Microwave Frequency Effect for Reduction of Magnetite

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This paper describes experimental investigations on pig iron production by use of microwave heating. In order to explore possible effects of the microwave frequency identical mixtures of magnetite and carbon powder were processed in different microwave systems, operating at 2.45 GHz and 30 GHz, respectively. High quality pig iron was obtained in the 30 GHz mm-wave system in air. However, at 2.45 GHz FeO was produced mainly at similar conditions. This result might suggest that the chemical reduction of magnetite is more efficient with higher microwave frequencies.

Keywords: microwave processing, millimeter-wave processing, magnetite, pig iron,

1. Inroduction

Microwave technology is not only a substitute for conventional heating, but it resides in the new domain of materials science, namely, microscopic and strong thermal non-equilibrium systems [1]. The application of microwaves in the iron industry may be characterized by a high potential for an essential reduction of carbon dioxide emission. Iron ore refinement by means of blast furnaces was realized with the same basic furnace structure based on the same principle for two centuries. We have conducted a series of experiments to prove effectiveness of rapid and high purity refinement under low temperature and oxygen-containing environment by means of microwave application, and achieved highly positive results.

Nagata and coworkers of the Tokyo Institute of Technology have been working on the development of unique ultra high purity iron refinement technology based on an ancient Japanese iron refinement method called "Tatara" [2]. Their findings during microwave sintering of powder metals led to the idea that rapid refinement of iron should be possible by application of 2.45 GHz microwaves instead of relying on burning of carbon for heat production. Joint experiments at the National Institute on Fusion Science (NIFS) and Forschungszentrum Karlsruhe (FZK) proved that high purity iron (1% carbon concentration) with less than 1/10 of impurities as compared to irons that the modern blast furnaces can produce. More over it reduced the carbon consumption to 2/3.

In order to investigate the effect of microwave frequency, samples of magnetite powder mixed with carbon powder were processed in different microwave systems. The following paper discusses recent experimental results obtained by millimeter-wave (mm-wave) processing and processing in a 2.45 GHz microwave system.

2. Experimental Setup of theMM-wave Process

For mm-wave experiments, the applicator of a compact 30 GHz gyrotron system was used as shown in Figure 1(a) [3]. The mm-wave power generated by a so called gyrotron oscillator can be controlled from 0 - 15 kW. This power is launched via a quasi-optical transmission line through a vacuum-sealed boron nitride window into the hexagonal applicator which is characterized by a very homogeneous field distribution. The samples used were mixed powder of magnetite and carbon. The weight ratio of magnetite and carbon in the power mixture was 89 to 11 weight%. According to the corresponding chemical equation this should allow to produce pig iron including 2 weight% of carbon. Such type of powder samples were filled into an alumina crucible surrounded by thermal insulation (see Fig. 1(b)). The temperature was measured by two S-type thermocouples, one sticking in the center of the powder sample, another near the crucible wall. The heating process was controlled along a preset temperature-time program with a heating rate of 70 °C/min using the temperature signal of the thermocouple placed near the sample surface.

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Fig.1 (a) 30GHz gyrotron system (top),
(b) Experimental setup of the mm-wave (bottom)

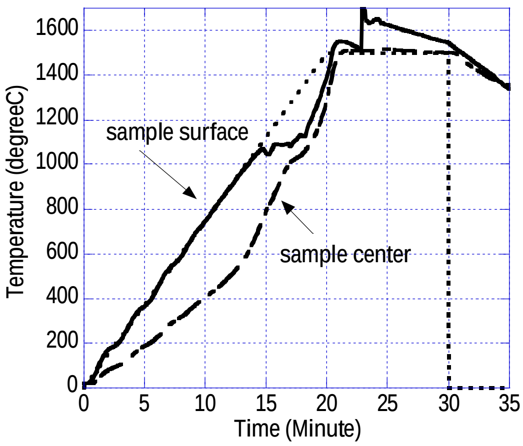

Fig.2 The measured temperatures of a mm-wave process.

