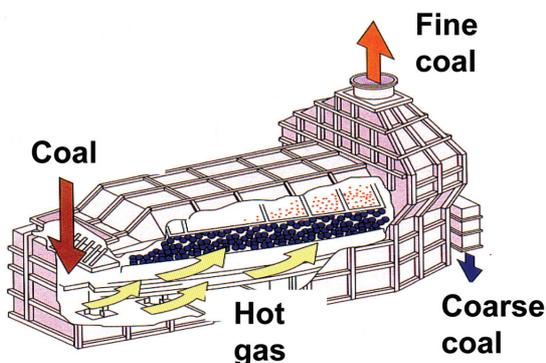


Fig. 2 Outline of fluidized bed dryer



Fig. 3 Outlook of Oita No.5 coke oven battery

In the initial fundamental research stage, the effect of rapid pre-heating of coal on the coke strength was studied. For the initial tests, a pneumatic, laboratory scale pre-heater was used to evaluate 200 g of coal. This shaft heater was 25 mm in diameter and 1.8 m in height and provided an upward flow of hot air. A series of tests using the laboratory heater made it clear that, when coal was rapidly heated to a temperature range of 330 to 380°C at a rate of about 3,000°C/min, the caking properties of the coal improved and led to an increase in the coke strength⁵⁾. Improving the caking properties of coal by rapid pre-heating was an unprecedented, innovative concept. The hardest part of the development lay in enlarging the process to a commercial scale. When the pneumatic pre-heater was enlarged, the gas flow and the heat transfer inside it changed. Laboratory tests were repeated more than 100 times in one year to define the conditions that would provide a homogeneous coal temperature during the rapid heating. Many numerical simulations were also carried out, and the range of optimum pre-heating conditions gradually became clear⁶⁾.

Thereafter, to confirm the commercial effectiveness of the developed process, a pilot plant with a 20 t-coal/h capacity was constructed at Nippon Steel's Nagoya Works, and trial operations were conducted for one year beginning in March 2002. It became clear at this stage that, by rapidly heating coal, it was possible to produce high-strength coke, even when mixing in as much as 50% non- or slightly-caking coals⁷⁾. The first commercial plant using the SCOPE21 process was then constructed at Nippon Steel's Oita Works, and entered into operation in February 2008 (see Fig. 3)⁵⁾.

It is expected that many new coke oven plants using this process developed in Japan will be constructed in and outside of the country to help the steel industry development further.

3. Selective Granulation of Limonitic Ores with Poor Sintering Properties

As the high-quality iron ore resources around the world become depleted, the importance of effectively utilizing abundant limonitic ores for steel production is increasing. Limonitic ores are ores that contain alumina and combined water in high percentages, and consist mainly of fine grains that require sintering, but that adversely affect the productivity and energy consumption of the sintering process and the quality of the sintered ore.

The selective granulation process is a method of iron ore treatment in which the limonitic ores are separated from the other types of ore.

The fine powder of the limonitic ores is then granulated so that it can serve as the cores in the formation of pseudo-particles in the drum mixer for the sinter feed, as shown in Fig. 4. This treatment prevents the alumina from adversely affecting the reactions that produce the molten liquid phase that serves as the binder during sintering. As a result, the selective granulation process enables the effective use of limonitic ores⁸⁾.

Although the name “selective granulation” suggests an agglomerating process, it is not simply a process to improve the agglomerating properties of the ore. Instead, its key purpose is the prevention of alumina from interfering with the reactions that form the molten liquid phase, because this interference is the main reason for the poor sintering properties of limonitic ores.

The poor sintering properties of limonitic ores are due mainly to two factors. First, the fine limonitic ore powder contains much alumina; in order to obtain the liquid phase that is required to form the sintered ore from the limonitic powder, it is necessary to heat it to temperatures higher than those required for other types of ore⁹⁾. Secondly, the coarse grains of limonitic ores are prone to cracking during the heating step of the sintering process due to the evaporation of the combined water that is inside them, which lowers the strength of the sintered ore. These problems occur at high temperatures, and it was necessary to work out measures to radically improve the sintering reactions of this type of ore.

