

Recent Advances in Ultralow-Carbon Steel Refining Technology by Vacuum Degassing Processes

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Abstract:

The vacuum degassing technology introduced into Japan early in the 1960s has developed into a multifunctional process, including flux refining and temperature compensation. In recent years, technology development in this area has focused on the manufacture of ultralow-carbon steel. A mathematical model has been built to calculate the decarburization rate at each reaction site in the decarburization step of the RH process. Comparison of the values calculated by the model with actual RH vacuum degasser operating data has clarified the effects of the recirculation rate, evacuation rate, and effective reaction surface area on the decarburization rate. On the basis of these findings thus obtained, technologies have been developed to increase the evacuation rate by prevacuum system, increase the reaction surface area by increasing the gas injection rate, and prevent skull buildup by installing a top burner or multifunctional burner. Using these technologies, a new technology has been established for efficient mass production of ultralow-carbon steel with a carbon content of 10 ppm or less.

1. Introduction

DH and RH vacuum degassers were introduced into Japan at the beginning of the 1960s and have made remarkable progress into larger and more general-purpose refining furnaces in line with the increase of crude steel production and the growth of demand for higher-purity steel. **Table 1** summarizes the history of the DH and RH vacuum refining technologies at Nippon Steel. The DH and RH vacuum degassing processes have grown from 70 to 300 tons in vessel capacity and have functionally advanced

from only dehydrogenation into a comprehensive steel refining process combining decarburization, desulfurization, dehydrogenation, heating, and composition adjustment. This history is reviewed below.

1.1 1960s to early 1970s (establishment of vacuum refining technology)

Japan's first DH and RH vacuum degassers that started full-fledged operation in 1961 and 1963, respectively, were introduced for the primary purpose of dehydrogenating steel for large forgings. From the beginning, research was made into various refining phenomena characteristic of pressure reduction, such as decarburization and inclusion removal, and into fine compositional control by alloy additions through the vacuum vessel. To-

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Table 1 Development of vacuum refining technologies at Nippon Steel

1960 to early 1970s	Establishment of vacuum refining technology	<p>DH process Start of full-fledged operation of 70-ton DH (at Yawata in 1961) Adoption of MgO-Cr₂O₃ brick (at Yawata in 1965) Adoption of twin-vessel system (at Hikari in 1969) Scale up to 180 tons (at Yawata in 1969) Gas injection through metal tuyere (at Yawata in 1974)</p> <p>RH process Full-fledged operation of 100-ton RH (at Hirohata in 1963) Adoption of twin-vessel system (at Nagoya in 1968) Stainless steel refining by RH-OB (at Muroran in 1972)</p>
Late 1970s	Expansion of equipment scale	<p>DH process Construction of 350-ton DH (at Yawata in 1979)</p> <p>RH process Development of RH-OB immersion tuyere (at Muroran in 1975) Construction of 350-ton RH (at Oita in 1976) Development of RH light treatment (at Oita in 1977) Establishment of aluminum heating (at Nagoya in 1978) Two-stage argon injection through snorkel (at Hirohata in 1978)</p>
Early 1980s	Expansion of refining functions	<p>RH process Total RH treatment operation (at Oita in 1980) Desulfurization by flux addition into vessel (at Nagoya in 1980) Establishment of fully automatic treatment (at Oita in 1984) Increase in steel recirculation rate (at Nagoya in 1984) Adoption of oval snorkel (at Hirohata in 1984) Development of RH injection process (at Oita in 1985) Development of RH-PB process (at Nagoya in 1986)</p> <p>Other Development of V-KIP process (at Kimitsu in 1986)</p>

day's multifunctional refining equipment was founded in this way. In the initial stage of development, DH and RH vacuum degassers were short in refractory life and required frequent vessel maintenance, were low in vessel availability, and were applicable only to some high-grade steels. Adoption of magnesia-chromite brick and spread of the twin-vessel system improved the vessel availability and expanded the range of steel grades to which they could be applied. Through these technological developments, the RH process gradually secured its position as a predominant vacuum degassing process. The reason is that metallurgical control is easier to perform in the RH process with a continuous circulating flow of steel than in the DH process featuring a reciprocating flow of steel¹⁾.

