ISIJ International, Vol. 46 (2006), No. 10, pp. 1403–1409

Production of Pig Iron from Magnetite Ore–Coal Composite Pellets by Microwave Heating

Kotaro ISHIZAKI,[1] Kazuhiro NAGATA[1] and Tetsuro HAYASHI[2]

1) Department of Chemistry and Materials Science, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo 152-8552 Japan. 2) Seki Branch Laboratory, Gifu Research Institute of Industrial Product Technology, Oze 1288, Seki-shi, Gifu 501-3265 Japan.

(*Received on March 27, 2006; accepted on July 23, 2006*)

Magnetite ore–coal composite pellets with about 10, 15 and 20 mm diameter were rapidly smelted to produce pig iron by microwave heating in N_2 gas. A microwave generator with 5 kW maximum power at 2.45 GHz was employed. Carbon content in pig iron was about 2 mass% near the liquidus line in the Fe–C system. Slag was easily separated from pig iron. By XRD analysis it is realized that the reduction of magnetite started at about 800°C and was completed to be pig iron at about 1 350°C. The heating rate of pellets was independent of their mass but dependent on applied power because of self heating. According to the increase of heating rate, the level of impurities in pig iron decreased less than blast furnace.

KEY WORDS: pig-iron; magnetite ore; coal; composite pellets; microwave heating.

1. Introduction

Owing to the shortage of suitable energy resources and the introduction of more stringent environmental laws a search for new pig iron and steelmaking processes is important. One approach is to shorten the time and/or to lower the temperature for producing iron in comparison with blast furnace methods.

To obtain pig iron from iron ore there are two main reactions; one is reduction by carbon and the other carburization. Both of them are endothermic reactions and therefore to make pig iron rapidly it is necessary to heat reactants quickly. Fine iron ore powder is very effective to accelerate the reduction of iron oxide by CO gas because finer particles have larger surface area relative to the volume. In the case of blast furnace, heat supply to iron ore is conducted by high temperature gas. Hot air at about 1 200°C is blasted into the blast furnace from tuyers located in the lower part of the furnace and hot air burns coke. If a fine iron ore powder is loaded in blast furnace, it flies away with hot air and prevents hot gas from flowing through blast furnace. Because of these reasons fine iron ore powders have not been used in blast furnace. Therefore it is difficult to improve further the efficiency of blast furnace for producing pig iron. Consequently it is necessary to investigate new iron making methods.

Nagata *et al.*[1] have clearly shown that by using a conventional electric furnace, pig iron can be produced from magnetite ore pellets containing coal at furnace temperatures higher than 1 325°C within about 16 min. These findings have been partially applied in the FASTMET direct-reduction processes.[2] In the FASTMET process, fine iron ore is pelletized together with a carbonaceous reductant. The pellets of magnetite ore with coal are heated to approximately 1 300°C in a reverberatory rotary hearth furnace and reduced to 90% metallization after about 15 min.[3] In this FASTMET process it is not possible to pile up pellets in multilayer because the production time becomes longer and heat is not uniform.[4] Using a conventional heating method, it is not possible to obtain pig iron from pellets placed in multilayer in a short time because the reaction is endothermic. A new technology of coal-based iron making, the HiQIP (High Quality Iron Pebble) process to produce pig iron pebbles for electric arc furnace has been developed. These processes are characterized by producing reduced iron directly from the mixture of fine ore and fine carbonaceous material, and by melting reduced iron to separate metal and slag in a rotary hearth furnace.[5,6]

In the present study, the production method of pig iron by microwave as an alternative heating way is investigated. Microwave energy has potential for the speedy and efficient heating of minerals and may provide savings in both time and energy.[7] The frequency range of microwave is from 0.3 to 300 GHz corresponding to a wavelength range from 1 mm to 1 m between radio wave and infrared radiation.[8] Microwave heating has a number of advantages over conventional heating. It is volumetric, rapid, as well as selective without contacting materials.[9] When microwaves are applied to a dielectric material, internal electric fields generated within the affected volume induce translational motions of free or bound charges and rotate charge complexes such as dipoles. The absorbed energy of microwave is dissipated as heat. Microwave heating is therefore internal meanwhile conventional is external.[10] In microwave heating temperature in a pellet is different from particle to particle because of their different efficiency to absorb mi-

Table 1. Chemical composition of pellet in mass%; (a) compounds and (b) elements.