Fig. 5 compares limonitic and hematite ores in terms of their alumina and combined water contents at different size ranges; limonitic ore contains more of both than hematite ore does. In particular, with respect to the fine powders that form the molten liquid phase only at high temperatures, the limonitic ore samples (Australian ores A and B), had a high content of both clayey gangue and alumina. To evaluate the difference in the liquid phase forming reactions of the fine powders of the two ore types, samples were melted in a laboratory infrared heater after the addition of 20% lime stone. Fig. 6 shows the results; the temperature at which the limonitic sample containing as much as 3.2% alumina formed the liquid phase was about 100°C higher than that of the hematite sample containing 0.5% alumina, and the fluidity of the liquid phase of the limonitic sample was poorer than that of the hematite sample¹⁰⁾. These results indicated that the sintered ore made from the limonitic ore was likely to have poorer bonding properties and lower strength than that made from the hematite ore. The idea for selective granulation originated from the question: Wouldn't it be possible to block the alumina in the fine

powders of the limonitic ore by granulating them?

To granulate the fine powders of limonitic ore, it is necessary in the first place to separate them from the coarser grains. The part of the ore responsible for the formation of the liquid phase are the very fine powders of 0.5 mm in diameter or less, but these powders contain clayey gangue in high percentages, and therefore, it is difficult to separate them from the rest of the grains by sieving, especially on a scale that is meaningful for a sintering plant, which is hundreds of tons per hour. Various tests were therefore conducted to evaluate different methods of classification, and eventually the sieving size for the actual process was set at 3 mm for the following reasons: (1) the aim was to block the alumina and not to classify powders of 0.5 mm or less in size; and (2) it became clear that it was possible to block these fine powders with a high alumina content by using grains of 0.5 to 3 mm in size as the cores for the granulation. Sizing the ore to <3 mm using undulating urethane screens proved viable without causing clogging.

Because the portion of the fine limonitic ore that is <3 mm in size contains plastic clayey gangue, it can be easily granulated by kneading with water without the addition of a binder. It can then be pelletized into 2 to 5 mm core grains that are strong granules

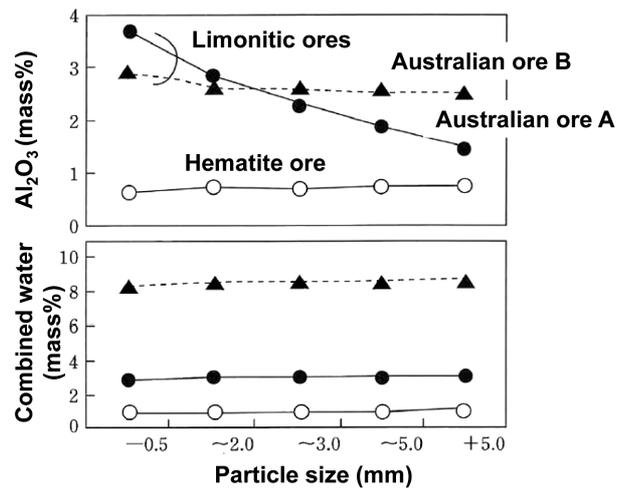


Fig. 5 Distribution of chemical composition for typical iron ores

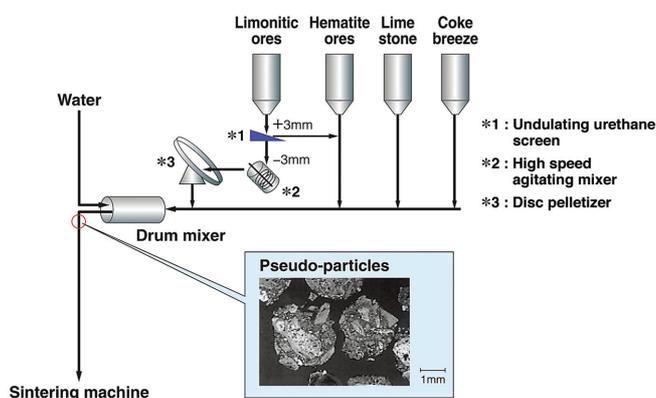


Fig. 4 Selective granulation process and the pseudo-particles

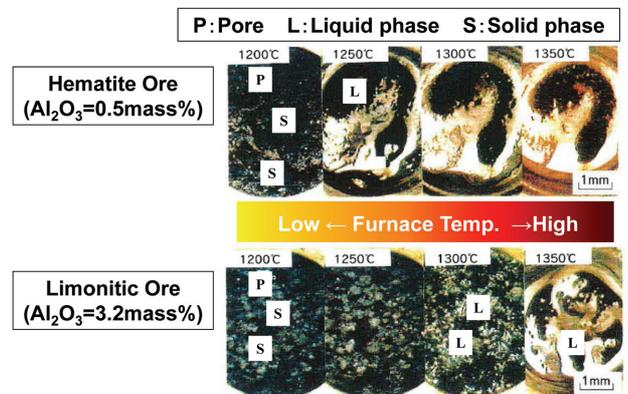


Fig. 6 Change in melt formation of hematite and limonitic ores with infra-red heating furnace

