Complex Permittivity of Graphite, Carbon Black and Coal Powders in the Ranges of X-band Frequencies (8.2 to 12.4 GHz) and between 1 and 10 GHz

Masahiro HOTTA,1) Miyuki HAYASHI,2) Michael Thomas LANAGAN,3) Dinesh Kumar AGRAWAL4) and Kazuhiro NAGATA5)

1) Department of Chemistry and Materials Science, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo, 152-8552 Japan. E-mail: hotta.m.ab@m.titech.ac.jp 2) Department of Metallurgy and Ceramics Science, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo, 152-8552 Japan. 3) Center for Dielectric Studies, Materials Research Institute, The Pennsylvania State University, University Park, PA 16802, USA. 4) Microwave Processing and Engineering Center, Materials Research Institute, The Pennsylvania State University, University Park, PA 16802, USA. 5) Department of Conservation Science, Tokyo University of the Arts, 12-8 Ueno Kouen, Taito-ku, Tokyo, 110-8714 Japan.

(Received on April 4, 2011; accepted on August 10, 2011)

The real and imaginary parts of the relative complex permittivity (\(\varepsilon'_r\) and \(\varepsilon''_r\)) were measured in the ranges of X-band frequencies (8.2 to 12.4 GHz) and between 1 and 10 GHz for graphite, carbon black and coal powders at room temperature so as to clarify the relation between the complex permittivities of carbonaceous materials and their characteristics, i.e., graphitization, porosity (i.e., specific surface area) and ash contained in coals. It is found that the complex permittivities increase with increasing crystallite size and specific surface area. It is also found that the dependency of the permittivities on the ash content seems be negligible within the range of the ash content in the present study.

KEY WORDS: permittivity; carbonaceous materials; specific surface area; graphitization; purity; microwave frequency.

1. Introduction

Microwave is an electromagnetic wave, the frequency range of which is from 0.3 to 300 GHz (corresponding to wavelengths ranging from 1 mm to 1 m). Microwave could be applied to a new heating source for iron making to reduce the CO2 emission and energy consumption.1–4) In order to improve the energy efficiency of iron making by microwave heating, it is important to know the microwave absorptivity of raw materials and the relation between the absorptivity and the material characteristic.

Microwave absorption can be described by the following equation.

\[ P = \frac{1}{2} \sigma |E|^2 + \pi f \varepsilon''_r |E|^2 + \pi f \mu''_r |H|^2 \] .......(1)

Where \(|E|\) electric field amplitude, \(|H|\) magnetic field amplitude, \(\sigma\) electrical conductivity, \(f\) microwave frequency, \(\varepsilon''\) imaginary part of relative permittivity, \(\mu''\) imaginary part of relative permeability. The first, the second and the third terms of the right hand side of Eq. (1) correspond to the microwave absorptions due to the Joule loss, the dielectric loss and the magnetic loss, respectively. Among raw materials of iron making, i.e., iron oxides and carbonaceous materials, the authors previously focused on the microwave absorption of iron oxide powders.

We have measured the relative complex permittivities of \(\alpha\)-Fe2O3, Fe3O4 and Fe1–xO powders over the frequency range between 0.2 and 13.5 GHz using a coaxial transmission line method.5) It has been found that the magnitudes of the real and imaginary parts of relative permittivity are in the hierarchy Fe1–xO > Fe3O4 > \(\alpha\)-Fe2O3 for the powders having an identical relative density over the measurement frequencies. We have also measured the relative complex permittivity and permeability of Fe3O4 powders with various particle sizes and have found that the imaginary parts of relative permeability of Fe3O4 powders with a particle size of 50 nm–180 \(\mu\)m show the peaks in the frequency range of 0.706 GHz to 2.99 GHz, depending on the particle size.6) It has also been found that the imaginary parts of relative permittivity of Fe3O4 powders are quite large having a peak around 10 GHz.