a) Compounds

mass%	Fe_3O_4	FeO	Fe_2O_3	SiO_2	Al_2O_3	CaO	MnO	S	C	Volatiles
Pellet	73.10	0.31	0.10	3.41	1.27	0.42	0.34	0.13	15.68	4.29

b) Elements

	Fe	O		Si	Al	Ca	Mn	S	C	Volatiles
mass%	53.21	22.92		1.60	0.67	0.30	0.26	0.13	15.68	4.29

crowave energy, but in conventional heating heat absorption is almost uniform for all components. Roy *et al.* have indicated that elements in a sample heated by microwave are under an-isothermal conditions.[11)]

The reduction of iron oxides using microwave energy as heating source has been previously investigated. Standish and Huang[12)] first demonstrated that the carbothermic reduction of both magnetite concentrates and hematite fines could be satisfactorily and rapidly carried out with microwave heating. Zhong *et al.*[13)] reported on the reduction of a low-silica taconite concentrate by coke or coal using an industrial microwave generator. Maurao *et al.*[14)] investigated the carbothermic reduction of composite pellets containing a hematite iron ore, coke or charcoal as carbonaceous material. Nagata *et al.*[15)] showed that pig iron was obtained from magnetite ore–coal composite pellets by microwave heating. Chen *et al.*[16)] indicated that microwave heating with carbothermal reduction can increase the metallization rate of iron ore concentrates containing coal.

Morita *et al.*[17)] could successfully recover about 70% of iron and 25% of phosphorous from factory steel-making slag by carbothermal reduction using microwave irradiation.

The present study shows that the production of pig iron from magnetite ore–coal-composite pellets using microwave heating is possible at a lower temperature and shorter time than by electric furnace.[1)] The effects of different pellet size and microwave input power on the production process and the content of impurities in pig iron have been investigated.

2. Experimental

2.1. Sample Preparation

Spherical composite pellets with about 10, 15 and 20 mm diameter were prepared using a pelletizer from a mixture of magnetite ore (named Romeral) (*ca.* 50 μm diameter), coal (named Robe River) (*ca.* 40 μm diameter) and bentonite powder as a binder.

The chemical compositions of raw materials are given in **Table 1**. The utility of carbon for reduction of iron ore is ideally to be pure CO_2 and about 70% CO+30% CO_2 for blast furnace. It may be possible to increase the utility for this process. The amount of coal was determined by taking into account the amount of carbon required for reducing iron ore completely to form 87% CO+13% CO_2 gas and for producing pig iron with 2 mass% of carbon. The content of bentonite was 2 mass%. The weight and apparent density of the pellets are shown in **Table 2**.

2.2. Experimental Procedure

A schematic illustration of the experimental apparatus is shown in **Fig. 1**. A microwave generator with 5 kW maximum power at 2.45 GHz was employed. As the shape of the microwave oven chamber is pentagon, the electric field (*E*-field) and the magnetic field (*H*-field) modes of microwave could be mixed. One or four pellets were set in the middle of the microwave oven chamber and covered with an alumina insulator. Silicon carbide as an exothermic auxiliary substance was pasted inside of the alumina insulator to heat it and compensate the heat lost from the pellet.

The chamber was first evacuated to 0.03 torr and then filled with 99.9% N_2 gas. The temperature of pellets above 600°C was monitored using a two color radiation thermometer from the upper part of the chamber through a window as shown in Fig. 1. The reaction progress of a pellet was observed from a small window located in the sidewall of the chamber.

The pellet setup shown in "Sample A" of Fig. 1 corresponds to one pellet with 20 mm diameter placed in an alumina dish and microwave heated by the step power supply pattern shown in **Fig. 2**(a). The pattern was composed of initial power of 0.2 kW, after 2 min 0.7 kW, followed by 1 kW, 1.5 kW, 1.75 kW and 2 kW in 2 min intervals. The

Table 2. Specifications of pellets and heating pattern.

Sample No.	Weight (g)	Density*(g/cm^3)	Heating pattern
10-ST-C	1.36~1.81	1.81~2.34	Step up to 2 kW
15-ST-C	3.66~4.30	2.04~2.28	Step up to 2 kW
20-ST-C	7.57~9.63	1.95~2.04	Step up to 2 kW
20-C1-C	7.83	2.00	Const. 1 kW
20-C2-C	7.68	1.76	Const. 2 kW
20-C3-C	8.19	1.89	Const. 3 kW