containing alumina that is blocked inside (hereinafter called the selective granules). Next, in a process of mixing and granulation with hematite and other low-alumina ores, the selective granules serve as the cores for formation of dual structure pseudo-particles. Fig. 7 schematically compares a pseudo-particle prepared by this selective granulation method with another produced by the conventional method.

The results of electron probe X-ray microanalysis (EPMA) analysis of a pseudo-particle prepared using the selective granulation method are shown in Fig. 8. Here, one can clearly see the dual structure of the pseudo-particle: a core of the selective granule composed of a high-alumina powder and a layer of adhering low-alumina fine ore with a high limestone content and coke breeze around the core.

During sintering reactions, the dual-structured pseudo-particles produce the following effects on the molten liquid phase formation: (1) the high concentration of the coke breeze secures effective heating; (2) the low alumina concentration and the high limestone concentration of the adhering fines lead to formation of a great amount of highly fluid molten liquid phase at temperatures lower than those required in the conventional method; and (3) the liquid phase penetrates the gaps between the ore grains and the voids inside of the

grains, acting as bonds, which makes the sintered ore stronger¹¹⁾.

Fig. 8 also shows the reaction between the selective granule and the liquid phase from the outer layer that takes place when a dual-structured pseudo-particle is heated. The pseudo-particle, consisting of a core of the selective granule composed of the high-alumina fine ore and an outer layer of the adhering low-alumina fine ore, is changed through the sintering process into a particle consisting of a core of hematite and calcium ferrite and an outer layer of calcium ferrite, with retention of a clearly distinguishable dual structure. The alumina content remains blocked inside the surface of the selective granule, and does not adversely affect the outer liquid phase. Assuming that the existence of the CaO indicates the presence of the liquid phase, it can be seen that the liquid phase intruded from the outer layer into the selective granule, imparting bonding strength to form the sintered structure.

The idea of blocking the alumina originated from a thorough investigation of the fundamental characteristics of the limonitic ore. Commercial facilities for selective granulation were realized, starting with fundamental studies and continuing through the process design phase. A critical point in the development of the process was the need to determine whether the microscopic granulation mechanisms could be enlarged to the scale of an operating, industrial sintering plant, where hundreds of tons of raw materials are processed every hour. The equipment for continuous and selective granulation, operating automatically, was made a practical reality through arduous testing and evaluation, and was eventually installed in many sintering plants¹²⁾.

The development of the selective granulation process made it possible to effectively utilize limonitic ore, which had been regarded as a low-grade ore because of its sintering issues. The available recoverable reserves of iron ore in the world are shrinking, and for this reason, it is necessary to develop methods for effective use of materials that require complicated pretreatment.

4. Development of a Dust Recycling System Using a Rotary Hearth Furnace

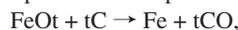
A large amount of dust, sludge and scale arise from steel production processes; these materials consist of iron oxide, carbon, and other compounds. For reasons of environmental protection and resource and energy savings, efficient methods for recycling of the oxide wastes have been sought, but conventionally, only up to 80 to 90% have been reused effectively, mainly because a portion of the oxide wastes from iron- and steelmaking processes contain elements that evaporate at high temperatures, such as zinc, lead, alkali metals, etc. To address this issue, Nippon Steel embarked upon a project to develop an energy-efficient and economical process for the recycling of oxide wastes via the economic removal of zinc.

Zinc (lead) in the dust can be removed by reducing its oxide at high temperatures into metallic zinc, and then evaporating the metal.

