Present Status of Research and Development of Nuclear Steelmaking in Japan*

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I. Introduction

The steel industry consumes an enormous amount of energy. The Japanese steel industry, being no exception, accounts for about 18% of the energy consumed in Japan as a whole. Further, about 70% of the primary energy it consumes is obtained from coal, of which about 90% is imported. Statistics show that in 1977, Japan imported about 56 million tons of coking coal at a cost of approximately \$3.3 billion.

As is generally known, the Japanese steel industry has always had the problem of procuring such huge quantities of coking coal at a stable rate. Other serious problems are price negotiations with overseas coal suppliers, transportation from collieries to Japan, and storage of coal at home.

In addition, management of the steel industry is under pressure from environmental problems caused by the consumption of such enormous quantities of coking coal. Therefore, an important subject for the Japanese steel industry is to push forward technological development in order to save coking coal or to develop steelmaking techniques which will eliminate the need of coking coal. It is towards this end and to contributing to diversification of national energy resources, that introduction of atomic energy into steelmaking processes is attempted on a grand scale.

The high-temperature gas-cooled reactor (HTGR), currently under development, is capable of supplying a large quantity of heat at high temperatures unobtainable by conventional means utilizing carbon dioxide, light water, heavy water, and fast breeder reactors. Once the utilization of the HTGR to provide process heat besides that for power generation is established, it is conceivable that this heat source will exert a great influence upon the present energy structure.

High-temperature nuclear heat can be utilized advantageously in processes to convert coal, petroleum, or natural gas into hydrogen, carbon monoxide, etc. Therefore, it is possible for the steel industry to use a large quantity of these converted gases for direct reduction or injection into blast furnaces.

In 1963, Schenck *et al.* of Aachen Technical College proposed the application of HTGR heat to the process of steelmaking. This proposal was the turning point from which the study of nuclear steelmaking began in many countries. In the initial stages of the study, research was carried out by British Steel Corporation, the Jülich Nuclear Research Center, and EC.

In Japan, Committee on Utilization of Nuclear Energy, the Iron and Steel Institute of Japan, was established in September, 1968. Based on the results of the investigations of this committee, Research and Development of Nuclear Steelmaking was inaugurated in July, 1973, as a national research and development program by the Agency of Industrial Science and Technology, Ministry of International Trade and Industry (MITI).

Elsewhere, the Nuclear Steelmaking Task Force was established by American Iron and Steel Institute (AISI) in 1973. In the same year, the European Nuclear Steelmaking Club (ENSEC) was established. In 1974, Arbeitsgemeinschaft Nukleare Prozesswärme (ANP) was established followed by Prototypanlage Nukleare Prozesswärme (PNP) in the Federal Republic of Germany (F.R.G.).

II. Nuclear Steelmaking

Atomic energy has been utilized mainly for power generation to date. However, the emergence of the HTGR in which nuclear heat is removed by helium gas coolant has opened up the possibility of utilizing the 700° to 1 000°C temperature heat as the process heat. In this temperature range, reducing gas, namely, hydrogen, or a mixture of hydrogen and carbon monoxide is efficiently produced from coal, petroleum or natural gas. In addition, ample heat in the 700° to 1 000°C temperature range, which is necessary for direct reduction of iron ore also, is obtainable quite readily from an HTGR. Thus the nuclear heat available from the HTGR can be utilized both in the production of reducing gas and in the reduction of iron ore.

Conventional techniques to convert petroleum or natural gas into reducing gas have already been established. Likewise, techniques for the direct reduction of iron ore have been established such that commercial scale shaft furnaces producing 1 000 to 1 200 t/day of reduced iron are already in operation in many parts of the world. However, it is necessary to carry out research and development in nuclear steelmaking to connect these shaft furnaces to the reactors. The extensive engineering technology and expertise resulting from these studies will lead to a safe, compatible and economical nuclear steelmaking system.