1.2 Late 1970s (equipment scale expansion)

In the late 1970s, 300-ton converters came to be constructed and continuous casting of steel made rapid progress. The same period saw the appearance of vacuum degassers of capacities befitting the new converters. For the secondary refining with degassers, coordination with discreet between the converter operation and continuous casting steel temperature control became a major

issue. The RH-OB process continued to develop for stainless steel production, and established stable operating technology by 1972. A top-blowing oxygen lance was initially employed, but was later replaced by bottom blowing by a submerged oxygen injection system, which improved the decarburization oxygen efficiency and allowed the process to be used also in the manufacture of carbon steel. The aluminum heating method developed as an RH-OB applied technology in the carbon steel refining shortened the molten steel heating time and provided the whole steelmaking process with large thermal flexibility. On the other hand, the RH light treatment technology made it possible to treat the entire steel tonnage by the RH-OB process, and to substantially reduce the converter load and ferroalloy consumption. These innovations in the secondary refining process supported the enlargement of converters and increase of continuous casting ratio during this period.

1.3 1980s (expansion of refining functions)

In the 1980s, the demand for high-purity steel increased, and the vacuum degassing ratio of Japan's converter steel production jumped from 16% at the end of the 1970s to over 50% in 1984. The mass production of high-purity steel started in earnest. In 1980, the vessel flux addition method was established to enable the RH process to perform desulfurization and deoxidation. Then were developed the RH injection process and RH-PB (PB for powder blowing) processes that make use of highly reactive powder/metal interface reactions, and the V-KIP process which is a dedicated vacuum flux refining facility. Technologies were thus established for stably making ultralow-sulfur and low-oxygen steel.

The development history described above tells the important position vacuum refining now holds in the whole chain of steel-making processes. In recent years, demand for ultralow-carbon steel has steeply increased as typically seen in automotive steel and electrical sheet, and at Nippon Steel ultralow-carbon steel has come to account for about 30% of the total vacuum-degassed steel production (about 1 million tons per month as of 1991). Technology development in the past several years has focused on operating technology for enhanced decarburization in the vacuum degasser and related equipment technology. As a result, technology has been established to attain a ultralow carbon content of C ≤ 10 ppm within the decarburization treatment time of 15 min.

This article focuses on ultralow-carbon steelmaking technology in describing improvements made in the operation and equipment of steel vacuum degassing, and presents the results of overall verification made of the resultant degassing performance on the basis of decarburization model analysis.

2. Theoretical Analysis of Ultralow-Carbon Steel-making Technology

2.1 Construction of decarburization reaction model

When improving the operation of an actual process, it is of extreme importance to correctly understand factors determining the reaction rate at the plant concerned and to select the most effective means. Several reaction models have been proposed to explain the decarburization mechanism involved. For example, Sumida et al.²⁾ modeled the effect of steel recirculation flow rate on the reaction rate and discovered an index to improve the operation of the RH process. Harashima et al.³⁾ quantified from the results of basic test the effect of surface active elements on the

decarburization rate in the ultralow-carbon region on the basis of an adsorption model. Many of these models are composed of relatively simple equations and are easy to handle. In an actual process, however, the degree of vacuum and carbon concentration considerably change during treatment and rate-governing factors change their effects from time to time. It is thus extremely difficult to analyze the reaction behavior from the start of evacuation through the ultralow-carbon region by a simple model. A new dynamic model capable of predicting the reaction rate throughout the decarburization treatment of the RH process was built and used to study factors governing the reaction rate in the RH process.

Fig. 1 outlines the decarburization reaction model. Its characteristics may be summarized as follows.

- (1) The free surface exposed to the vacuum environment (bath surface decarburization), the surface of injected argon bubbles rising through the steel bath (argon bubble surface decarburization), and the evolution of CO gas from the bulk of the steel bath with supersaturation pressure (bulk steel decarburization) are considered as reaction sites.
- (2) The reactions of bath surface decarburization and argon bubble surface decarburization are described by a model that assumes that their rates are controlled by the combination of four processes, namely, liquid-phase mass transfer of carbon, liquid-phase mass transfer of oxygen, gas-phase mass transfer of CO, and interfacial chemical reaction, as expressed by Eqs. (1) to (4).

$$N_1 = ((A \cdot \rho / 100) / 12) \cdot k_c \cdot ([C]^b - [C]^i) \quad \dots\dots(1)$$

$$N_2 = ((A \cdot \rho / 100) / 16) \cdot k_o \cdot ([O]^b - [O]^i) \quad \dots\dots(2)$$

$$N_3 = -(A / (RT)) \cdot k_g \cdot (P_{co}^b - P_{co}^i) \quad \dots\dots(3)$$

$$N_4 = ((A \cdot \rho / 100) / 12) \cdot k_r \cdot ([C]^i \cdot [O]^i - P_{co}^i / E) \quad \dots\dots(4)$$