This reaction is expressed as:



This process accompanies another reaction that yields reduced iron:



where t is the oxygen rate with a value between 0.95 and 1.5. When the reduced iron is used as an iron source without re-oxidation, the energy consumed for the iron reduction reaction is efficiently utilized in the smelting process, and the energy loss for the entire process from oxide waste processing to smelting is minimized. Rotary kilns, submerged arc furnaces (SAFs) and rotary hearth furnaces (RHF) are used for this process. As a result of our investigations of the process

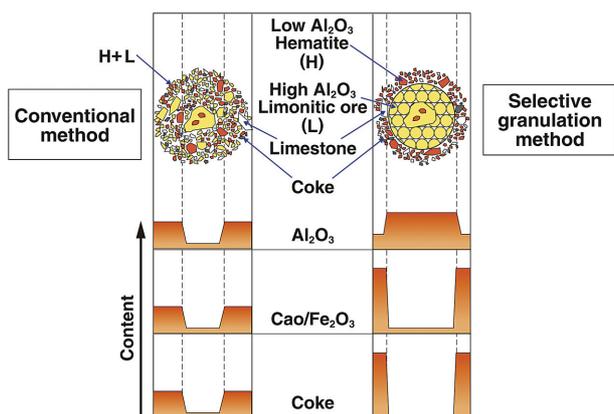


Fig. 7 Pseudo-particle structure of conventional and selective granulation method

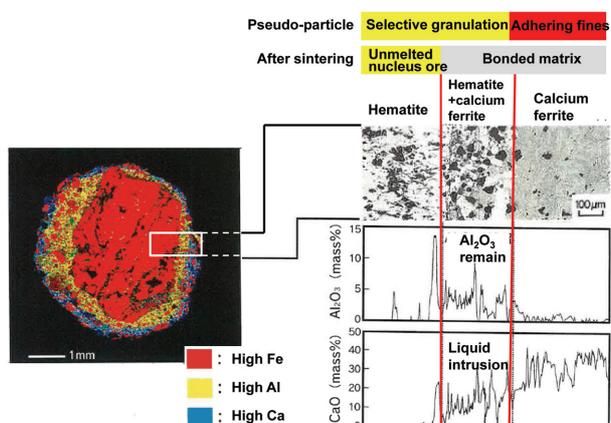


Fig. 8 Dual structure pseudo-particle and sinter structures by selective granulation method

performance, the RHF method was determined to be the most energy-efficient and economical¹³⁾.

The RHF method is a high-temperature reduction process using a furnace having a torus-shaped hearth that turns around a vertical axis, as shown in Fig. 9.

The process consists of three steps: (1) pretreatment and agglomeration of the dust and scale into pellets; (2) reduction in an RHF; and (3) exhaust gas treatment. The oxide waste is first formed into pellets containing the oxides of iron and other elements and carbon, which are then reduced in the RHF.

As shown in Fig. 10, the formed pellets are fed to the furnace so as to cover the hearth, which rotates at a constant rate and is heated first in the heating zone and then in the reducing zone. The reduction reactions take place between the oxides of iron, zinc, etc. and the carbon in the pellets.

Fig. 11 shows the in-furnace reactions of the oxide waste pellets. They are heated by the radiative heat, and the FeO and C in them react with each other to yield metallic iron and CO. Because of the rapid heat transfer by the radiative heat and high reactivity of the fine particles comprising the pellets, the reactions are completed in 10 to 20 min, and therefore the RHF exhibits a high productivity per hearth area. Through this process, the oxide waste pellets are turned directly into reduced iron (DRI) with a high concentration of metallic iron.

Although the RHF was originally developed exclusively for producing DRI, its possible application for dust treatment also

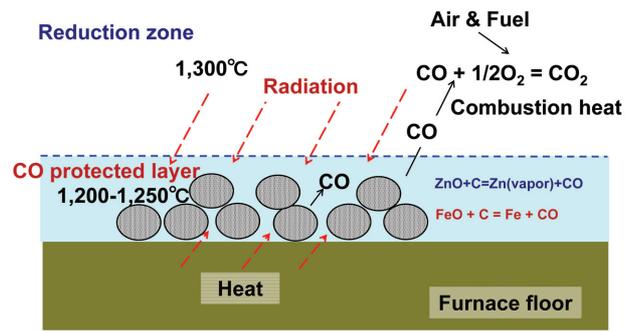


Fig. 11 Conceptual idea for pellet reduction

attracted attention. At the time when Nippon Steel started its development activities, there were two RHF's used for dust recycling in the world, but both were closed after about one year of operation due to technical problems.

At that time, the operation of the RHF's was still unstable, because the technology for granulating oxide wastes of widely different chemistries and properties and converting them into feedstock for blast furnaces, melting furnaces, etc. was still immature.