The tremendous amount of development cost accompanying this research and development makes it difficult for a private enterprise or a single industry

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to launch such a project without the support of a more influential organization. It is regarded as being timely, therefore, that nuclear steelmaking was taken up in Japan by the Agency of Industrial Science and Technology of MITI, as the subject of a national research and development project.

The pattern of nuclear steelmaking processes varies with the selection of the material and process used for producing reducing gas, the selection of the type of reduction furnace, and whether intermediate heat exchangers are used or not. An additional but important factor to be determined is whether the site of the plant for producing reducing gas should be located remotely from the site of the steelmaking plant or the former should be an integral part of the latter.

Figure 1 shows concisely the nuclear steelmaking system which is being developed in Japan. In this process, the nuclear heat is transferred to the secondary helium through the medium of an intermediate heat exchanger. The primary helium, which comes out of the high-temperature gas reactor at 1 000°C and 40 atm, returns to the reactor at 400°C, while the secondary helium comes out of the intermediate heat exchanger at 925°C and 45 atm and returns at 300°C. The reducing gas heater, the steam reformer, and the steam heater are installed in parallel on the high-temperature side of the secondary helium loop, and the steam generator is located on the low-temperature side. Since Japan lacks fossil fuel, vacuum residual oil derived as a by-product of oil refining is selected

as the raw material to produce reducing gas. Therefore, the major portion of the reducing gas, hydrogen and carbon monoxide, is produced by steam reformation of naphtha and gasification of pitch that is obtained by the cracking of vacuum residual oil by steam. The reducing gas is heated to 800°~850°C in the reducing gas heater and then directed into the shaft furnace. As shown in Fig. 1, the top gas of the shaft furnace is recycled in a closed system after dust elimination and other treatments.

Figure 1 also shows an integrated system in which both the reactor and the steelmaking plant are located on the same site. Whereas, Fig. 2 is a schematic diagram of a non-integrated system drawn by

Fig. 2. A nuclear reactor gas-making and electricity generating complex serving various steelworks and other industries (ENSEC)

Fig. 1. Nuclear steelmaking process diagram (Japan)

ENSEC¹⁾ where the reducing gas is piped out at a low temperature. This gas is supplied not only to the steelmaking plant but also to chemical and other plants after being heated with fossil fuels. An intermediate heat exchanger is not used in this system.

AISI is considering that the use of natural gas and fuel coal as the material for producing reducing gas and has formulated a plan for a metallurgical-chemical complex based on a Very High Temperature Gas Cooled Reactor (VHTR) system as shown in Fig. 3.²⁾ Because of absence of intermediate heat exchanger, this plan is similar to that presented in Fig. 2. It is proposed that the natural gas be replaced by SNG in the reforming unit.

The PNP project is furthering the research to gasify bituminous and brown coals by nuclear heat. The PNP position is that this research is not solely for the purpose of nuclear steelmaking but is intended for both industrial and household uses. Accordingly, the steelmaking industry would be merely one of the consumers of the gasified bituminous or brown coal. Figure 4³⁾ shows the PNP process of gasifying bituminous coal. Heat transferred *via* an intermediate heat exchanger to secondary helium is used for the gasifying reaction.

The process for the hydro-gasification of brown coal is presented in Fig. 5.⁴⁾ In this process, helium gas at 950°C from the HTGR is directed to a steam reformer. The hydrogen gas obtained through the steam reformer and shift converter is fed into the brown coal gasifying unit. Part of the methane obtained in treating crude gas is sent into the steam reformer.

The processes to utilize nuclear energy as process heat elaborated above show that each reflects the particular conditions of the respective countries.

III. High-temperature Gas-cooled Reactor (HTGR)

An HTGR can supply a large quantity of high-

temperature heat suitable for steel and chemical industries which are intensive energy consumers. The nuclear fuel used in these reactors is a coated-particle type consisting of a number of particles, approximately 1 mm diameter, dispersed in a graphite matrix and molded into nuclear fuel elements. Using helium gas as the coolant, the HTGR is considered to be a clean reactor due to the low emission of fission products.