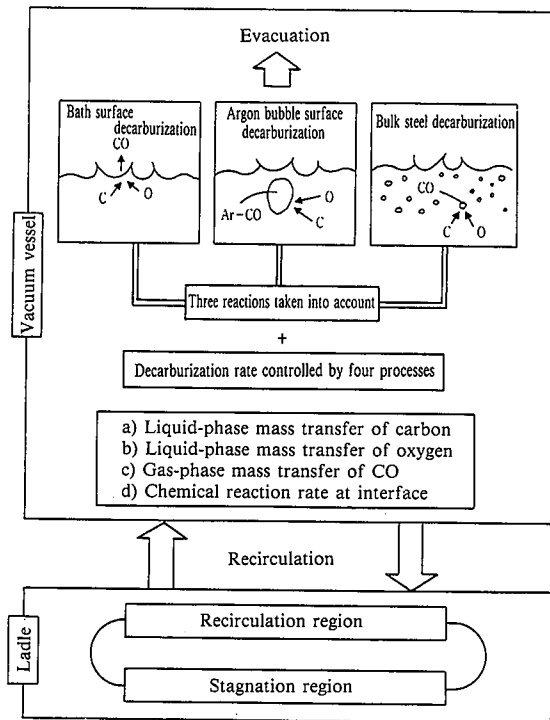


Fig. 1 Outline of decarburization reaction model

where N_x is the amount of reaction per unit time (mol/s); $[C]$ is carbon concentration (mass%); $[O]$ is oxygen concentration (mass%); k_c is liquid-phase mass transfer coefficient of carbon (m/s); k_o is liquid-phase mass transfer coefficient of oxygen (m/s); k_g is gas-phase mass transfer of CO (m/s); k_r is chemical reaction rate constant as calculated by Harashima's equation³⁾ (m/(%/s)); ρ is density of molten steel (kg/m³); E is equilibrium constant ($[C] + [O] \rightarrow CO$); A is reaction interface area (m²); P_{co} is partial pressure of CO (Pa); T is temperature (K); b is bulk; and i is interface.

- (3) Bulk steel decarburization is based on the model of Kuwabara et al.⁴⁾ If the ferrostatic pressure at a given bath depth is smaller than apparent P_{co} , the CO gas is assumed to instantaneously evolve to a given bulk steel decarburization end condition (supersaturation pressure). Bulk steel decarburization is considered to end when apparent P_{co} falls below 2,670 Pa (about 20 Torr) and the carbon concentration falls below 15 ppm⁵⁾.

- (4) A stagnation zone in the ladle is assumed in addition to a reaction zone in the vacuum vessel and a mixing zone in the ladle. Perfect mixing is assumed in each zone and exchange of the molten steel is considered to occur between the zones. The rate at which the molten steel is exchanged between the reaction zone and mixing zone is determined according to an existing recirculation rate equation⁴⁾ The volume of stagnation zone is put at 10% of the heat size, and the rate at which the molten steel is exchanged between the stagnation zone and mixing zone is specified to suit the measured mixing time.

- (5) The relationship between the degree of vacuum and evacuation capacity peculiar to specific vacuum degassers is programmed so that the degree of vacuum can be changed to meet the amount of gas evolution.

For argon bubble surface decarburization, k_c and k_o are specified according to penetration theory, and A is specified according to the equation of Sano et al.⁶⁾ to give the bubble diameter in a rising bubble swarm. The value of liquid rising velocity necessary to calculate the bubble residence time, for example, is calculated from the recirculation flow rate and the cross-sectional area of the upleg snorkel. For bath surface decarburization, k_c and k_o are put at a constant level of 0.015 m/s, a representative value calculated from penetration theory. To specify A , the new parameter Π that indicates the effective reaction surface area of free surfaces is defined by Eq. (5) from the liquid rising velocity u (m/s) calculated by the equation of Sawada et al.⁷⁾ and from the result of a basic experiment⁷⁾ saying that the bubble rising region (bubble active surface) has a larger effective reaction surface area than other free surfaces. Assuming that injected bubbles spread to an angle of 12°⁸⁾ on each side, the bubble active surface area is calculated on a geometrical cross section.