*apparent density

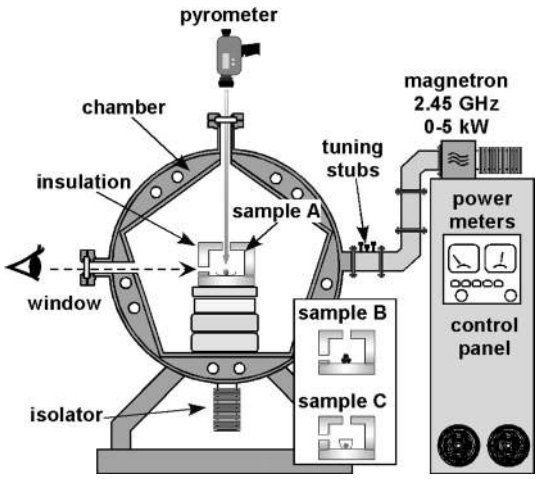

Fig. 1. Schematic illustration of the experimental apparatus. The inside of the insulator was coated with SiC as an auxiliary heater. A, B and C indicate; one pellet in an alumina dish, four pellets pilled in pyramid placed on an alumina plate and one pellet in an alumina crucible respectively.

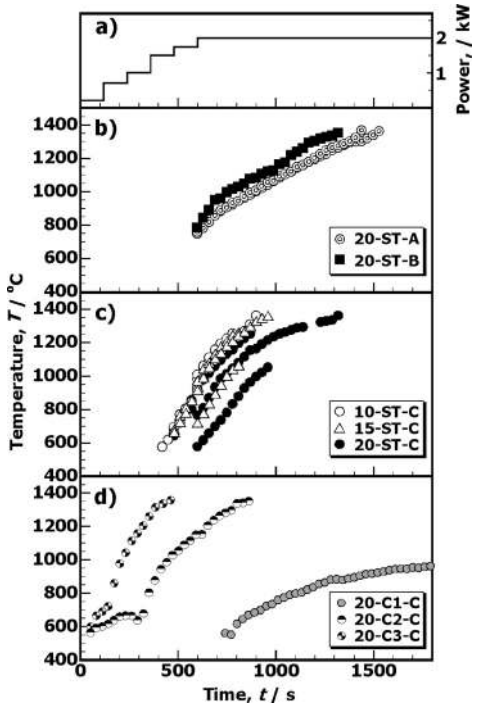

Fig. 2. (a) The step power supply pattern (ST). Temperature increase of pellets heated by microwave; (b) and (c) step power supply (ST), and (d) by constant power supplies (C1, C2 and C3 corresponding to 1 kW, 2 kW and 3 kW, respectively. 10, 15 and 20 are diameter of pellets in mm. A, B and C indicate; one pellet in an alumina dish, four pellets pilled in pyramid placed on an alumina plate and one pellet in an alumina crucible respectively.

constant power of 2 kW was kept until the end of heating. The sample setup shown in "Sample B" of Fig. 1 corresponds to four pellets with 20 mm diameter piled in pyramid on an alumina plate and heated by the step power supply. "Sample C" of Fig. 1 indicates one pellet placed in an alumina crucible. The specifications of pellets and heating patterns for "sample C" are given in Table 2. In order to investigate the progress of reaction, pellets were also heated up to 800, 1 050, 1 150 and 1 250°C by the step power supply under the "sample C" condition and cooled in the chamber.

2.3. Analysis

The initial weight of each pellet and the final weight after heating were measured.

The pellets cooled from 800, 1 050, 1 150 and 1 250°C were cut in quarters. The pig iron pebbles were cut in half. One quarter of the pellet and half of the pig iron were fixed in resin and the cross sections were mechanically polished. The impurities in pig iron were analyzed using a sequential X-ray florescence spectrometer (XRF); Shimadzu XRF-1800.

The other quarter pieces were analyzed for identifying the crystal phases of reactants and products using X-ray diffractometer (XRD); Rigaku RINT-TTR-3C/PC.

3. Results

3.1. Temperature Increase of Pellets

Figure 2(b) shows the time–temperature profiles of one and four pellets heated by the step power supply without crucible. Figure 2(c) shows the profiles of pellets with 10, 15 and 20 mm diameter heated in an alumina crucible by the step power supply. The pellet size does not seem to affect the temperature increase rate. On the other hand the temperature increase rate for pellets with 20 mm diameter heated under constant power increased with increasing microwave power, as shown in Fig. 2(d). 1 kW power was not enough to heat a pellet with 20 mm diameter up to the temperature for producing pig iron. In the case of 2 and 3 kW power supplies, the pellets heated very rapidly up to approximately 600°C.