We have carried out the measurements not only at room temperature but also at elevated temperatures. We have successfully measured the relative complex permittivity and permeability of Fe3O4 powders over the temperature range of 25–575°C and have proposed a heating mechanism of Fe3O4 powders; Fe3O4 powders can be heated by the combination of the magnetic loss at lower temperatures and the Joule loss at higher temperatures.7)

On the other hand, carbonaceous materials are good absorbers of microwave and could be main heating sources for carbothermic reductions of iron oxides by microwave heating. Therefore, it is necessary to measure the complex
permittivities of carbonaceous materials to estimate their performance as a heating source. The aim of this study is to measure the complex permittivities of graphite, carbon black and coal powders at room temperature so as to clarify the relation between the complex permittivities of carbonaceous materials and their characteristics, i.e., graphitization, porosity (i.e., specific surface area) and ash contained in coals. These three factors are considered to predominantly affect the reactivity, i.e., the reducing power of carbonaceous material. The reactivity is one of the most important properties among high temperature properties of carbonaceous materials for iron making as it controls the overall reduction rate of iron ores. The degree of graphitization controls the chemical reaction rate. It is generally known that carbonaceous materials with higher degree of graphitization show lower reactivity. Specific surface area of carbonaceous material controls the gas diffusion rate. The reactivity of carbonaceous materials would increase with an increase in the specific surface area. Ash contained in coals may also affect the reactivity since alkalis contained in ash catalyze the gasification reaction.

2. Experimental

2.1. Sample

Three graphite (AT-No.5, artificial graphite and pure carbon), one carbon black and five coal (coals A–E) powder samples were used. Table 1 summarizes the purities and the particle sizes reported by the suppliers for the samples employed in this study. Here, the purities of coal samples were measured by the authors using the X-ray fluorescence analysis (XRF). Table 2 shows the chemical compositions of the coal samples measured by the XRF analysis.

The shapes of particles were observed using the scanning electron microscope (SEM). With respect to the sample preparation for the SEM observation, a sample was submerged in ethanol and the ethanol containing powder was subjected to an ultrasonic bath treatment. After that, a drop of the ethanol was placed on a glass plate and dried in air. Figure 1 shows the SEM images of some samples. Figures 1(a) through 1(d) and Fig. 1(f) show the SEM images of AT-No. 5, artificial graphite, pure carbon, carbon black and coal B, respectively. It can be found that the observed particle sizes are in good agreement with the nominal particle sizes.

### Table 1. Purities, particle sizes and crystallite sizes of the samples employed in this study.

| sample name | purity, % | particle size, μm | Le, Å |
|-------------|-----------|------------------|------|
| AT-No.5     | 99        | 40               | 372.0|
| artificial graphite | 99         | 80               | 577.4|
| pure carbon | 99.9      | 20               | 340.9|
| carbon black | guaranteed reagent grade | 20          | 10.6 |
| coal A      | 96.84     | 80               | 41.9 |
| coal B      | 95.99     | 80               | 15.3 |
| coal C      | 95.97     | 80               | 18.7 |
| coal D      | 94.75     | 80               | 29.1 |
| coal E      | 92.77     | 80               | 28.0 |

### Table 2. Chemical compositions of the coal samples measured by the XRF analysis (mass%).

|     | C    | S    | Si   | Al   | Fe   | Ni   | Ca   | Mg   | Ti   | K    | V    | P    | Cr   | Sr   |
|-----|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| coal A | 96.84 | 0.58 | 1.64 | 0.73 | 0.05 | 0.04 | 0.03 | 0.06 | 0.02 | 0.00 | 0.01 |
| coal B | 95.99 | 1.14 | 1.20 | 0.93 | 0.35 | 0.30 | 0.06 | 0.08 | 0.05 |
| coal C | 95.97 | 1.05 | 1.16 | 0.94 | 0.31 | 0.55 | 0.06 | 0.04 |
| coal D | 94.75 | 0.80 | 1.95 | 1.45 | 0.65 | 0.13 | 0.07 | 0.09 | 0.09 | 0.04 | 0.01 | 0.01 |
| coal E | 92.77 | 6.96 | 0.06 | 0.04 | 0.02 | 0.04 | 0.00 | 0.01 |

Fig. 1. SEM image of powder samples. (a) AT-No.5, (b) artificial graphite, (c) pure carbon, (d) carbon black (primary particle), (e) carbon black (secondary particle), and (f) coal-B.
in Table 1 except for carbon black (d). The particle size of carbon black is observed to be around 0.2 μm, which is much smaller than the nominal particle size of 20 μm. In fact, the average particle size of the carbon black powders without an ultrasonic bath treatment is around 20 μm, as shown in Fig. 1(e). Therefore, it is considered that a carbon black particle is an agglomeration of much smaller particles.