$$\Pi = u \times [(\pi d^2 / 4) + 6.5 \times S] \times (P_0 / P)^{2/3} \quad \dots\dots(5)$$

where π is circle ratio; d is inside diameter of the vacuum vessel (m²); S is bubble active surface area (m²); P_0 is standard pressure (= 133 Pa); and P is pressure in the vacuum vessel (Pa). A regression equation is derived between the value of Π and the decarburization curve fit to various RH operating conditions, and A is defined by Eq. (6).

$$A = 0.65 \times \Pi \quad \dots\dots(6)$$

2.2 Clarification of factors governing decarburization reaction

Fig. 2 shows the results of applying the model to an actual RH vacuum degasser. The calculated values agree well with the measured values. The decarburization ratio of each reaction site at each point of time is included. As indicated, bulk steel decarburization predominates in the initial stage of decarburization, while bath surface decarburization occurs principally in the final stage of decarburization. The model can be used to analyze the decarburization behavior of the RH vacuum degasser where various factors are involved. Given this capability, the model was applied to two RH vacuum degassers (A-RH and B-RH) operated under the conditions given in Table 2 and was used to study the factors concerned.

The decarburization rate constant K_1 for the time t_1 (or period I) (min) during which the carbon concentration falls from 150

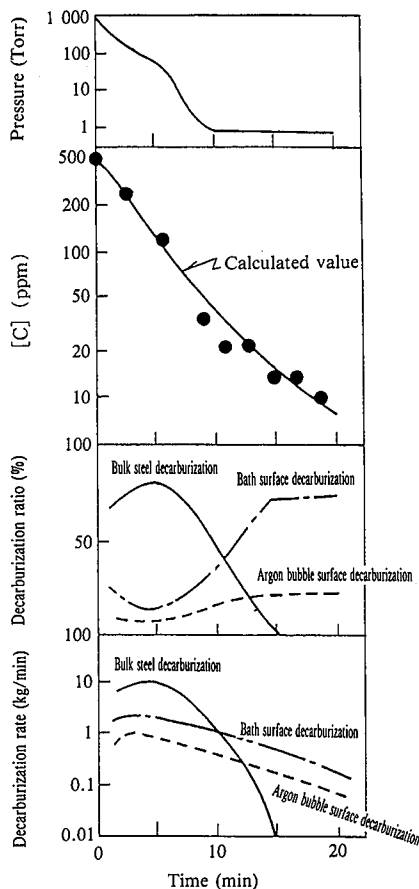


Fig. 2 Comparison of values calculated by decarburization reaction model and measured values (A-RH, lift gas: 150 Nm³/h, stirring gas: 600 Nm³/h)

Table 2 Equipment and operating conditions of RH vacuum degassers analyzed

	A-RH	B-RH
Heat size (tons)	260	340
Snorkel inside diameter (mm)	730	600-750
Lift gas flow rate (Nm ³ /h)	150	150-240
Stirring gas flow rate (Nm ³ /h)	0-1000	0-390
Evacuation capacity at 300 Torr (kg/t·h)	90	35
Initial oxygen content (ppm)	500-650	250-400

to 50 ppm and the decarburization rate constant K_2 for the time t_2 (or period II) (min) during which the carbon concentration falls from 30 to 15 ppm are given by

$$K_1 = \ln(150/50)/t_1 \quad \dots\dots(7)$$

$$K_2 = \ln(30/15)/t_2 \quad \dots\dots(8)$$

Fig. 3 shows the relationship between the decarburization rate constant K_1 (min⁻¹) during period I calculated by the model and the recirculation ratio (recirculation tonnage W_e /heat tonnage W). The decarburization rate improves with increasing recirculation ratio. When compared at the same recirculation ratio, the A-RH vacuum degasser with a higher evacuation rate provides higher values than the B-RH vacuum degasser. If the evacuation rate of the B-RH degasser is improved, its treatment rate during period I can be brought closer to that of the A-RH degasser. The reaction involved can be accelerated by bringing the recirculation ratio of the A-RH degasser close to the maximum recirculation ratio of the B-RH degasser. Fig. 4 shows the effect of the effective reaction surface area of bath surface decarburization per unit steel weight A_s/W (m²/t) on the decarburization rate during period II as calculated by Eqs. (5) and (6). The calculated values can explain well the actual data of the two RH vacuum