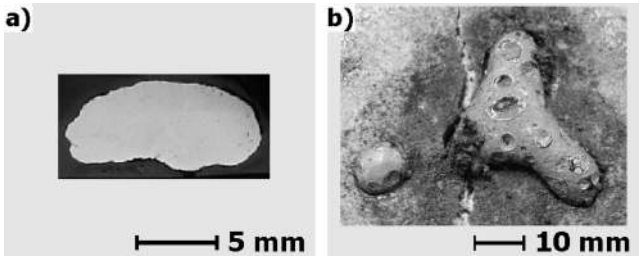

Fig. 3. Pig iron products; (a) cross section of pig iron obtained from 20-ST-A and (b) product of 20-ST-B.

3.2. Reaction Products

One pellet placed on an alumina dish and four pellets on a plate ("sample A, and B"), respectively were heated by the step power supply. After the start of heating, the inside of alumina insulator brightened and pellet was clearly observed. Soon, soot came out of pellet and ceased. During the step-up of microwave power, a yellow flame was generated from pellet during heating, pellet gradually brightened in red to yellow. When the temperature of pellet attained at about 1 350°C, pellet melted down to be pig iron and disappeared from the view. After the temperature of pig iron reached 1 360°C, the power was shut down and then products were cooled down to room temperature in nitrogen atmosphere. For one pellet it took between 10 to 15 min to cool down to room temperature, and for four pellets 20 min. On the surface of pig iron in pebble slag was partially adhered, as shown in **Fig. 3**(b) and **Fig. 4**. From the cross section of pig iron pebble, slag and gang materials did not include in pig iron, as shown in Fig. 3(a). **Figure 5**(a) shows an original pellet with 20 mm diameter. Pellet heated by 1 kW constant power could not give pig iron (Fig. 5(b)). For pellets heated by 2 and 3 kW constant power, as shown in Figs. 5(c) and 5(d), respectively, gave pig iron pebbles and powder composed mainly of slag droplets and residual carbon.

3.3. Carbon Content in Pig Iron

The carbon concentration of pig iron is plotted in the Fe–C phase diagram[18] of **Fig. 6**. The carbon concentrations of pig iron from one pellet heated by the step power supply in an alumina crucible are in the coexisting phase of solid and liquid and by constant power supply in the liquid phase near the liquidus-line. In the case of one and four pellets heated by the step power supply without alumina crucible the carbon concentrations are on the liquidus-line.

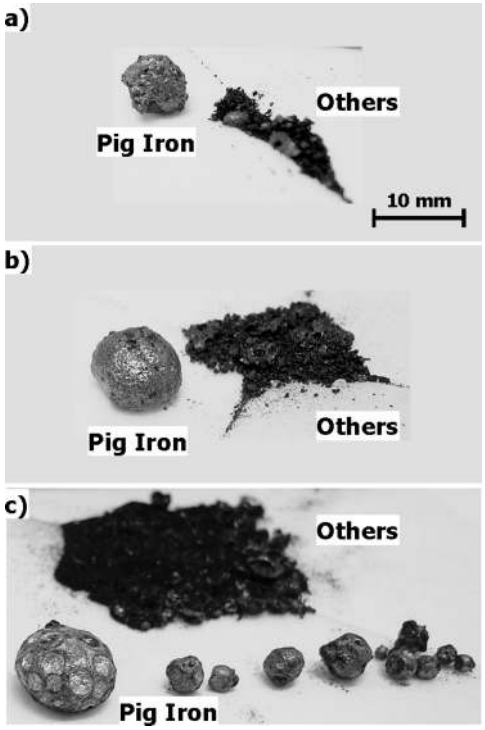

Fig. 4. Reaction products from (a) 10-ST-C, (b) 15-ST-C and (c) 20-ST-C.

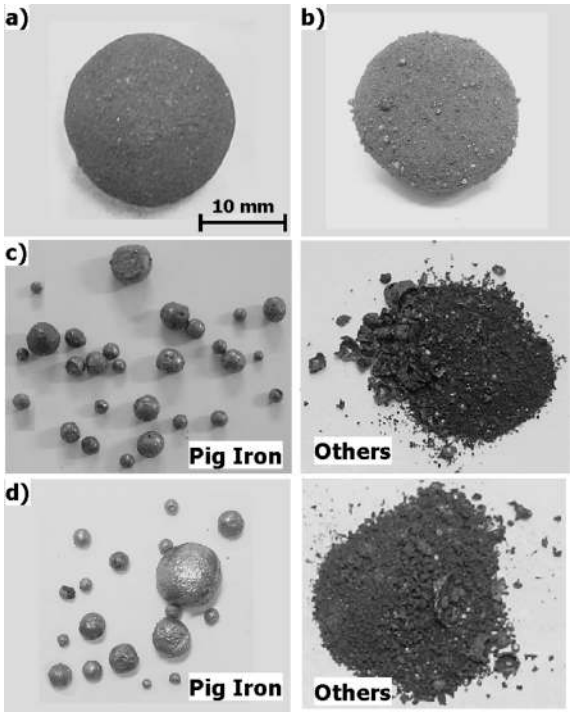

Fig. 5. View of pellet and products; (a) original pellet. (b), (c) and (d) reaction products from 20-C1-C, 20-C2-C and 20-C3-C, respectively.