The experimental reactor, AVR (13 MW_{el}), installed at the Jülich Nuclear Research Center, has successfully produced heat at 950°C.

In Japan, the Japan Atomic Energy Research Institute is promoting research and development of a multi-purpose HTGR with an output helium temperature of 1 000°C. The plans call for the construction of an experimental reactor with a thermal output of 50 MW. Figure 6⁵⁾ is a schematic diagram of this experimental reactor.

A 330 MW HTGR has been in operation at Fort St. Vrain, U.S.A. and a THTR with an almost equal capacity is to be completed in 1981 in Schmehausen, F.R.G. The helium temperature of both these reactors is about 750°C. However, these installations are intended for electric power generation and not

Fig. 4. Gasification of bituminous coal with steam using heat from HTRs (PNP)

Fig. 3. A VHTR-based metallurgical-chemical complex (AISI)

Fig. 6. Design concept of the multi-purpose high temperature gas-cooled experimental reactor

- ① Control rod driving device
 - ② Stand pipe
 - ③ Biological shield
 - ④ Orifice device
 - ⑤ Fuel element
 - ⑥ Reflector element
 - ⑦ Pressure vessel
 - ⑧ Core barrel
 - ⑨ Back-up cooling system
 - ⑩ Plenum element
 - ⑪ Diagrid
 - ⑫ Coolant outlet pipe
- changer
- 6) Development of a helium circulator
 - 7) Development of a 3 MW helium heater
 - 8) Development of a thermal insulation system in the very high temperature region
 - 9) Research on the hydrogen permeation phenomenon
 - 10) Development of an emergency shut-off valve and a bypass valve
 - 11) Development of a helium purification system to remove hydrogen and other impurities
 - 12) Research on a system for the continuous measurement of hydrogen and the analysis of impurities in helium

Photo. 1. High-temperature helium test loop

Table 2. Design data of high temperature heat exchanger

Shell	Outside diameter	1 560 mm
	Height	13 000 mm
Heat Transfer Tube	Size	25.4 mm O. D.×4.0 mm thick
	Number	30
Temperature Entering	Shell side	1 000°C
	Tube side	327°C
Temperature Leaving	Shell side	379°C
	Tube side	925°C

- 13) Sliding wear and fretting wear tests of metallic materials.

These efforts have culminated in a 1.5 MW high-temperature helium test loop shown in Photo. 1. The loop comprises an helium purification unit (foreground), a pair of helium circulators, one for the primary line and the other for the secondary (middle ground), a 1.5 MW heat exchanger, a 3 MWt helium heater, a steam heater, and a steam generator (background), and completes with control-cum-data processing system and auxiliary units, some of which are installed outside this housing. Table 2 shows the elementary data on the high-temperature heat exchanger. Operation of this test loop has commenced October, 1978.

2. Heat Resistant Superalloy

Primary importance is attached to the creep rupture strength which is to be no lower than 1 kg/mm² at 50 000 hr (ultimately 100 000 hr) in a helium atmosphere at 1 000°C. New superalloys for manufacturing pipes with 25 mm O.D., 15 mm I.D. and over 7 m in length are being developed.

Table 3⁶⁾ shows an estimation of the creep rupture strength of conventional wrought alloys in air at 1 000°C. It is impossible to find any alloys having the necessary strength mentioned above. In addition, it has been observed in some cases that the creep rupture strength of an alloy in helium atmosphere is lower than that in air. Therefore, it has been

Table 3. Estimated creep rupture strength of superalloys (by Larson-Miller method)

Alloy	Creep rupture strength (kg/mm ²)	
	10 000 hr	100 000 hr
Incoloy 800	0.72	0.45
Incoloy 807	0.55	—
Inconel 617	1.05	0.60
Inconel 625	0.82	0.52
Hastelloy X	0.82	0.48

judged that to manufacture heat transfer pipes using existing alloys is difficult, to say the least.