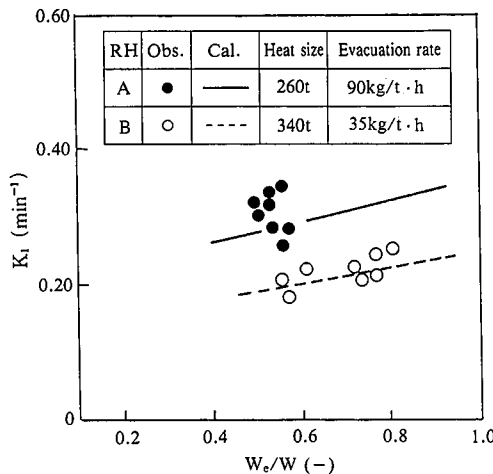


Fig. 3 Effects of recirculation rate and evacuation rate on decarburization rate during period I

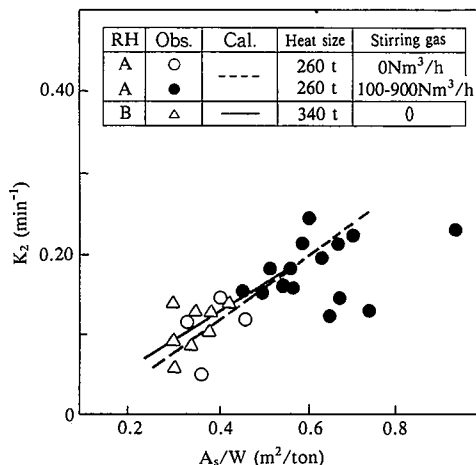


Fig. 4 Effect of effective reaction surface area on decarburization rate during period II

degassers. The closed circles indicate the data obtained when a large amount of gas was injected into the vacuum vessel of the A-RH vacuum degasser. It is obvious that the gas injection increased the effective reaction surface area and thus substantially improved the decarburization rate during period II. Under intense stirring with $A_s/W > 0.6$ some measured values are smaller than the corresponding calculated values. This is probably because the violent splash caused by the intense stirring action melted the high-carbon vessel skull and contaminated the steel bath with the resultant carbon. Therefore the following requirements must be met to increase the decarburization rate in the RH process:

Decarburization period I

- (1) Increase the evacuation rate and create a vacuum high enough to cause bulk steel decarburization.
- (2) Increase the recirculation rate and bring the carbon concentration in the vacuum vessel close to that in the ladle.

Decarburization period II

- (1) Impart violent surface turbulence and increase the effective reaction surface area of bath surface decarburization.
- (2) Prevent carbon contamination from the skull and alloy additions.

3. Improvements Made in RH Process Equipment and Operation to Promote Decarburization Reaction; Result

Nippon Steel has made various modifications to the operation and equipment of the RH process to promote the decarburization reaction according to past findings and the above-mentioned results of analysis. Major technologies developed in this line are described below.

3.1 High-speed evacuation

To improve the decarburization rate during period I, the recirculation rate and evacuation rate must be increased as noted above. To increase the recirculation rate in No. 2 RH vacuum degasser at Nagoya Works, the snorkel diameter and lift gas flow rate were increased, resulting in a marked rise of decarburization rate in 1984.⁹⁾ In 1992, high-speed evacuation technology using a prevacuum system was established to further improve the decarburization reaction.

The prevacuum system is schematically illustrated in Fig. 5. Before the vacuum treatment is started, the exhaust system standby valve is closed, and the exhaust system and alloy addition hopper (310 m³) downstream the standby valve are kept at a reduced

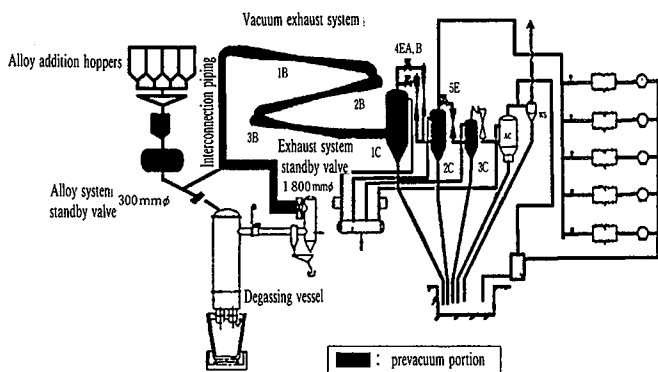


Fig. 5 Schematic illustration of prevacuum system

pressure of about 100 Torr by the vacuum pumps. As soon as the treatment is started, the standby valve is opened, and the degassing vessel is rapidly evacuated. Using the prevacuum system, the vacuum of about 400 Torr at which the molten steel recirculation is started can be reached in about 4 s, cutting down about 30s from the normally required time. Fig. 6 shows the relationship between the decarburization rate constant and the carbon concentration as measured at two sampling points. In the case of conventional treatment, decarburization is arrested by the evacuation rate in the initial high-carbon region. This arrest of decarburization in the high-carbon region can be completely eliminated by introducing the prevacuum system technology.

