Quantification of Isothermal Phase Transformation in Solid Metals Based on Measurement of Magnetic Susceptibility

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A new method that provides quantitative information of phase transformation in solid metals by measuring a magnetic susceptibility has been proposed. The principle of the method is based on that the magnetic susceptibility is different from one phase to another and additive identity holds in the magnetic susceptibilities. This method directly reflects the phase transformation and provides more accurate information in comparison with conventional methods, which are based on indirect measurements of various physical properties or micro-structure observation. This method not only enables continuous measurement of phase transformation, but also can clearly indicate the start and end points of it.

In this study, this new method was applied to measure the \( a/\gamma \) isothermal transformation rate in an Fe–C alloy above the Curie temperature.

KEY WORDS: electromagnetic processing of materials; phase transformation in solid metals; \( a/\gamma \) phase transformation in Fe–C alloy.

1. Introduction

For measuring the phase transformation taking place in solid metals, there are several methods such as a direct way of micro-structure observation, and indirect ways through the measurement of physical properties such as hardness, electric resistance and thermal expansion. The former method is not suitable for continuous measurement of phase transformation and the latter ones are difficult for the clear determination of the start and end point of it.1) In this study, a new method has been developed to directly evaluate the phase transformation rate by measuring the magnetic susceptibility. The principle adopted in this method is based on the additivity of the magnetic susceptibility. That is, each composition ratio of mixed substances can be calculated by measuring the magnetic susceptibility of the composite material composed of mixed substances. The method proposed here has been applied to measure the \( \gamma/\alpha \) phase transformation rate during the isothermal phase transformation of an Fe–C alloy above the Curie temperature. This method allows to draw TTT (Time-Temperature Transformation) diagrams by continuously measuring the transitional variation of the magnetic susceptibility during the phase transformation.

2. Measurement of Magnetic Susceptibility

A magnetic susceptibility was measured by use of the Gouy method.3) A magnetization force acting on a sample in a magnetic field \( B_z \) is given as Eq. (1)

\[
F_z = \frac{m}{2L \mu_0} \chi (B_z^2 - B_0^2)
\]

where \( L \) is the sample height [m], \( \mu_0 \) the magnetic permeability in vacuum [H/m], \( m \) the mass of a sample [kg], \( \chi \) the magnetic susceptibility [m^3/kg], \( B_z \) and \( B_0 \) the magnetic flux densities [T] at the top and bottom positions of the sample, respectively. The weight difference measured by an electronic mass balance under the conditions with and without magnetic field, namely the magnetization force, is given as Eq. (2)

\[
F_z = M_{C+S} - M_{\alpha+S}
\]

where \( M_{C+S} \) and \( M_{\alpha+S} \) are the weights of a crucible and sample measured without and with magnetic field, respectively. Equation (2) includes the gravity and magnetization forces contributed from not only the sample but also the crucible. That is, it is not a pure magnetization force acting on the sample itself. Thus, the weight difference measured by an electronic mass balance under the conditions with and without magnetic field, namely the magnetization force, is given as Eq. (3)

\[
F_z = (M_{C+S} - M_{C+S}) - (M'_{C} - M_{\alpha})
\]

where \( M'_{C} \) and \( M_{\alpha} \) are the weight of the crucible measured
Derivation of Phase Transformation Rate

Magnetism in a substance is based on the motion of electrons in it, and magnetic susceptibility directly reflects the motion of electrons. Based on the assumption that the motion of electrons in one substance in the composite material composed of two substances is not affected by the existence of the other substance, the magnetic susceptibility of the composite material is expressed as the sum of products of the magnetic susceptibility and fraction of each substance. Thus, the magnetic susceptibility in the composite material composed of A and B substances can be expressed by Eq. (5).

\[ \chi = \chi_a f_a + \chi_b f_b \] ..............................................................(5)

where \( \chi \), \( \chi_a \) and \( \chi_b \) are the magnetic susceptibility of the composite material, and the A and B substances, respectively, and \( f_a \) and \( f_b \) the fraction of the A and B substances, respectively.

Let us apply the new method based on the magnetic susceptibility measurement to the \( \gamma/\alpha \) isothermal phase transformation in an Fe–C alloy above the Curie temperature. Figure 1 shows the equilibrium phase diagram of Fe–C system. A sample held at the austenite phase was quickly cooled from the single phase region to the \( \alpha+\gamma \) binary region and then isothermally kept at a given temperature in the \( \alpha+\gamma \) region. During the phase transformation, three phases of the \( \alpha \) phase, the original austenite phase \( \gamma \) with carbon concentration of \( c_0 \), and the new \( \gamma \) phase with carbon concentration of \( c \), coexist in the sample. If each fraction of the three phases is indicated by \( f_\alpha, f_\gamma \) and \( f_\gamma \), the following equation holds among them.

\[ f_\alpha + f_\gamma + f_\gamma = 1 \] ..............................................................(6)

On the basis of the additive identity of the magnetic susceptibility, the magnetic susceptibility \( \chi \) of the sample measured in the \( \alpha+\gamma \) binary region is expressed by Eq. (7).

\[ \chi = \chi_\alpha f_\alpha + \chi_\gamma f_\gamma + \chi_\gamma f_\gamma \] ..............................................................(7)

where \( \chi_\alpha \), \( \chi_\gamma \) and \( \chi_\gamma \) are the magnetic susceptibilities of the \( \alpha \), \( \gamma \) and \( \gamma \) phases, respectively. By introducing the assumption that the isothermal \( \alpha/\gamma \) transformation would take place under the lever rule, i.e. \( f_\gamma f_\gamma = b/a \) as seen in Fig. 1, the ratio of the new \( \gamma \) phase \( f_\gamma \) can be expressed by Eq. (8).