Thereupon, since 1973, eleven new alloys have been proposed, of which several have already been omitted from further development because the required creep rupture strength could not be obtained in trial manufacturing and various tests. Among the proposed alloys, details have been made public on the following types:

- 1) 0.05C-16Cr-20W-30Co-Ni⁽⁷⁾
- 2) 0.03C-23Cr-18W-Ni⁽⁸⁾
- 3) 0.06C-18Cr-15W-Ni⁽⁹⁾
- 4) 0.03C-15Cr-25W-Ni⁽¹⁰⁾
- 5) 0.03C-27.5Cr-5W-5Mo-Ni⁽¹¹⁾
- 6) 0.06C-20Cr-20W-Ni⁽¹²⁾

Hydrogen permeation tests as well as creep rupture and corrosion tests in various atmospheres are being performed at the National Research Institute for Metals.

3. High-temperature Heat Insulating Materials

The characteristics of heat insulating materials in atmospheres of high-temperature helium, steam or reducing gas are different from those in air. It is necessary, therefore, to develop heat insulating materials which can maintain stable characteristics for a long time in these atmospheres, particularly in a high-temperature helium atmosphere. In this project, blocks made of fused silica refractories and Al₂O₃-SiO₂ in fibrous shape have been selected as the materials for development. Research on manufacturing methods as well as characteristics tests of the materials manufactured in trial are in progress. The aim of the research and development is to improve the insulating performance of these materials in helium gas at over 1 000°C and at 40 atm and in a reducing gas or steam atmosphere at approximately 900°C. Besides the obvious requirement that no substance detrimental to the operation must be discharged, any change in properties occurring through a 100 000 hr service must be:

- 1) Increase in thermal conductivity: less than 10%
- 2) Increase in gas permeability: less than 10%
- 3) Shrinkage: less than 0.1%
- 4) Resiliency from 50% compression (for fibrous material): greater than 20%.

Fused silica refractories are being studied with the accent on restraining devitrification and minimizing shrinkage. Restraining of devitrification will present

Fig. 7. Resiliency of newly developed insulators as heated for 20 000 hr in helium

Fig. 8. Resiliency under a thermal gradient

no problem since some materials show no evidence of devitrification after heat treating for more than 27 000 hr at 1 000°C in a helium atmosphere. Also, holding shrinkage to within the specified limits seems possible through the selection of particle size, baking temperature and time. It appears also possible to achieve the required thermal conductivity and permeability at 1 000°C in helium atmosphere.

The Al₂O₃-SiO₂ fibrous insulator are being studied with the emphasis on improving resiliency. Figure 7 shows the result of a resiliency test conducted for 20 000 hr keeping the temperature uniform through the sample. Apparently, it is difficult to obtain the target value of 20% resiliency at temperatures over 900°C, but, since a temperature gradient always exists through the heat insulating material, the uniform temperature tests are too severe. Actually, from the results presented in Fig. 8, it seems possible to achieve the target value given a temperature gradient of 1 000°C at the hot surface and 500°C at the cold surface.

In addition, several heat insulating structures combining the fused silica refractories and the Al₂O₃-

SiO₂ fibrous materials are being examined together with their fabrication methods. Photograph 2 shows the test apparatus of the structure to determine the heat insulating characteristics in helium atmosphere at 1 000°C and 40 atm.

4. Reducing Gas Production Unit

As mentioned earlier, Japan, a country having little fossil fuel resources, has a very limited option to select raw materials for the reducing gas production. The selection of vacuum residual oil, a by-product of oil refining, is significant because it is a mere remainder of the costly imported oil.

For shaft furnaces, the desired characteristics of the reducing gas are $H_2/(H_2+CO)>0.5$, $(H_2O+CO_2)/(H_2+CO)<0.1$, $CH_4<3\%$ and a temperature of between 800° and 850°C. In addition, since these shaft furnaces can utilize only 30% of the resident reducing gas, re-cycling of the top gas is a necessity.