Prior to the permittivity measurement, all powder samples were dried at 373 K for 72 hours to remove the adsorbed moisture.

2.2. Permittivity Measurement

The real and imaginary parts (ε′ and ε″) of relative permittivity were determined from transmitted electromagnetic wave in the ranges of X-band frequencies (8.2 to 12.4 GHz) and between 1 and 10 GHz. As for the measurement in the X-band frequency range, powder samples were filled in the cross section of the rectangular waveguide using transparent tape, as shown in Fig. 2. The ports of the network analyzer (Hewlett-Packard 8510T) were attached to a coaxial cable, and the sample holder was adjoined to a coax-waveguide adapter (see Fig. 3(a)). Determination of the transmitted electromagnetic wave by this system enables us to obtain the ε′ and ε″ values averaged over the frequency range between 8.2 and 12.4 GHz. As for the measurement over the frequency range between 1 and 10 GHz, a 7 mm coaxial sample holder (sample holder length is 100 mm) was configured vertically, as shown in Fig. 3(b).

Material in sample holders alters the magnitude and phase of the transmitted microwave, which are observed by the network analyzer. The complex transmission data were transferred to a computer for dielectric property calculation. The measurement technique has been described in elsewhere.11)

Graphite and carbon black powders were measured both in the ranges of X-band frequencies and between 1 and 10 GHz with the coaxial line. However, coal samples were only measured in the X-band frequency range because, as it will be described later, the ε′ values of coal powders are very low and independent of frequency.

2.3. Crystallinity and Specific Surface Area Measurements

X-ray diffraction measurements were carried out for graphite, carbon black and coal powder samples to determine the average crystal size along the structural c-axis, Lc. The Lc values were evaluated from the full widths at half maximum of X-ray diffraction peak intensities of the (002) reflection using the Scherrer equation.12) These crystallite

![Sample holder assembly for rectangular waveguide measurement.](image)

![Schematic of the network analyzer system with a X-band rectangular waveguide sample holder (a), and a coaxial type sample holder (b).](image)

![Frequency dependency of relative complex permittivity (ε′ and ε″) for the graphite and carbon black samples: (a) AT-No.5, (b) artificial graphite, (c) pure carbon, and (d) carbon black.](image)
sizes are generally used as a measure of the degree of graph-
itization.\textsuperscript{12,13}) The specific surface area was determined by the gas adsorption method (Micromeritics Instrument Corp., TriStar3000) using the N\textsubscript{2} gas at 77 K.

3. Results

Figures 4(a) through 4(d) show the frequency dependen-
tcies of the relative complex permittivity of (a) AT-No.5, (b) artificial graphite, (c) pure carbon, and (d) carbon black in the range of X-band frequencies (8.2 to 12.4 GHz) (broken lines) and between 1 and 10 GHz (solid lines). The measurements in the ranges of X-band frequencies and between 1 and 10 GHz were carried out for five and three different relative densities, respectively. The relative densities of powdery samples for all the measurements are included in Fig. 4, which were obtained by dividing the apparent density by the bulk density (graphite: 2.25 g/cm\textsuperscript{3},\textsuperscript{1,4} carbon black: 2.1 g/cm\textsuperscript{3}).\textsuperscript{1,4} For each relative density, three to five runs were carried out to confirm the reproducibility. The error bars represent the standard deviation of the experimental values. The \( \varepsilon' \) values of all the samples in Fig. 4 monotonically decrease with increasing frequencies to and approach a constant value above 6.5 GHz. The \( \varepsilon'' \) values also monotonically decrease with increasing frequency in the similar fashion as the \( \varepsilon' \) values.

Table 3 shows the relative complex permittivity in the X-
band frequency range for five coal samples. The measure-
ments were carried out for five relative densities, and for
each relative density, three to five runs were conducted. The relative densities, \( \rho_r \), were obtained by dividing the apparent density by the bulk density, 1.3 g/cm\textsuperscript{3}.\textsuperscript{1,5,16} It is found that the \( \varepsilon' \) values of coal samples are much smaller than those of graphite and carbon black samples.