Fig.3 Pig iron from the 30GHz mm-waves process.

Figure 2 shows the process temperatures measured during mm-wave heating of a powder sample of about 80 g mixed magnetite and carbon. It can be seen that during the initial step of heating that means during the first 15 minutes, the temperature measured at the sample surface was higher than the temperature measured within the sample volume. However thereafter, at a temperature of about 1100 °C the absorption behavior of the powder is changing dramatically. According to the Iron-Carbon phase diagram this temperature is close to the point where liquefaction of iron with 2% carbon content starts. That means microwave power is consumed by the melting process and heating of the material stagnates. Due to this melting process a strong rearrangement of the processed material happens, leading to changes in the thermocouple positions as well. Finally, when the material is completed molten, the thermal conductivity of the material increased. Thus the measured temperatures converge to each other. Figure 3 shows a picture of the obtained pure pig iron. EDX analysis along the cross section revealed a carbon content of 1 weight%. No oxygen could be detected.

3. Experimental Setup for CM-wave Process

The multimode test furnace at the National institute for Fusion Science shown in Figure 4 was employed for the present study at 2.45 GHz. According to the concepts developed in Germany in order to improve the homogeneity of the electromagnetic field, the applicator shape is hexagonal [4]. The furnace with 0.92 m^3 in volume is equipped with five magnetrons. The microwave power of a single magnetron is 2.5 kW. Two mode stirrers scatter the standing wave. The samples used had the same weight ratio of magnetite and carbon power as for the mm-wave process. The powder samples were filled into an alumina crucible surrounded by thermal insulation (see Figure 4) similar to the setup used for the mm-wave process. The temperature was measured by an IR pyrometer. The heating process was controlled along a preset temperature-time program with a heating rate of 70 °C/min using the temperature signal of the IR pyrometer measured at the top of the sample surface. In addition, the multi-point emission spectroscopic diagnostic was conducted through the viewing port on the furnace.