3.2 Technology to increase free reaction surface area

To promote the decarburization reaction during period II, the effective reaction surface area must be increased by injecting an inert gas into the vacuum vessel. If the inert gas is injected at a high rate through two or more points, the effective reaction surface area can be made larger than when the lift gas is injected through one snorkel as normally done.

The RH-OB vacuum degasser is equipped with oxygen blowing tuyeres through oxygen is blown into the degasser vessel to speed up decarburization in the high-carbon region while the steel is heated by aluminum oxidation. In the low-carbon region of $C < 100$ ppm, the mass transfer rate of oxygen normally is high enough and has nothing to do with the decarburization reaction. Oxygen therefore need not be blown through the OB tuyeres. Instead, argon can be blown through the OB tuyeres at a high rate. Fig. 7 shows the effect of the total argon flow rate on the decarburization rate constant during period II. When the total argon flow rate is increased from 500 to 1,000 Nm³/h, the decarburization rate constant is almost doubled. This result verifies the effectiveness of this technology in substantially improving the decarburization rate during period II.

3.3 Prevention of skull buildup

Skull buildup on the inside wall of the degasser vessel causes carbon contamination during treatment and incurs the additional task of deskulling. It is therefore important to guard against the skull buildup at all times. A fuel burner can melt down and remove the skull with relative ease. Here are described a top burner that can melt down and remove the skull between vacuum treat-

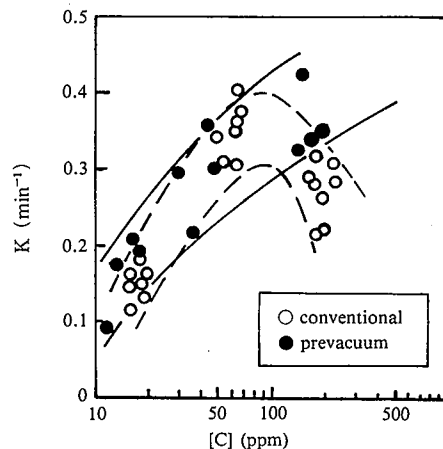


Fig. 6 Relationship between steel carbon concentration and decarburization rate

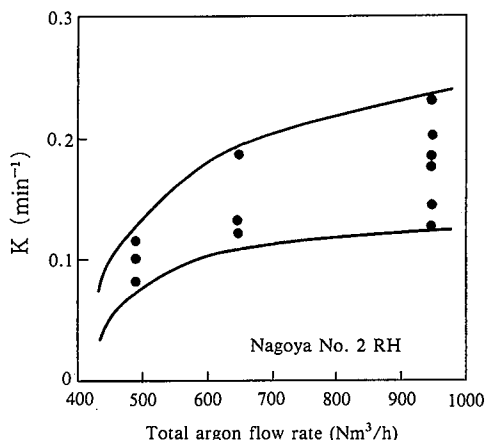


Fig. 7 Effect of total argon flow rate on decarburization rate

ments, and a multifunctional burner that can melt down and remove the skull irrespective of whether or not the vacuum vessel is treating the molten steel.

3.3.1 Top burner

Table 3 presents the main specifications of the top burner equipment of No. 2 RH vacuum degasser at Nagoya Works^{10,11}. The top burner is inserted through a port in the dome of the degassing vessel down to 5 m below the dome. It melts down the skull and preheat and dry the inside wall of the vacuum vessel down to the snorkels. Using the top burner, the RH degasser vessel can be preheated and deskulled as required so that it can operate at the maximum possible rate and prevent carbon contamination from the skull during the vacuum treatment of ultralow-carbon steel.