Consequently, Nippon Steel began to study the issue in 1996, and in order to examine the technical matters that could be clarified only on a large scale, constructed two commercial-scale RHF plants in 2000 at the Kimitsu and Hirohata Works, each with the capacity to process approximately 200,000 t of oxide wastes per year. Through trial operations and verification tests at those plants, the equipment and operation technologies for the recycling of oxide wastes were finally established in 2002¹⁴⁾.

The basic understandings gleaned from the R&D efforts targeting the recycling of oxide wastes using the RHF was that the system was an integrated process comprised of several steps, including raw material pretreatment, granulating, heating, reducing, exhaust gas treatment and recovery of zinc from the flue dust, and that it was essential to design the conditions for all of these process steps from the raw material preparation to the reduction in the furnace in an integrated manner. Based on this understanding, the following needs were identified: (1) a method for the pretreatment and granulation of low-quality material with fluctuating chemistries and properties into pellets suitable for processing in the RHF; (2) a method for reducing the oxide waste pellets into DRI suitable as a feedstock for steel production; and (3) stable operation technology for the equipment, which is essential for a commercial plant facility. The development progressed as follows.

(1) Pretreatment and granulation

A method for the mixing of dust and scale with fluctuating grain sizes, moisture content and chemistries was developed, and the conditions for granulating them were revealed. Operational technologies were established for pelletizing, briquetting and extrusion forming processes. In addition, for the pre-treatment of high-moisture sludge with fine particles, a combined continuous dewatering and extrusion forming process was developed, and was the first process of this kind to allow direct charging of wet agglomerates into the RHF without causing bursting through rapid heating.

(2) Reduction

The reduction and smelting furnaces that consume DRI are blast



Fig. 9 Outlook of rotary hearth furnace

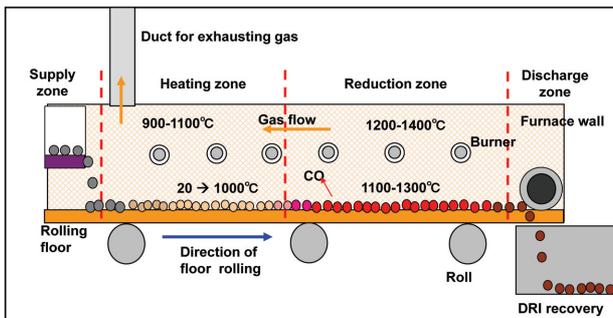


Fig. 10 Schematic view of several reactions accompanied with floor rolling direction in RHF process

furnaces, scrap melting furnaces, electric arc furnaces, etc., and the required DRI quality is different for each type of furnaces; high strength for blast furnaces, and a high reduction rate for scrap melting and electric arc furnaces.

(2-1) Production of high-strength DRI for blast furnaces

Use of DRI lowers the reducing agent ratio of blast furnaces, while improving the energy consumption and decreasing CO₂ emissions. On the other hand, blast furnaces require high strength feedstock. To clear this hurdle, a method for producing high-strength DRI was developed, in which sintered networks of metallic iron were formed in the DRI pellets. This approach allowed, for the first time, the direct charging of the produced DRI into blast furnaces.

(2-2) Production of high-reduction DRI for melting and electric arc furnaces

Melting and electric arc furnaces require a high reduction rate for DRI. In the laboratory, it was not difficult to obtain a high reduction rate, but in the large-scale plants it was a challenge, because CO₂ in the furnace atmosphere interfered with the reduction reactions. In this case, the optimum reducing conditions were studied on the Hirohata RHF. By increasing the furnace temperature through improvement of the refractory quality, enhancing the furnace temperature control and improving the reactivity of the pellets through modification of the material chemistry, it became possible for the first time for a commercial plant to produce DRI having metallic iron ratios of 85% or more.

(3) Stable operation

Long-term stable operation is essential for a commercial dust treatment plant, and various measures had to be developed to achieve this goal. Those actually put into practice included hearth condition control, measures against dust deposition in the exhaust gas system, and improvement of the furnace refractories. Three problems were particularly difficult to solve.

Due to reactions in the furnace, the oxide waste pellets adhered to the hearth refractory surface. Measures were taken to avoid the chemical composition range in which the melting point of the oxide waste pellets decreased during the reduction process. In order to reduce the generation of fines that were more likely to stick to the surface of the hearth refractory, the granulation method was also modified. Another important issue was prevention of adverse effects on the furnace operation due to impurities in the oxide waste pellets. The alkali content in the dust pellets evaporated and intruded into the furnace refractories, causing spalling. As a countermeasure, the composition of the refractory was modified to prevent alkali intrusion and allow higher operating temperatures. Volatile substances such as zinc, lead, alkalis, halogens, etc. evaporate in the RHF and are deposited in the exhaust gas system. To avoid this problem, the material chemistry was adjusted in order to raise the melting points of the deposited substances, and the exhaust gas system was modified to make dust removal easier.