3.4. Impurities in Pig Iron

The impurities in pig iron are shown in **Figs. 7**(a) and 7(b) for the step and constant power supplies, respectively. Pig irons from pellets placed in a crucible and heated by the step power supply have higher concentration of impurities than those heated by constant power supply. The content of impurities in pig iron from one and four pellets heated without alumina crucible by the step power supply are

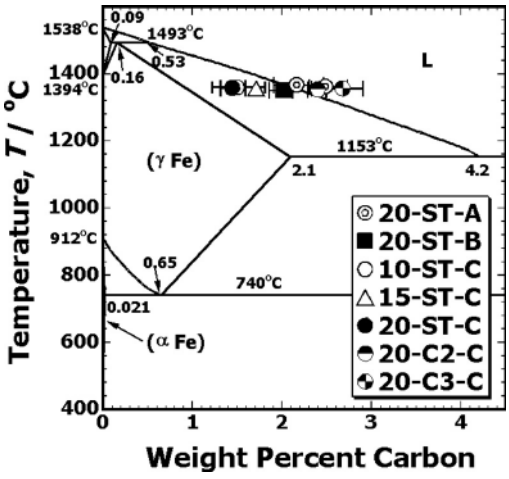

Fig. 6. Carbon content in pig iron produced by microwave heating in step and constant power supplies.

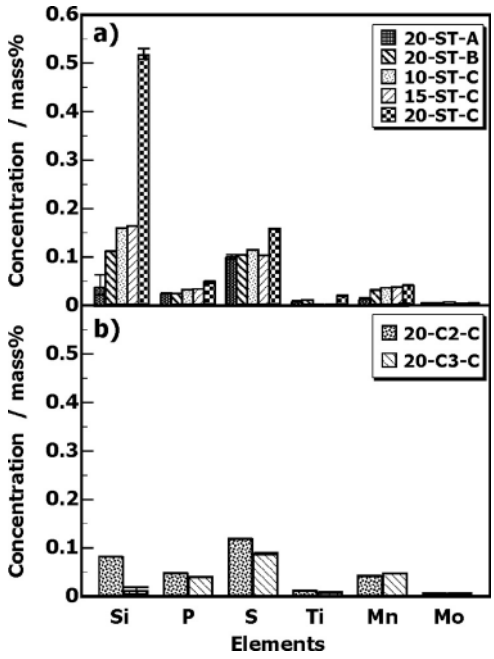

Fig. 7. Contents of impurities in pig iron produced by microwave heating; (a) step power supply and (b) constant power supply

slightly lower than those from pellets heated in an alumina crucible. Pig iron produced in faster heating contains lower content of impurities.

3.5. Transformation of Iron Oxides and Weight Loss

In order to follow the reduction process, pellets were heated by the step power supply up to different temperatures and the products analyzed by XRD. The analysis was carried out in several points between the center and the surface of pellets. No significant difference was detected in the results. As an example the XRD of the 20 mm pellet heated up to 1 050°C taken in the center a) and near the surface b) of the pellet are shown in **Fig. 8**. **Figure 9** shows the XRD patterns of products in pellets with 10 mm, 15 mm and 20 mm diameter heated up to 800°C, 1 050°C, 1 150°C and 1 250°C taken in a point between the center and the surface of each pellet. For all pellets at 800°C a little amount of wustite (FeO) is observed. At 1 050°C, a little amount of

the initial magnetite (Fe_3O_4) is present and the majorities are wustite (FeO) and iron (Fe). At 1 150°C, the proportion of reduced iron (Fe) increased and at 1 250°C only reduced iron (Fe) was detected.

The weight loss percent of pellets with 10, 15 and 20 mm, heated by the step power supply was plotted *versus* heating time as shown in **Fig. 10**. In this figure the weight loss percent includes volatiles evolved at the beginning of heating.