Fig. 8 shows the decarburization rate without skull buildup in comparison with that with skull buildup. When the vacuum degasser is operated without skull buildup, the decarburization rate is high throughout the treatment. Particularly, the drop of decarburization rate in the low-carbon region is greatly eased. The carbon pickup, presumably from the skull, averaged 2 ppm

Table 3 Main specifications of top burner equipment

Preheating method	Top-inserted oxygen combustion COG burner
Combustion capacity	6.0×10^6 kcal/h
Fuel and flow rate	COG, 1,340 Nm ³ /h

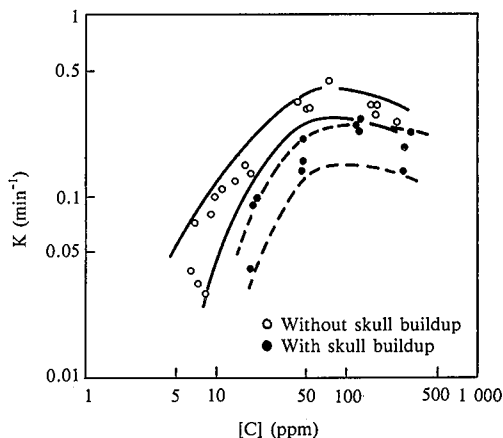


Fig. 8 Effect of vessel wall skull buildup on decarburization rate

during the alloy addition after the end of decarburization and posed a serious problem. The skull-free operation can solve this problem and prevent the carbon contamination of the molten steel during and after the decarburization treatment.

3.3.2 Multifunctional burner

Hirohata Works makes sheet steel and electrical steel, two grades with completely different chemical compositions, in the same RH vacuum degasser. It was thus imperative to develop technology of molten steel temperature compensation without skull buildup.

A burner with the heating function to compensate for the molten steel temperature and prevent skull buildup, and with the oxygen top blowing function to promote decarburization and heat the steel with aluminum was devised. It is made up of a single lance. When plant tested, the multifunctional burner performed satisfactorily.

Table 4 shows the service conditions and functions of the multifunctional burner. The plant test confirmed that the multifunctional burner can stably operate both in atmosphere and under vacuum if three conditions, namely, ignition temperature of over 800°C, presence of fuel and presence of oxygen, are met.

Fig. 9 shows the results of combustion test made of the multifunctional burner under the atmospheric pressure while the degasser was out of operation. The refractory heat transfer rate with present electrode heating is 400,000 kcal/h and the refractory heat transfer efficiency with LNG heating is 57 to 71%. With the multifunctional burner, a steel heating rate equivalent to that in electrode can be obtained by burning inexpensive LNG at the flow rate of 70 Nm³/h. At the maximum LNG flow rate, a heating rate four times as high as that obtainable in electrode heating can be realized. Fig. 10 gives the combustion test results of the multifunctional burner during vacuum treatment. The efficiency of heat transfer to the molten steel is about 52%, the molten steel is reheated at a rate of 0.5 to 1.5°C/min, and the

Table 4 Purposes of multifunctional burner

	Oxygen + LNG	Oxygen blowing
Under vacuum (during treatment)	Steel heating Vessel refractory heating	Promotion of decarburization Aluminum heating
In atmosphere (off treatment)	Vessel refractory heating	

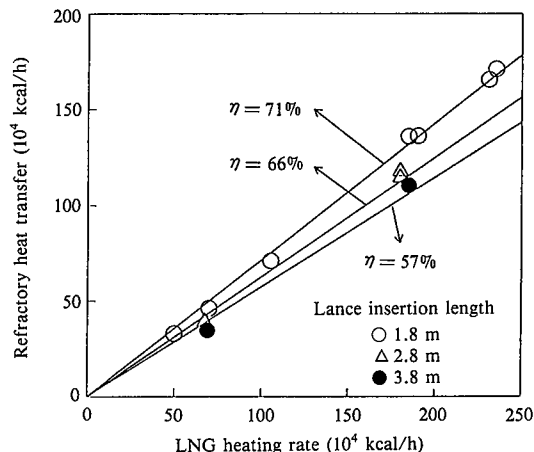


Fig. 9 Effect of LNG heating rate on refractory heat transfer rate (off treatment)

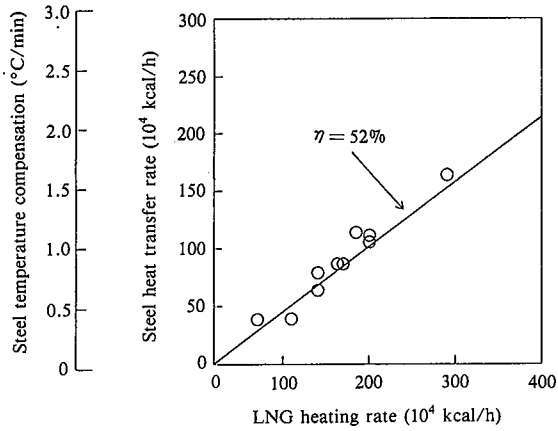


Fig. 10 Steel temperature compensation by LNG burner heating (during treatment)

combustion gas has such a small impact on the vacuum level that there is no need to augment the vacuum system.