The world's first dust recycling system with RHF's at an integrated steel mill was thus established on a commercial and permanent basis through systematic integration of these developed technologies.

For economically viable recycling of the oxide wastes arising from steelmaking processes using RHF's, material handling systems before and after the furnace are also important. In view of this need, a system to collect dust and scale and classify them by their grain size, iron content, carbon, zinc, alkali and moisture levels, etc., and to minimize costs based on the local conditions of the steel mill, was constructed.

At Kimitsu Works, which has an annual steel production capacity

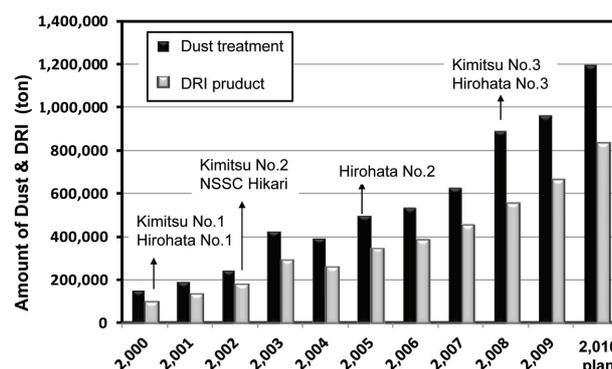


Fig. 12 Transition of the amount of dust treatment with RHF process in Nippon Steel group

Table 1 Effect of reduction merit for natural resource and energy saving obtained with RHF process

		Unit	Result of 2009fy	Final target
Reduction merit	Dust treatment	10,000t/y	95	120
	Iron ore	10,000t/y	66	81
	Coal	10,000t/y	7.4	9.0
	Zinc concentrate	10,000t/y	2.5	3.4
	Energy saving	Tera J/y	2,200	2,622
	CO ₂ reduction	10,000t/y	17	20

of 11 million tons, the DRI from the RHF is fed to blast furnaces, and accordingly, the technical developments focused on the production of high-strength DRI. Presently, three RHF's are in operation there, and are processing 600,000 tons per annum of oxide wastes containing zinc. Charging of the DRI into the blast furnaces, which translated into effective utilization of the energy for reducing the oxide wastes in the RHF's, successfully lowered the reducing agent ratio, realizing overall energy savings.

The recycling system of the Hirohata Works has three RHF's and recycles oxide wastes produced internally and from other mills. The collective recycling is possible because Hirohata produces steel from cold iron sources and does not operate blast furnaces; for this reason, there is no need to worry about zinc accumulation due to dust recycling. The system is capable of achieving high reducing ratios, and due to a rapid transfer system linking the RHF's to the melting furnaces, the DRI is charged while still hot, remarkably decreasing the overall energy consumption.

As stated above, Nippon Steel constructed six RHF's up until 2010 and completed a structure to recycle all dust and scale generated from its production activities, as shown in Fig. 12; presently, the annual recycling capacity is 1.2 million tons. As seen in Table 1, nearly 1 million tons of dust and scale were recycled in 2009, greatly contributing to energy and resource savings.

5. Closing

In addition to the above, Nippon Steel's technologies for the effective utilization of low-quality resources include chemical recycling of waste plastics and tires using coke ovens^{15,16)}, intensive granulation of fine iron ores using a polymer dispersant¹⁷⁾, etc. All of

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these technologies were developed through thorough examination of the fundamental characteristics of the materials in question. Ideas arising from such studies and efforts to overcome problems that enable mere ideas to become actual industrial practice are expected to continue to drive technical developments that will save natural resources and further increase the recycling of materials.

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Tetsuzo HAGA
General Manager, Dr.Eng.
Plant Engineering Div.
Plant Engineering and Facility Management Center
20-1, Shintomi, Futtsu, Chiba 293-8511



Tetsuharu IBARAKI
General Manager, Ph.D.
Ironmaking Technical Div.



Kenji KATOH
General Manager, Ph.D.
Plant Engineering Div.
Plant Engineering and Facility Management Center