Microwave Permittivity, Permeability, and Absorption Capability of Ferric Oxide

Zhiwei PENG,1) Jiann-Yang HWANG,1,*) Chong-Lyuck PARK,1,2) Byoung-Gon KIM,2) Matthew ANDRIESE1) and Xinli WANG3)

1) Department of Materials Science and Engineering, Michigan Technological University, Houghton, MI, 49931 USA. E-mail: zpeng@mtu.edu, clpark@mtu.edu, mdandrie@mtu.edu  2) Mineral Processing Division, Korea Institute of Geoscience and Mineral Resources, Daedeon, 305-350 Korea. E-mail: bgkim@kigam.re.kr  3) School of Technology, Michigan Technological University, Houghton, MI, 49931 USA. E-mail: xinlwang@mtu.edu

(Received on December 31, 2011; accepted on March 30, 2012)

The permittivity and permeability measurements of ferric oxide (Fe₂O₃) were carried out over a broad temperature range from 24°C to above 1000°C at 915 and 2450 MHz. The real part and imaginary part of complex relative permittivity (ε' and ε'') of ferric oxide slightly increase with temperature below 450°C, above which ε' increases significantly while ε'' presents a broad dielectric loss peak between 450 and 1000°C. Contrary to ε' and ε'', the real part and imaginary part of complex relative permeability (μ' and μ'') remain relatively invariable (1 and 0, respectively) until 700°C. The μ'' values subsequently exhibit a decreasing tendency due to the increased electrical conductivity at higher temperatures while the μ'' values stay negligible as temperature increases. The results demonstrate that the dielectric loss is the primary factor contributing to microwave absorption of Fe₂O₃. The calculation of microwave penetration depth shows that Fe₂O₃ undergoes a transition from a microwave transparent material to a good microwave absorber with increasing temperature.

KEY WORDS: permittivity; permeability; microwave absorption; microwave penetration depth.

1. Introduction

The increasing applications of microwave energy in ironmaking and steelmaking in recent years urgently demand the characterizations of microwave absorption properties of raw materials such as iron oxides.1–3 This is because of microwave heating having strong dependences on the physical properties of materials, namely permittivity (ε) and permeability (μ). The relation between ε and μ and energy loss (P) in microwave heating due to the microwave-matter interaction can be illustrated by the following equation:

\[ P = \pi f \left( \varepsilon_0 \varepsilon_r \left| E \right|^2 + \mu_0 \mu_r \left| H \right|^2 \right) \]

where \( f \) is the microwave frequency (Hz), \( \varepsilon_0 \) and \( \mu_0 \) are the dielectric permittivity of free space (8.854×10⁻¹² F m⁻¹) and magnetic permeability of free space (4π×10⁻⁷ H m⁻¹), \( \varepsilon_r \) and \( \mu_r \) are the imaginary part of complex relative permittivity (dielectric loss factor) and imaginary part of complex relative permeability (magnetic loss factor), and \|E\| and \|H\| represent the electric and magnetic fields within the sample. As shown by Eq. (1), the energy loss in microwave heating is proportional to the dielectric loss factor and magnetic loss factor. Thus, the permittivity and permeability are of great significance for the energy conversion in materials subjected to microwave irradiation. Detailed characterizations of these properties are essential to advance the understanding of the microwave heating process.

Recently, due to realization of importance of the material properties in microwave assisted ironmaking, several scientists have investigated permittivity and permeability of iron oxides. Hotta et al. reported the permittivity and permeability of Fe₃O₄, Fe₂O₃, and Fe₁₋ₓO (0.05≤x≤0.15) powders in the microwave frequency range between 0.2 and 13.5 GHz and studied the temperature dependences of microwave permittivity and permeability of Fe₂O₃ up to 575°C using the coaxial transmission line method.4–7 Peng et al. investigated the effect of temperature on the microwave absorption capabilities of magnetite concentrate and ferrous oxide (Fe₃O₄·FeO·2O) covering a broad range from room temperature to above 1000°C through the permittivity and permeability measurements using the cavity perturbation technique.5,9) These studies showed that the investigated parameters closely depend upon microwave frequency and temperature. Meanwhile, the effect of temperature is comparatively stronger than that of frequency for iron oxides, especially above 500°C. This indicates that the high temperature permittivity and permeability are extremely important for the technology of microwave metallurgy and full investigations on these properties are crucial for the whole ironmaking process.