4. Mass Production Test Results of Ultralow-Carbon Steel

On the basis of the above discussion, modifications were made to the equipment and operation of Nagoya No. 2 RH vacuum degasser, as shown in Table 5, and nine heats of ultralow-carbon steel were test processed in the RH vacuum degasser. The major improvements are an increase in snorkel diameter, improvement in evacuation rate by use of the prevacuum system, increase in lift gas flow rate, injection of stirring gas through the OB tuyeres, and complete preliminary deskulling by the top burner to preclude the adverse effects of skull contamination.

Fig. 11 shows the change of carbon concentration with time. The average carbon concentration was reduced to 10 and 5 ppm at 12 and 20 min into the treatment cycle, respectively. The decarburization rate and the ultimate carbon concentration were substantially improved as compared with the results of the conventional process. Fig. 12 shows the carbon content of the nine heats as determined in the tundish after deoxidation and alloy addition. The carbon pickup after decarburization was very small in magnitude and variability. It was thus made possible to sequence cast ultralow-carbon steel with a product carbon content of ≤ 10 ppm.

5. Conclusions

When the factors governing the decarburization reaction in the RH process were clarified by a decarburization reaction model and operational improvements were made accordingly, it was found feasible to mass produce ultralow-carbon steel with a carbon content of 10 ppm or less by the RH process. It is predicted that the proportion of ultralow-carbon steel will increase and that steels will be more finely graded. From a point of view of production technology, emphasis will shift to ensuring mass production

Table 5 Ultralow-carbon steel vacuum processing test conditions

Snorkel inside diameter	730 mm
Evacuation capacity	2,000 kg/h (at 1 Torr) Prevacuum system used
Lift gas flow rate	180 Nm ³ /h
Stirring gas flow rate	340 Nm ³ /h
Deskulling	Top burner used

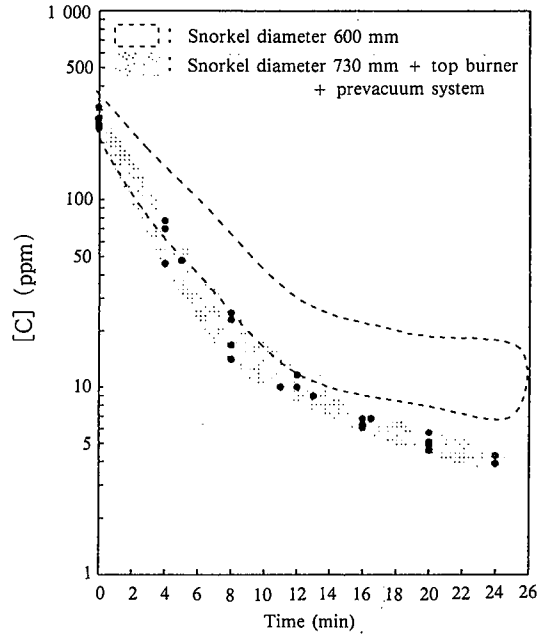


Fig. 11 Change with time in carbon concentration

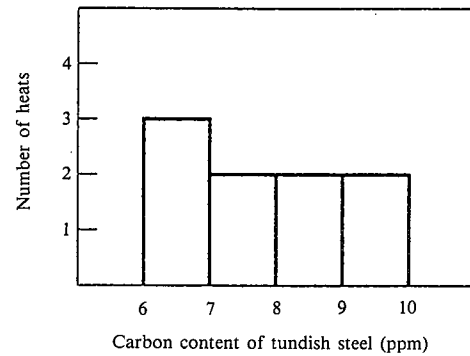


Fig. 12 Results of ultralow-carbon steel mass vacuum processing test

capacity by shortening the total operation time, including maintenance, and providing multiple secondary refining functions for treating many steel grades with one facility. For example, the top burner and multifunctional burner described in the foregoing part are expected to find increasing usage in these two aspects. Given the present trend of the steelmaking process toward expansion, the steelmaking functions should be diversified to cover the whole process from the converter down to the continuous caster including the RH vacuum degasser that undertakes secondary refining. The last named, in particular, should keep progressing always with a view to optimization for the total steelmaking process.

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