4. Discussion

4.1. Effect of Pellet Mass and Microwave Power on Temperature Increase Rate

For pellets heated with the constant power of 2 and 3 kW, the patterns of temperature increase can be classified into four stages; I) very rapid increase up to about 600°C, II) stagnant from 600 to 700°C, III) rapid increase to about 1 000°C and IV) slow increase until pig iron melts down at about 1 350°C. It is clear that by the constant power supply of 2 kW and 3 kW, heating was very fast up to about 600°C, as shown in Fig. 2(c). Stages I and II could not be recorded for pellets heated by the step power supply but the starting time of the stage III is around 400 to 600 s, when the power attained 1.5 to 2 kW, meanwhile it is about 300 s for pellet heated by the constant power supply of 2 kW. This probably means that the temperature increased in steps according to the power in step increase. Despite the different weight of pellets and the different ways of holding them on a dish (A), a plate (B) or in a crucible (C), temperature increases of pellets after 700°C are similar to that of pellet heated by the constant power supply of 2 kW, as shown in Fig. 2. The time lag of heating up to about 600°C for some pellets heated by the step power supply may be caused by the different reflection of microwave due to the condensation of

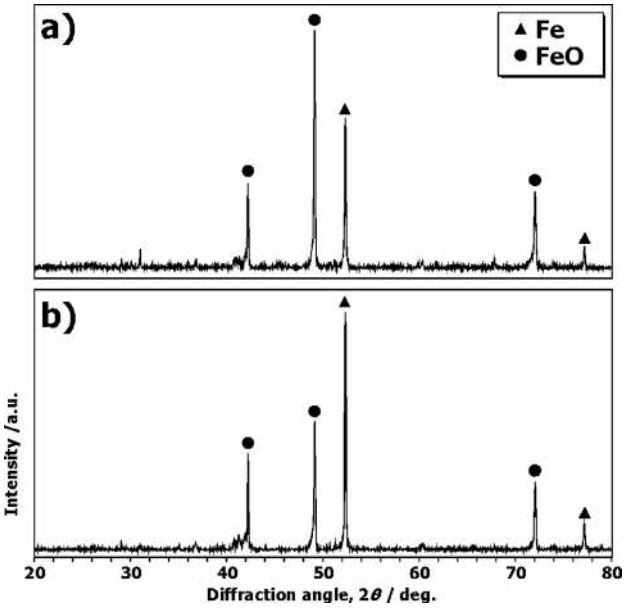

Fig. 8. XRD patterns of the 20 mm diameter pellet heated up to 1 050°C taken at the center (a) and near the surface (b).

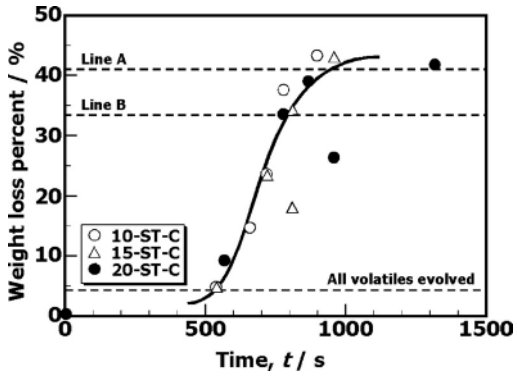

Fig. 10. Weight loss % as a function of heating time for 10, 15 and 20 mm diameter pellets heated by microwave in step power supply. Hatch lines A and B corresponds to the weight loss % including volatiles and 87% CO gas+ 13% CO_2 gas or 100% CO_2 gas respectively.