To date, there is little reported on the effect of temperature on the permittivity and permeability of ferric oxide (Fe₂O₃). Also, although the frequency dependences of these
parameters were studied, a comprehensive evaluation of microwave absorption capability of Fe₂O₃ based on the parameter measurements has not been reported. In response to these concerns, the present study was devoted to characterize the dielectric and magnetic properties of Fe₂O₃ from room temperature to above 1000°C by measuring the permittivity and permeability. A microwave absorption capability evaluation of Fe₂O₃ was then presented by determining the microwave penetration depths in the tested temperature range.

2. Experimental

Ferric oxide powders (99.98% purity) were supplied by Sigma-Aldrich Corp., St. Louis, MO. The chemical composition was confirmed by X-ray diffraction (XRD) using a conventional Scintag XDS2000 powder X-ray diffractometer (Scintag Inc., Cupertino, CA) with a graphite monochromator and Cu Kα radiation. Sample microstructure and particle distribution were characterized by using a Hitachi S-4700 field-emission scanning electron microscope (FE-SEM, Hitachi Ltd., Tokyo, Japan).

The permittivity and permeability of Fe₂O₃ sample were measured by the cavity perturbation technique. The main components of the measurement system include a resistive heating furnace and a cylindrical TM₀₅₀₀ resonant mode cavity. The system measures the differences (frequency shift and change of quality factors) in the microwave cavity response between a cavity with an empty sample-holder and the same cavity with a sample-holder plus the sample at each specified temperature. These differences are recorded in a Hewlett Packard 8753B vector network analyzer and then used to calculate the permittivity or permeability. The details about this technique and apparatus used for the measurements can be found in the published literature.

As for the permittivity test, Fe₂O₃ powders were first uniaxially pressed at ~207 MPa in a die lined with tungsten carbide to form pellets with a diameter of ~3.62 mm having a total stacked length (height) of 12.97 mm. The bulk density (room temperature) of the sample was 2.79 g cm⁻³. During the measurements, the sample was step-heated in the conventional resistance furnace to the designated temperatures in 0.01 L min⁻¹ flowing argon. The permittivity measurements started at room temperature (24°C) and heated in 50°C steps to ~1100°C. The variation of sample bulk density during the measurements was identified. The sample mass and dimensions were measured at the beginning and end of the dielectric properties measurement sequence. The mass and dimensions of the sample at each step during the dielectric measurement run were estimated by observing the time/temperature where the onset and end of significant changes in permittivity were occurring, and fitting a non-linear (power of 1.5) dimension reduction curve between the initial and final values of mass and dimensions. In the permeability measurements, the same punch/die unit was used to form pellets with a diameter of ~3.62 mm and total length of 13.38 mm. The bulk density (room temperature) of pellets was 2.80 g cm⁻³. The measurements were performed in the same argon, starting at room temperature, then in 50°C steps to ~1000°C.

It is widely known that microwaves are electromagnetic radiation having a broad frequency range of 0.3–300 GHz. However, to avoid interference with communication networks, all microwave heaters (domestic or scientific) are designed to work at either 915 or 2450 MHz. Due to this reason, this study focuses on the characterizations of the permittivity and permeability at these two frequencies.

3. Results and Discussion

3.1. Permittivity and Permeability of Fe₂O₃

Figure 1 shows the XRD pattern of ferric oxide. It is found that all of the marked peaks belong to the α phase, indicating the sample having rhombohedral structure. The morphology of Fe₂O₃ was characterized using FE-SEM, as presented in Fig. 2. It shows the sample powders having spherical shape with particle size between 0.05 and 0.2 μm. Figure 3 illustrates the temperature dependences of real
part \(\varepsilon'_r\) and imaginary part \(\varepsilon''_r\) of complex relative permittivity of Fe\(_2\)O\(_3\) at 915 and 2450 MHz. It is seen that \(\varepsilon'_r\) and \(\varepsilon''_r\) slightly increase with temperature up to 450°C. The examination and calibration of experimental data demonstrate that the dried Fe\(_2\)O\(_3\) has \(\varepsilon''_r\) values of about 0.014 and 0.012 at 24°C for 915 and 2450 MHz, respectively. It suggests that Fe\(_2\)O\(_3\) possesses a very low dielectric loss at room temperature.