Steam raised to about 800°C with nuclear heat is used to crack the vacuum residual oil. The reducing gas is produced through the steam reforming process of light hydrocarbons with nuclear heat and through gasifying pitch with high-temperature steam and oxygen. A low-sulfur fuel oil is produced by converting heavy oil into hydrogen followed by a desulfurizing step. Since the steam cracking process of vacuum residual oil and hydrodesulphurization process of heavy oil are already established and have been in operation commercially, it was determined in this project to develop a heat exchange type steam reformer and a pitch gasifying unit. A concentrated effort is being made to develop a steam reformer which produces reducing gas at 850°C by utilizing helium gas as the heating fluid, reforming light hydrocarbons with a suitable catalyst. In 1974, the design stage began on a selected model of the steam reformer. The basic tests necessary for this design, such as material and prototype tests, were carried out. Further, a testing unit was installed to obtain information concerning heat transfer characteristics and pressure losses. Also, basic tests concerning the chemical reactions are being conducted. Based on the results of these tests, a test plant of a steam reformer treating 100 kg/hr of naphtha was built. Photograph 3 illustrates the test plant, which is in operation since June, 1978, and Table 4 shows the elementary data of the plant.

Photo. 2. Thermal barrier testing equipment

The unit for gasifying pitch converts super-heavy hydrocarbons containing extremely small percentage of hydrogen into coke. The reducing gas is produced by gasifying this coke with the water-gas reaction. On the basis of the tests conducted with a micro-reactor of 400 g/hr capacity and a bench scale reactor of 1 kg/hr capacity, a test plant to treat 200 kg/hr of pitch is being constructed. This test plant is to be completed in September, 1978, and will be capable of generating from 1 t of vacuum residual oil 0.5 t of low sulfur fuel oil as well as reducing gas enough to produce 3 t of reduced iron.

5. Reduced Iron Production System

During the planning stage, both a shaft furnace and a fluidized bed reactor were in operation, each producing about 1 000 t/day of reduced iron using reducing gas. After comparing and considering these two systems, however, the shaft furnace system was selected for this nuclear steelmaking project. High pressure operation was required for the shaft furnace to be compatible with other units in the total nuclear steelmaking system, but at that time there was no instance of a shaft furnace operating at such pres-

Photo. 3. Heat exchange type steam reformer test plant

Table 4. Design data of heat exchange type steam reformer

Shell	Outside diameter	1 580 mm
	Height	14 000 mm
Tube	O. D.	72 mm
	I. D.	50 mm
	Effective height	8 760 mm
	Number	8
Capacity	100 kg/hr (Naphtha)	

tures.* Therefore the problems considered were the improvement of productivity and energy efficiency resulting from the application of high pressure and the determination of the optimum range of the reducing gas composition. Also, the development of new equipment to transfer solids and gases into and out of the shaft furnace and to control the distribution of gas flow was required.

Iron ore reduction experiments¹³⁾ were conducted at atmospheric pressure for various kinds of pellets with the JIS reducing apparatus (JIS M8713). Valuable data were obtained on the effects of temperature and of the CO_2 , H_2O and CH_4 contents on the reduction rate and reduction degree attained after a specified time. Other investigations completed were on the effect of the $\text{CO}:\text{H}_2$ ratio in the reducing gas, carbon deposition, pellet degradation and the strength of the reduced iron.