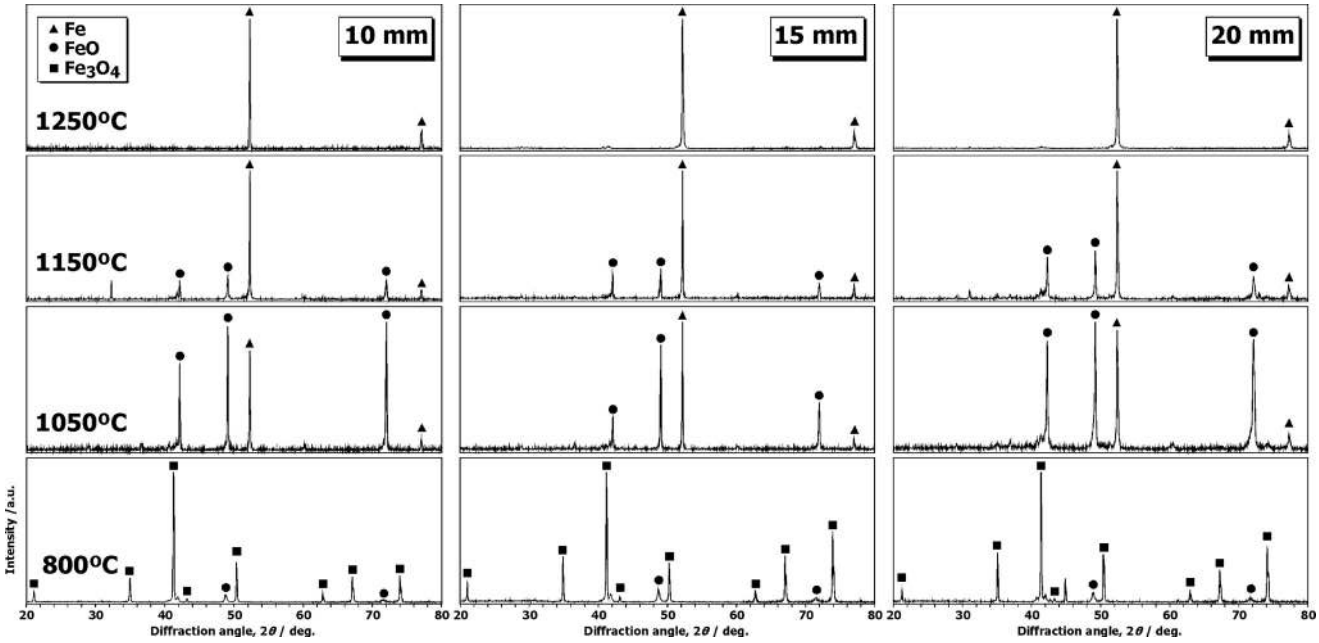

Fig. 9. Identification of iron oxide products reduced by carbon at 800°C, 1 050°C, 1 150°C and 1 250°C for 10, 15 and 20 mm diameter pellets heated by microwave in step power supply.

evolved volatiles on the chamber walls of the oven. Magnetite and carbon in pellet absorb microwave and generate heat.[10] Cheng *et al.*[19] reported the temperature–time profiles of magnetite heated by electric and magnetic fields separately. In magnetic field the profile shows that magnetite was heated up to about 650°C and stayed at this temperature. It is well known that magnetic power is not absorbed in magnetite above the Curie temperature of 587°C,[20] which is very close to the initial temperature rise by the microwave constant powers of 2 and 3 kW. Then, the temperature of pellet stagnates at 600 to 700°C in the stage II. As the electric conductivity of wustite[21] increase with increasing temperature and larger than that of magnetite, wustite are heated by electric field and the temperature of pellet rises in the stage III.

From the XRD results shown in Fig. 9, the reduction of iron oxide in pellets heated by the step power supply started at about 800°C. Since the reduction of iron oxide by carbon is endothermic reaction and reduced iron absorbs little microwave energy, above 800 to 1 000°C the temperature increase rate slows down in stage IV.

Since the XRD patterns taken at the center and near the surface of heated pellets at different temperatures were similar as shown in Fig. 8 as an example, it is realized that compared to the results of Nagata *et al.*[1] using conventional heating, in the case of microwave heat is generated inside the pellet. The results also show that the generation of heat and the temperature increase rate of pellet are dependent on microwave power.

4.2. Reaction Rate of Pellets

Most of the weight per unit original weight of pellets with 10, 15 and 20 mm diameter heated by the step power supply decrease after 550 s, as shown in Fig. 10. At this time the power reached a value of 1.75 kW. The XRD patterns in Fig. 9 show that iron oxide is reduced above 800°C. In **Fig. 11** the weight loss percent of pellets excluding the volatile fraction of 4.4% are plotted against the corresponding temperatures reached by the pellets. This plot includes data up to 1 250°C because according to the XRD results of Fig. 9, the reduction to Fe has finished for all pellets. From this figure it is possible to estimate the temperature at which the reduction starts. In the case of the conventional heating, Nagata *et al.*[1] reported that pellets with 20 mm diameter started to reduce at about 1 000°C and carburized at about 1 300°C. By the step power supply of microwave, the reduction could starts at about 820°C and the carburization at about 1 250°C.

The results of the present work are in agreement with previous reports.[12–14] They indicate that microwave energy dissipates rapidly throughout the volume of material and heats it directly with a significant increase in the reduction rate in comparison with conventional heating system.

4.3. Low Content of Impurities in Pig Iron

Figures 7(a) and 7(b) show the contents of impurities in pig iron produced by the step and constant power supplies of microwave, respectively. The contents of impurities are lower than those of pig iron produced by blast furnace (JIS G2201-1976). With the exception of Si, the concentrations of all other impurities are in a similar level independent of the weight of pellet. Pig iron with high Si content was produced by the step power supply with long contact to slag.