4. Discussion

4.1. Mechanisms of Polarization

Polarization mechanisms are the mechanisms with which
atoms and molecules respond to an electric field. Figure 5
shows a schematic illustration of frequency dependency of real and imaginary parts of permittivity. There are essentially four basic mechanisms which contribute to the permittivity:\textsuperscript{17} (i) Space charge polarization (Interfacial polarization) characterized by a space charge polarizability, \( P_s \); this polarization occurs in a dielectric material containing mobile charge carriers. In an applied electric field, charge carriers migrate and are impeded in their motion because they become trapped at surfaces, grain boundaries, and so on, resulting in the dielectric permittivity. (ii) Dipolar polarization characterized by a dipolar polarizability, \( P_d \); this arises from molecules with a permanent dipole moment that can change orientation in an applied electric field. (iii) Atomic polarization characterized by an atomic polarizability, \( P_a \); this arises from the displacement of electron clouds with respect to atomic nuclei. The net polarizability, \( P \), of the dielectric material consists of the four terms.

\[
P = P_s + P_d + P_a + P_e \tag{2}
\]

In general, the resonant frequencies of electronic and atomic polarization are at ultraviolet or visible light region and at infrared or far infrared region, respectively. As micro-
wave frequencies are far below the resonant frequencies of electronic and atomic polarizations, the permittivity due to

| Table 3. Relative complex permittivity of coal A–E. |
|------------------|--------|--------|--------|
| sample          | relative density | \( \varepsilon' \) | \( \varepsilon'' \) |
| coal A          | 0.37    | 2.08   | 0.25   |
| coal A          | 0.39    | 2.15   | 0.36   |
| coal A          | 0.46    | 2.28   | 0.11   |
| coal A          | 0.48    | 2.21   | 0.20   |
| coal A          | 0.52    | 2.21   | 0.29   |
| coal B          | 0.36    | 2.18   | 0.29   |
| coal B          | 0.37    | 2.13   | 0.33   |
| coal B          | 0.44    | 2.10   | 0.32   |
| coal B          | 0.48    | 2.03   | 0.29   |
| coal B          | 0.49    | 2.12   | 0.35   |
| coal C          | 0.44    | 2.16   | 0.34   |
| coal C          | 0.46    | 2.01   | 0.59   |
| coal C          | 0.54    | 1.97   | 0.21   |
| coal C          | 0.57    | 2.02   | 0.24   |
| coal C          | 0.61    | 2.08   | 0.52   |
| coal D          | 0.45    | 1.90   | 0.18   |
| coal D          | 0.46    | 1.90   | 0.30   |
| coal D          | 0.46    | 1.94   | 0.28   |
| coal D          | 0.51    | 1.91   | 0.32   |
| coal D          | 0.54    | 1.94   | 0.40   |
| coal E          | 0.44    | 2.20   | 0.36   |
| coal E          | 0.45    | 2.01   | 0.09   |
| coal E          | 0.45    | 2.07   | 0.27   |
| coal E          | 0.46    | 2.21   | 0.04   |
| coal E          | 0.49    | 2.05   | 0.15   |

Fig. 5. Schematic illustration of frequency dependency of real and imaginary parts of permittivity.
these polarizations is almost independent of the frequency. In addition, their contributions to the real and imaginary permittivity magnitudes are small in comparison to space charge and dipolar polarizations. Therefore, the frequency dependency of permittivities of the present samples are attributed to the space charge polarization as carbonaceous materials do not have permanent dipoles. In fact, it is widely known that in microwave susceptible solids heat is produced via space-charge polarization.\textsuperscript{18–25}

Atwater and Wheeler, Jr.\textsuperscript{26–28} have measured the complex permittivities of activated carbon powders and graphitized carbon black powders over the frequency range between 0.2 and 26 GHz, and have found that the permittivities show the relaxation type frequency dependencies and the relation between the $\varepsilon_r'$ and $\varepsilon_r''$ values obeys the Cole-Cole model. They have also found that the frequencies of $\varepsilon_r''$ maxima range from 2 to 6.8 GHz depending on the samples. For the present samples, the $\varepsilon_r''$ values do not show any peaks but monotonically decrease with increasing frequencies within the measurement frequency range, which indicates that the $\varepsilon_r''$ values could have peaks below 1 GHz. The peak frequencies may be dependent on the material characteristics such as particle size and conductivity. The peak frequencies may also be affected by the contact conditions of powders although the relative densities of the samples have not been reported in the Atwater and Wheeler, Jr.’s papers.