Inspection of Fig. 3 shows that \(\varepsilon''_r\) and \(\varepsilon''_r\) vary similarly below 450°C. As temperature increases, however, they begin to show different variation behaviors. The \(\varepsilon'_r\) value rapidly increases with temperature beyond 450°C while the \(\varepsilon''_r\) values show a broad dielectric loss peak between 450 and 1000°C. This is known as a typical relaxation/interfacial polarization phenomena behavior, usually indicating a change in the material associated with the loss of an insulating barrier between particles, or the presence of a transient species during a phase change. Such observation is associated with microstructure change due to sintering occurred at high temperatures, which can be demonstrated by the variation of sample bulk density during the measurements, as depicted in Fig. 4.

Figure 4 shows that the sample bulk density \((\rho)\) remains constant until 800°C and then increases proportionally with temperature. This suggests that sintering starts at 800°C and becomes more evident at higher temperatures. Sintering makes the sample particles adhere together, resulting in a strong surface densification with the possibility of grain growth during extended heating time. The increase in the average grain size, as shown in Fig. 5, may hinder the diffusion of iron and oxygen ions, decreasing dielectric polarization. This effect is found to be more obvious above 1000°C. However, it should be emphasized that the measured dielectric loss of material comprises of dipole/ion contribution \((\varepsilon''_r)\) and conductivity \((\sigma)\) contribution, as given by the following equation:

\[
\varepsilon''_r = \varepsilon''_{rd} + \frac{\sigma}{2\pi f \varepsilon_0} \tag{2}
\]

Thus, one can expect that a possible increase in electrical conductivity due to thermal activation at high temperatures may overwhelm the adverse effect of sintering, leading to a high dielectric loss. This possibility can be confirmed by the fact that electrical conductivity of Fe\(_2\)O\(_3\) increases with temperature, as expressed by the formula:

\[
\sigma = 2.7 \times 10^8 e^{-(0.96/\theta)} \tag{3}
\]

where \(\sigma\) is the electrical conductivity (S m\(^{-1}\)), \(k\) is the Boltzmann constant \((1.38 \times 10^{-23} \text{ m}^2 \text{ kg} \text{ s}^{-2} \text{ K}^{-1})\) and \(\theta\) is the temperature (K).

Figure 6 presents the permeability change in the temperature range between 24 and 1000°C. It reveals that both the real part \((\mu'_r)\) and imaginary part \((\mu''_r)\) of complex relative permeability keep relatively constant below 700°C. Their values vary around 1 and 0, respectively. It is also observed that there are small variations of permeability \((\pm 0.05)\) in the temperature range, resulting from the statistical error in the measurements. These results are consistent with the fact that Fe\(_2\)O\(_3\) becomes weakly ferromagnetic above the Morin transition at \(-13°C\) and below its Neel temperature at 675°C. Continuous heating to higher temperatures gives rise to different variation behaviors of \(\mu'_r\) and \(\mu''_r\). The \(\mu'_r\) value decreases with increasing temperature beyond 700°C, which can be attributed to the increased electrical conductivity. As discussed before, the conductivity of Fe\(_2\)O\(_3\) increases with temperature, making the sample more “conductive”. According to Lenz’s law, a time-varying magnetic field \((e.g.\: \text{microwave})\) induces large currents near the surface of a good conductor, producing a magnetic field opposite to the external magnetic field, leading to zero magnetic field inside. This means that a substance has an “effective magnetic susceptibility” of \(-1\), and thus appears to have a permeability value of \(\mu'\approx 0\) when it exhibits high conductivity. Therefore, in the present case, the \(\mu'_r\) values of Fe\(_2\)O\(_3\) show a decreasing tendency as temperature increases. Contrary to \(\mu'_r\), it is noticed that the \(\mu''_r\) values stay negligible.
at temperatures higher than 700°C. This is because Fe₂O₃ exhibits paramagnetism above its Néel temperature.²

From above results, it can be inferred that Fe₂O₃ does not show noticeable magnetic response at 915 and 2450 MHz below 1000°C. The dielectric loss is the primary factor contributing to the microwave absorption of Fe₂O₃ and the contribution of magnetic loss to microwave heating of Fe₂O₃ can be ignored.