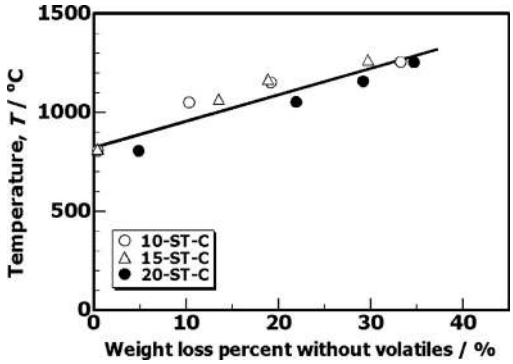

Fig. 11. Correlation between weight loss % and temperature for 10, 15 and 20 mm diameter pellets heated by microwave step power supply.

The contents of impurities decrease with increasing microwave power and increasing the temperature of pellet rapidly.

The low contents of impurities in pig iron produced by microwave heating are explained as follows. Nagata *et al.*[1] showed that by increasing the heating rate of pellet, the oxygen partial pressure in the pellet increases and impurity oxides in the pellet were not reduced. In this manner, impurity oxides remain in slag.

5. Conclusions

Pig iron was rapidly produced from the mixed powder of commercial magnetite ore and coal at low temperature (about 1 350°C) by heating in the step and constant power supply of 2 and 3 kW.

Carbon content in pig iron was of about 2 mass% near the liquidus line in the Fe–C system.

Slag separated easily from pig iron. The contents of impurities in pig iron were very low.

By XRD analysis it is realized that the reduction of magnetite started at around 820°C and it was completed to iron at the temperature between 1 150°C and 1 250°C.

REFERENCES

1) K. Nagata, R. Kojima, T. Murakami, M. Susa and H. Fukuyama: *ISIJ Int.*, **41** (2001), 1316.
2) J. Jimbo, H. Tanaka, T. Sakaguchi and Y. Kuwata: *Kobelco Techno. Rev.*, **22** (1999), 60.
3) T. Coetsee, P. C. Pistorius and E. E. de Villiers: *Miner. Eng.*, **15** (2002), 919.
4) T. Harada, H. Tanaka and H. Sugitatsu: *Kobe Steel Eng. Rep.*, **51** (2001), 23.
5) Y. Sawa, T. Yamamoto, K. Takeda and H. Itaya: *ISIJ Int.*, **41** (2001), S17.
6) K. Takeda: *Kinzoku*, **75** (2005), 547.
7) R. M. Kelly and N. A. Rowson: *Miner. Eng.*, **8** (1995), 1427.
8) M. A. M. Khraisheh, T. J. R. Cooper and T. R. A. Magee: *J. Food Eng.*, **33** (1997), 207.
9) A. Idris, K. Khalid and W. Omar: *Appl. Therm. Eng.*, **24** (2004), 905.
10) W. H. Sutton: *Ceram. Bull.*, **68** (1989), 376.
11) R. Roy, R. Peelamedu, L. Hurtt, J. Cheng and D. Agrawal: *Matel. Res. Innovat.*, **6** (2002), 128.
12) N. Standish and W. Huang: *ISIJ Int.*, **31** (1991), 241.
13) S. Zhong, H. E. Geotzman and R. L. Bleifuss: *Miner. Metall. Process.*, **300** (1996), 174.

14) M. B. Mourao, I. Parreiras de Carvalho, Jr. and C. Takano: *ISIJ Int.*, **41** (2001), S27.
15) K. Nagata, K. Ishizaki and T. Hayashi: Proc. of the 5th Japan-Brazil Symp. on Dust Processing-Energy-Environment in Metallurgical Industries, 1, (2004), 617.
16) J. Chen, L. Liu, J. Zeng, R. Ren and J. Liu: *Iron Steel*, **39** (2004), 1.
17) K. Morita, M. Guo, N. Oka and N. Sano: *J. Mater. Cycles Waste Manag.*, **9** (2002), 93.
18) Binary Alloy Phase Diagrams Second Edition, Vol. 1, ed. by T. B. Massalski, H. Okamoto, P. R. Subramanian and L. Kacprzak, Published by William W. Scott Jr., ASM Int., Materials Park, OH, (1990), 843.
19) J. Cheng, R. Roy and D. Agrawal: *Mater. Res. Innovat.*, **5** (2002), 170.
20) Rika Nenpyo (Chronological Scientific Tables), Vol. 70, ed. by National Astronomical Observatory, Maruzen Co., Ltd., Tokyo, (1996) 538.
21) K. Endo: Sanka-Butu-Binran, Nisso, Wakayama, (1970), 267.