4.2. Relation between Relative Density and Complex Permittivity

Figure 6 shows the $\varepsilon_r'$ and $\varepsilon_r''$ values of the graphite and carbon black as a function of relative density. The closed symbols denote the data measured in the X-band frequency range (8.2 to 12.4 GHz) using a rectangular waveguide sample holder, and the open symbols represent the average values of the data over the frequency range of 9–10 GHz measured using a coaxial sample holder. The error bars represent the standard deviation of the experimental values. As can be seen in Fig. 6, the $\varepsilon_r'$ values increase with increasing the relative densities, $d_r$. Scattering of the data may be caused by the measurement error in the relative densities as well as in the $\varepsilon_r'$ and $\varepsilon_r''$ values. In particular, the data from the coaxial sample holder (open symbols) show large errors. This is likely to have been caused by difficulties in measurements of the height of the powder in the coaxial line, resulting in erroneous densities. Therefore, the dependencies of the complex permittivities on the relative densities were obtained only using the data measured in the X-band frequency range (closed symbols). Although we have reported that the $\varepsilon_r'$ versus $d_r$ curve for Fe$_3$O$_4$ powders is convex downward,\textsuperscript{6} in the present study, the dependencies of the permittivities on the $d_r$ values are fitted to linear equations using a least-square method.

Figure 7 shows the $\varepsilon_r'$ and $\varepsilon_r''$ values of coal powders. Since the values are largely scattered, it is hard to determine the dependencies of the permittivities on the $d_r$ values. However, in the same manner as Fig. 6, the dependencies of the permittivities on the $d_r$ values are assumed to be expressed as the straight lines passing through the permittivity of vacuum (i.e., $\varepsilon_r'=1$, $\varepsilon_r''=0$) at $d_r=0$.

In the following sections, the relation between the complex permittivities of carbonaceous materials and their characteristics, i.e., ash contents in coals, graphitization (i.e., crystallization) and porosity (i.e., specific surface area) will be discussed. To compare the $\varepsilon_r'$ and $\varepsilon_r''$ values with the ash contents in coal samples, crystallite sizes and specific surface area, the $\varepsilon_r'$ and $\varepsilon_r''$ values at $d_r=0.25$ (or $d_r=0.45$) are either interpolated or extrapolated by the fitted linear equations, and are subjected to the comparisons.

4.3. Effect of Ash Content in Coal on Complex Permittivity

Figure 8 shows the relations between the ash contents and the $\varepsilon_r'$ and $\varepsilon_r''$ values at $d_r=0.45$. Within the range of the ash content in the present study, the dependency of the
The $\varepsilon'$ and $\varepsilon''$ values on the ash content seems to be negligible. Therefore, in the following sections the $\varepsilon'$ and $\varepsilon''$ values of coal are containing ash.

4.4. Effect of Crystallite Size on Complex Permittivity

Figure 9 shows the relations between the crystallite sizes $L_c$ and the $\varepsilon'$ and $\varepsilon''$ values at $d_r = 0.25$. It is seen that except for the data of carbon black, the complex permittivities increase with increasing the $L_c$ values. The reason that the $\varepsilon''$ values increase with increasing the degree of graphitization, i.e., the $L_c$ values can be explained as follows: It is known that the electrical conductivities of carbonaceous materials increase with an increase in the degree of graphitization (crystallization). Electrical conductivity $\sigma$ is relevant to the $\varepsilon''$ value by the equation $\sigma = \omega \varepsilon_0 \varepsilon''$, where $\varepsilon_0$ is permittivity of vacuum and $\omega$ is angular frequency. Therefore, the $\varepsilon''$ values of carbonaceous materials should increase with increasing the degree of graphitization. In fact, Nishioka and coauthors have heated carbon samples having various degrees of graphitization by microwave, and have found that the temperature increasing rate is faster, that is, the microwave absorption is larger for the sample with a larger degree of graphitization. This also supports our results that the $\varepsilon''$ values increase with increasing the $L_c$ values.