In order to further clarify the reducing reaction of iron ore at high pressures, a shaft furnace simulator, as shown in Fig. 9, was constructed, and experiments were initiated in February, 1975. With this equipment, the specimens were subjected to the same history of thermal and reducing conditions encountered by an iron ore pellet charged into the shaft furnace. By following the behavior of the specimen as it is charged into the top, converted into reduced iron and discharged out the bottom, a simulation model was established. The diameter of the reaction pipe was 120 mm, and the specimen composed a fixed layer weighing 3 to 5 kg. The maximum pressure and the highest temperature of the reducing gas were 15 atm and 1 000°C, respectively, and the composition was freely changeable. By utilizing this equipment, the reduction rate, the rate of formation and decomposition of methane, and the carbon deposition reaction were examined. Also, data for

optimizing the reducing gas conditions in the shaft furnace were obtained. The principal results were as follows:

(1) The pressure increase in the shaft furnace caused carbon deposition as well as the violent formation of methane.

(2) The desired conditions to prevent carbon deposition were in the low pressure side of the shaft furnace operation.

(3) The effects of composition, pressure and degree of oxidation of the reducing gas on the reducing time were clarified.

(4) Reducing gas conditions that promise stable operation of the shaft furnace and efficient productivity were established.

(5) A high precision mathematical models for simulation purposes was developed making it possible to estimate the shaft furnace capacity and the quantity of reducing gas needed.

As for auxiliary equipments for the high pressure furnace, experiments were conducted with several cold models. Out of those studies, a charging and discharging system, shown in Photo. 4, a seal valve and a stop valve, both special for this system, were developed. Also, a study on gas flow was conducted in preparation for scaling-up of the shaft furnace and for selecting the furnace configuration.

To develop a system to recover heat from the reduced pellets, experiments and simulation model exercises were carried out. The effect of changing the distance between the blast and suction nozzles on the gas flow pattern was investigated. Also, the number and diameter of the nozzles and the shaft furnace diameter were varied to clarify their effects on the gas flow.

On the basis of these results, a 2 000 t/day shaft furnace that is compatible with the projected experi-

Fig. 9. Basic concept of the shaft furnace simulator

* Subsequently, Hirohata Works, Nippon Steel Corp., operated a 500 t shaft furnace with a top pressure of 4.5 kg/cm²G.

Fig. 11. Response of steam reformer to secondary helium inlet temperature change (Secondary helium inlet temp. -20°C step change)

REFERENCES

- 1) R. S. Barnes: *Iron Steel Eng.*, **53** (1976), 5, 53.
- 2) D. J. Blickwede: *Iron Steel Eng.*, **53** (1976), 4, 38.
- 3) H. Juntgen and K. H. van Heek: *Nuclear Eng. and Design*, **34** (1975), 1, 59.
- 4) L. Schrader, W. Schtrauss and H. Teggars: *Nuclear Eng. and Design*, **34** (1975), 1, 51.
- 5) Status of Research and Development on Multi-purpose HTGR, ed. by Japan Atomic Energy Research Institute, (1976).
- 6) Second Report of the 122nd-123rd Joint Committee on Heat Resisting Materials, The Japan Society for the Promotion of Science, (1975).
- 7) R. Watanabe, Y. Chiba and T. Kuno: *Tetsu-to-Hagané*, **61** (1975), 2405.
- 8) R. Watanabe and Y. Chiba: *Tetsu-to-Hagané*, **63** (1977), 118.
- 9) Y. Hosoi, A. Shinoda, Y. Tsuchida, S. Sekino, N. Sakakibara and H. Honma: *Tetsu-to-Hagané*, **62** (1976), S803.
- 10) S. Ohta, K. Aota, T. Motoda and T. Honjo: *Tetsu-to-Hagané*, **62** (1976), S802.
- 11) T. Yukitoshi, K. Yoshikawa and T. Sawaragi: Report of the 123rd Committee on Heat Resisting Metals and Alloys, The Japan Society for the Promotion of Science, Rep. No. 18, (1977), 163.
- 12) T. Ohmura, Y. Sahira, A. Sakonooka and N. Yonezawa: *Tetsu-to-Hagané*, **61** (1975), S323.
- 13) A. Sasaki and K. Okabe: *Tetsu-to-Hagané*, **61** (1975), S3.