3.2. Microwave Absorption Capability of Fe₂O₃

As aforementioned, microwave energy loss in materials relies on the permittivity and permeability. This suggests that a reasonable evaluation of microwave absorption capability of Fe₂O₃ has to take the combined effect of dielectric and magnetic properties. According to the previous studies, a quick evaluation of microwave absorption properties of materials can be achieved by determining the microwave penetration depth \( D_p \).²,⁵ The equation for \( D_p \) calculation is given as:⁸

\[
D_p = \frac{\lambda_0}{2\pi} \left( \varepsilon_r' - \varepsilon_r'' \mu_r' - \mu_r'' \right)^{1/2} \left( \varepsilon_r'' \mu_r' - \varepsilon_r' \mu_r'' \right)^{1/2} \]  

(4)

where \( \lambda_0 \) is the microwave wavelength in free space (m).

Considering the negligible microwave magnetic loss of Fe₂O₃ (\( \mu_r' \approx 1 \) and \( \mu_r'' = 0 \)) in the tested temperature range, calculation of \( D_p \) is determined by the variation of permittivity.

**Figure 7** shows the variation of \( D_p \) with temperature. It is seen that Fe₂O₃ has large \( D_p \) (10.05 and 4.36 m at 915 and 2450 MHz, respectively) at room temperature, indicating a very slow microwave dissipation inside the oxide. This is due to the small permittivity and permeability of Fe₂O₃ at low temperatures. As temperature increases, the \( D_p \) value decreases rapidly mainly because of a significant increase in permittivity. Since the relative permittivity is much larger than the relative permeability at high temperatures, the permittivity dominates the variation of microwave penetration depth. Figure 7 indicates \( D_p \) having values less than 0.045 m for both frequencies beyond the temperature of 600°C. This shows that ferric oxide undergoes a transition from a microwave transparent material to a good microwave absorber with increasing temperature.

### 4. Conclusions

To characterize microwave absorption in ferric oxide, the permittivity and permeability measurements from 24°C to above 1000°C and calculation of microwave penetration depth were performed. The following conclusions can be drawn from the obtained results:

(1) The \( \varepsilon_r' \) and \( \varepsilon_r'' \) values of ferric oxide slightly increase with temperature below 450°C. Continuous heating to higher temperatures leads to a rapid increase in the \( \varepsilon_r' \) value and a broad dielectric loss peak between 450 and 1000°C formed by the \( \varepsilon_r'' \) values. The increase in permittivity at high temperatures is closely associated with sintering effect and the variation of electrical conductivity.

(2) The complex relative permeability is found to be relatively independent of frequency and temperature below 700°C. The \( \mu_r' \) and \( \mu_r'' \) values remain relatively invariable (1 and 0, respectively) in this temperature range, which are consistent with the fact that Fe₂O₃ becomes weakly ferromagnetic above the Morin transition at ~13°C and below its Néel temperature at 675°C. As temperature continues to rise, the \( \mu_r'' \) values present a decreasing tendency due to the increased electrical conductivity at higher temperatures. The \( \mu_r'' \) values, however, stay negligible due to the paramagnetism presented by Fe₂O₃ above its Néel temperature.

(3) The significant difference between the variation behaviors of permittivity and permeability of Fe₂O₃ indicates that the dielectric loss is the primary factor contributing to the microwave absorption of Fe₂O₃ and the contribution of magnetic loss to microwave heating can be ignored. The decrease in microwave penetration depth of Fe₂O₃ with increasing temperature suggests that Fe₂O₃ undergoes a transition from a microwave transparent material to a good microwave absorber as temperature increases.

**Acknowledgements**

The authors are indebted to Dr. Ron Hutcheon of the Microwave Properties North for helpful discussions. This work was supported by the Korea Institute of Geoscience and Mineral Resources, the Michigan Public Service Commission, U.P. Steel, and the United States Department of Energy under Award No. DE-FC36-01ID14209.

**REFERENCES**