On the other hand, the $\varepsilon'$ and $\varepsilon''$ values of carbon black deviate much upward from the relations between the complex permittivities and the $L_c$ values obtained from the residual samples. Carbon black has a smaller $L_c$ value, and in fact, much smaller electrical conductivity although the $\varepsilon'$ and $\varepsilon''$ values are larger. The reason will be discussed in the next subsection.

4.5. Effect of Specific Surface Area on Complex Permittivity

As described in the Introduction section, porosity, i.e., specific surface area may be relevant to the reactivity, i.e., the reducing power of carbonaceous materials. Therefore, it is important to elucidate the relation between the microwave absorptivity and the specific surface area of carbonaceous materials. Figure 10 shows the $\varepsilon'$ and $\varepsilon''$ values at $d_r = 0.25$ as a function of specific surface area. It is found that the complex permittivities increase with increasing the specific surface area except for the $\varepsilon'$ value of artificial graphite. This results may be explained as follows: Magnitudes of the $\varepsilon'$ and $\varepsilon''$ values in the X-band frequency range might be associated with the space charge polarization. The space charge polarization, as aforementioned, takes place by impeding charge carriers migrated in an applied electric field.
field. Namely, for the occurrence of the space charge polarization, heterogeneities of electrical properties in the samples, i.e., the mixtures of conductive and less-conductive parts are necessary.\textsuperscript{32) Charge carries of carbonaceous materials are electrons and holes, the number densities of which are nearly identical.}\textsuperscript{29} In the present samples, the contact electrical resistance between particles may play the role of the less-conductive parts; electrons and holes are impeded at interfaces between particles, yielding the space charge polarization. This possibility may be the reason that the $\varepsilon_r'$ and $\varepsilon_r''$ values increase with increasing the specific surface area.

In the former subsection, we have described that the $\varepsilon_r'$ and $\varepsilon_r''$ values of carbon back are much larger than the values estimated from the relations between the complex permittivities and the $L_s$ values deduced by the residual samples. In fact, the specific surface area of carbon black is the largest among those of all samples. The large space charge polarizability of carbon black due to the large specific surface area could lead to the deviation of the $\varepsilon_r'$ and $\varepsilon_r''$ values of carbon back from the relation between the crystallite sizes, $L_s$, and the $\varepsilon_r'$ and $\varepsilon_r''$ values, as shown in Fig. 9. Similarly, the deviation of the $\varepsilon_r'$ value of artificial graphite in Fig. 10 can be associated with its large $L_s$ value. As a consequence, it can be concluded that both crystallite size and specific surface area affect the complex permittivities of carbonaceous materials.

5. Conclusions

The real and imaginary parts of the relative complex permittivity ($\varepsilon_r'$ and $\varepsilon_r''$) were measured for graphite, carbon black and coal powders over the frequency ranges in the X-band frequency (8.2 to 12.4 GHz) and between 1 to 10 GHz using a rectangular waveguide sample holder and a coaxial type sample holder, respectively. The effects of microwave frequency and material characteristics, i.e., graphitization, specific surface area and ash content in coals on the complex permittivities were investigated. The obtained results are summarized as follows.

(1) The $\varepsilon_r'$ values of graphite and carbon black powders monotonically decrease with increasing frequencies to become fairly constant above 6.5 GHz. The $\varepsilon_r''$ values also monotonically decrease with increasing frequency in the similar fashion as the $\varepsilon_r'$ values. It is found that the $\varepsilon_r'$ values of coal samples are much smaller than those of graphite and carbon black samples.

(2) The complex permittivities increase with increasing crystallite size and specific surface area. It is also found that the dependency of the permittivities on the ash content seems to be negligible within the range of the ash content in the present study.

Acknowledgements

The authors would like to thank Mr. Steve Perini for his assistance in the microwave characterization.