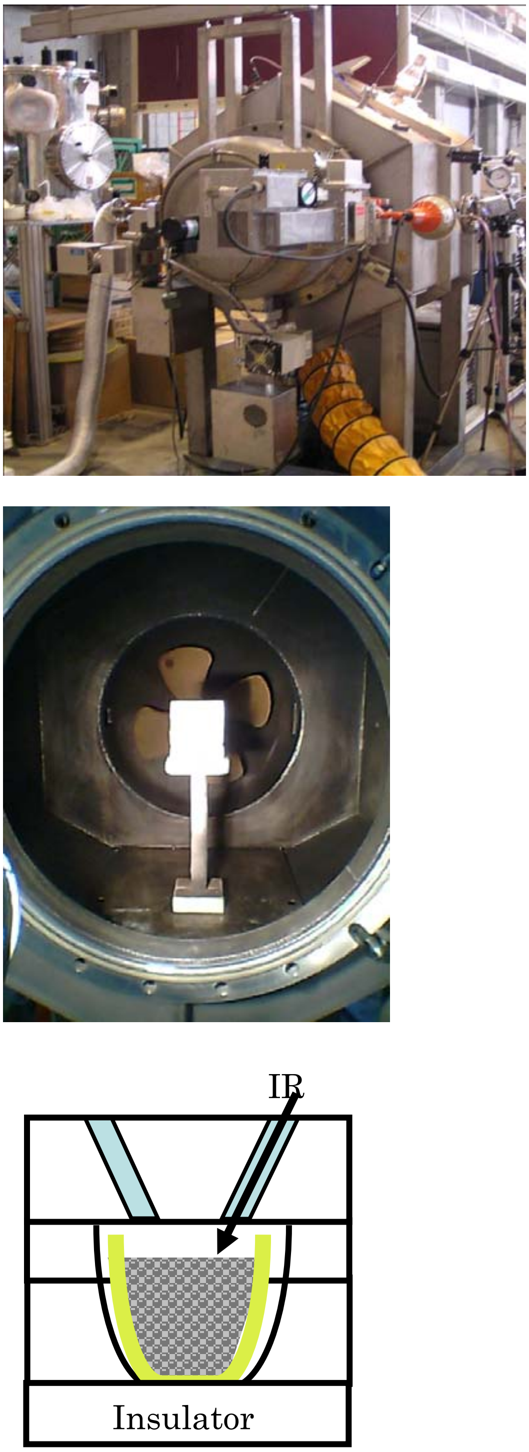

Fig.4. (a) 2.45GHz magnetron system (top), (b) Experimental setup of cm-wave process (middle), (c) Sketch of experimental setup (bottom).

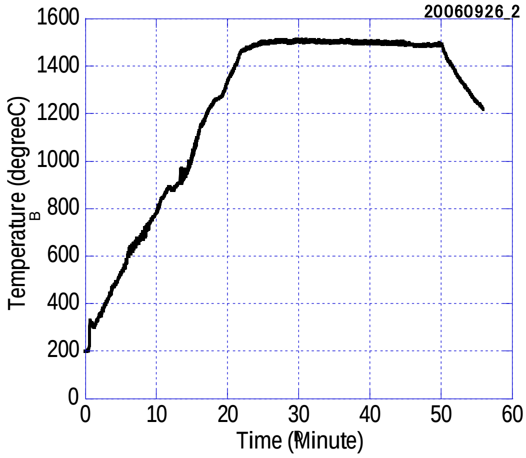

Fig.5 The measured temperatures of a cm-wave process.

Fig.6 Sample after 2.45GHz processing.

Figure 5 shows the process temperatures measured during cm-wave heating of a powder sample of mixed magnetite and carbon. After 15 minutes from start, strong radiation appeared due the ignition of a plasma at the sample surface [5]. The spectral intensity of the continuous spectrum is at least three orders of magnitude larger than that of black body emission for the temperature range of 860 °C – 1070 °C measured by the multi-point emission spectroscopic diagnostic conducted through the viewing port on the furnace. The pattern of the continuous spectrum is partly similar to a continuum of the emission light radiated by the free-band electron transition.

The origin for the continuous emission spectrum will be discussed. One candidate is a solid state fluorescence called cathodoluminescence induced by impingement of a plasma electron onto the sample surface of magnetite;

this results in the excitation of electrons from the valence band into the conduction band, and deexcitation with the broadband emission.

Figure 6, shows a picture of the material obtained by this process. The mainpart of the material was FeO as detected by XRD analysis.

4. CONCLUSIONS

High quality pig iron could be made from powder samples of mixed magnetite and carbon by 30 GHz mm-waves heating in air. However, in case of heating by 2.45 GHz at similar conditions, mainly FeO was produced. Therefore, we expect that there is frequency dependence in the reduction reaction.

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References

[1] R. Roy, R. Peelamedu, L. Hutt, J. Cheng, D.Agrawal, "Definitive experimental evidence for microwave effects: radical new effects of separated E and H fields", Mat. Res. Innovation **6**: 128-140, 2002.

[2] K. Nagata, R. Kojima, T. Murakami, M. Susa, H. Fukuyama, "Mechanisms of Pig-iron Making from Magnetic Ore Pellets Containing Coal at Low Temperature", ISIJ International **41**: 1316-1323, 2001.

[3] G. Link et al., "Sintering of advanced ceramics using a 30-GHz, 10-kW, cw industrial gyrotron"; IEEE Transactions on Plasma Science, 27 (2): 547-554, 1999.

[4] Feher L., Link G., DE-PS 19633245C1 (27.11.1997); Feher L.; Nuss V.; Seitz T.; Flach A. DE-PS 10329411 (31.1.2005).

[5] A. Matsubara, et al., submitted to Proc.11th International Conference on Microwave and High Frequency Heating, (3-6 Sep 2007, Oradea, Romania).