REFERENCES

1) K. Nagata, M. Hayashi, M. Sato, K. Ishizaki, T. Hayashi, A. Matsubara, S. Takayama and O. Motojima: Proc. of 4th Int. Cong. on the Science and Tech. of Steelmaking (ICS2008), ISIJ, Tokyo, (2008), 638.
2) K. Ishizaki, K. Nagata and T. Hayashi: ISIJ Int., 46 (2006), 1403.
3) K. Ishizaki and K. Nagata: ISIJ Int., 47 (2007), 811.
4) K. Ishizaki, K. Nagata and T. Hayashi: ISIJ Int., 47 (2007), 817.
5) M. Hotta, M. Hayashi and K. Nagata: ISIJ Int., 50 (2010), 1514.
6) M. Hotta, M. Hayashi, A. Nishikata and K. Nagata: ISIJ Int., 49 (2009), 1443.
7) M. Hotta, M. Hayashi and K. Nagata: ISIJ Int., 51 (2011), 491.
8) K. Uebo, H. Kano, T. Chikata and Y. Kashiwagi: in 4th European Coke and Ironmaking Congress Proc., Paris, Vol. 1, ATS, Paris, (2000) 174.
9) K. Kobayashi: The Introduction of Carbon Materials, Soc. Carbon Mater., Tokyo, (1972), 12.
10) Y. Kashiwagi, M. Takahata, K. Ishii, K. Yamaguchi, M. Naito and H. Hasegawa: Tetsu-to-Hagané, 87 (2001), 259.
11) M. T. Lanagan, J. H. Kim, D. C. Dube, S. J. Jang and R. E. Newnham: Ferroelectrics, 82 (1988), 91.
12) J. V. Dubrawski and W. W. Gill: Ironmaking Steelmaking, 11 (1984), 7.
13) Y. Kashiwagi and K. Ishii: ISIJ Int., 31 (1991), 440.
14) Rika Nenpyo (Chronological Scientific Tables), Vol. 69, ed. by National Astronomical Observatory, Maruzen Co., Ltd., Tokyo, (1995), 438.
15) W. V. Kotlensky and P. L. Walker, Jr.: Proc. 4th Conf. on Carbon, Peragamon Press, Oxford, (1960), 432.
16) D. W. van Krevelen and H. A. G. Chemin: Fuel, 33 (1954), 79.
17) A. V. Hippel: Dielectric materials and applications, Technology Press of MIT and Wiley & Sons, New York, (1954), 19.
18) P. Debye: Polar Molecules, Lancaster Press, Lancaster, Pennsylvania, (1929).
19) K. S. Cole and R. H. Cole: J. Chem. Phys., 9 (1941), 341.
20) D. W. Davidson and R. H. Cole: J. Chem. Phys., 19 (1951), 1484.
21) T. Ohgushi and K. Isimaru: Phys. Chem. Chem. Phys., 3 (2001), 3299.
22) C. J. F. Botcher: Theory of Electric Polarization, Elsevier, New York, (1952).
23) S. Havriliak, Jr. and J. S. Havriliiak: Dielectric and Mechanical Relaxation in Materials, Hanser, New York, (1997).
24) V. I. Gaiduk: Dielectric Relaxation and Dynamics of Polar Molecules, World Scientific, New Jersey, (1999).
25) A. K. Jonscher: J. Phys. D: Appl. Phys., 32 (1999), R57–R70.
26) J. E. Atwater and R. R. Wheeler, Jr.: J. Mater. Sci., 39 (2004), 151.
27) J. E. Atwater and R. R. Wheeler, Jr.: Appl. Phys. A, 79 (2004), 125.
28) J. E. Atwater and R. R. Wheeler, Jr.: Carbon, 41 (2003), 1801.
29) T. Iwashita: Shin-tansozairyonyumon, The Carbon Society of Japan, Tokyo, (2000), 26.
30) T. Kitano, K. Ohno, T. Maeda, K. Nishio and M. Shimizu: Proc. of 4th JEMEA meeting, 1, JEMEA, Tokyo, (2010), 78.
31) H. Kobayashi, S. Wang, F. Aida, K. Kagiwada and O. Hashimoto: IEICE Tech. Rep., 114 (2004), 31.
32) K. Iwauchi: Jpn. J. Appl. Phys., 10 (1971), 1520.