\[ f_\gamma = (a/b)f_\gamma \] ..............................................................(8)

From Eqs. (6), (7) and (8), the ratio of the \( \alpha \) phase \( f_\alpha \) is derived as Eq. (9).

\[ f_\alpha = \frac{b(\chi_\gamma - \chi_\gamma)}{b(\chi_\gamma - \chi_\gamma) + a(\chi_\gamma - \chi_\gamma)} \] ..............................................................(9)

Here, \( \chi \) is obtained by measuring the magnetic susceptibility of the sample during the isothermal transformation. In addition, \( \chi_\gamma \) and \( \chi_\gamma \) can be estimated by measuring the magnetic susceptibility of samples with various carbon concentrations in the \( \gamma \) single phase and extrapolating the measured values to a given temperature in the \( \alpha+\gamma \) region, on the basis of the Curie–Weiss law, namely, a magnetic susceptibility is inversely proportional to a temperature. Since the value of \( f_\gamma \) at the equilibrium state is given as \( f_\gamma = (a+b) \) from the equilibrium diagram shown in Fig. 1, and \( \chi_\gamma \) as the value of \( \chi \) at the equilibrium state can be obtained in the measurement, \( \chi_\alpha \) can be expressed as Eq. (10) by substituting \( f_\alpha \) and \( \chi_\gamma \) into Eq. (9), and rearranging.

\[ \chi_\alpha = \frac{(a+b)\chi_\gamma - a\chi_\gamma}{b} \] ..............................................................(10)

By substituting the values of \( \chi_\gamma \), \( \chi_\gamma \), \( \chi_\gamma \) and \( \chi_\gamma \) into Eq. (9), the transitional mass fraction of the \( \alpha \) phase \( f_\alpha \) with time can be continuously calculated. Hitherto, the \( \gamma \) and \( \gamma \) phases during the isothermal transformation have not been recognized as different phases since they could not be separately detected in a micro-structure observation. That is, this magnetic method enables to distinguish the \( \gamma \) and \( \gamma \) phases as different phases and this fact will lead to the better understanding of the \( \alpha/\gamma \) phase transformation.

4. Magnetic Susceptibility in \( \gamma \)-Phase

The magnetic susceptibilities of the \( \gamma \)-phase of the samples with different carbon concentrations were measured. Figure 2 shows an experimental apparatus. An alumina crucible loading each sample (pure-Fe, 0.1, 0.2, 0.4, 0.6 mass% C–Fe) was hung to an electronic mass balance. After discharging and filling Ar–5%H2 gas in a heater zone, the sample was kept at 1 000°C for 30 min under the imposition of a magnetic field of 2 T. The weights of the crucible with the sample and the empty crucible were measured by the electronic mass balance during cooling at a rate of 0.15 K/s, under the conditions with and without the magnetic field. Then, from these measured values, the magnetic susceptibilities of the sample were calculated by use of Eq. (4). Figure 3 shows the relationship between the inverse magnetic susceptibility in the \( \gamma \)-phase of the samples with
different carbon concentrations, and the temperature on the basis of the Curie–Weis law; namely, a magnetic susceptibility is inversely proportional to a temperature. Regarding to the pure Fe, the difference between the measured data and reference ones is about 1%. This small difference tells this new method gives comparatively accurate values. It is noticed that the magnetic susceptibility of the -phase increases with increasing of carbon concentration.

5. Isothermal Phase Transformation of 0.1mass%C–Fe

The experimental apparatus was nearly same as one shown in Fig. 2, but it equipped two heaters. The upper and lower heaters were used to austenitize a sample and to keep it at a given isothermal temperature in the α+γ binary region, respectively. A crucible having a sample of 0.1mass%C–Fe alloy was hung to the electronic mass balance through a pulley. After the air in the heater zones was ejected by discharging Ar–5%H₂ gas, the sample was kept at 950°C for 30 min in the upper heater zone. Then, the sample was slipped down to the lower heater zone by releasing the pulley and kept at the given temperatures of 800–850°C under a magnetic field of 2 T. The values of the weight indicated by the electronic mass balance were recorded in every 10 s interval, during the isothermal phase transformation. The cooling rate from 950°C to the phase transformation temperatures was about 1.5 K/s.

The transitional variation of the magnetic susceptibilities of the sample with carbon concentration of 0.1 mass% at different temperatures is shown in Fig. 4. The value of magnetic susceptibility at each temperature began to drastically increase after the incubation time of about 100 s and its increasing rate was suppressed between 100 s and 1000 s. The values of magnetic susceptibility at the equilibrium state are indicated by broken lines in this figure. The carbon concentration in the -phase at the transformation temperatures and that of samples are listed in Table 1 on the basis of the equilibrium phase diagram. Figure 5 shows the relation between the magnetic susceptibilities in the γ phase of the samples at the phase transformation temperatures and the carbon concentrations.

Table 1. Carbon concentrations in γ phase extrapolated to the transformation temperature.

<table>
<thead>
<tr>
<th>Transformation temperature (°C)</th>
<th>Extrapolated carbon concentration in γ phase (mass%)</th>
<th>Carbon concentration of the sample (mass%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>800</td>
<td>0.34</td>
<td>0.1</td>
</tr>
<tr>
<td>825</td>
<td>0.24</td>
<td>0.1</td>
</tr>
<tr>
<td>845</td>
<td>0.18</td>
<td>0.1</td>
</tr>
</tbody>
</table>
The solid points were obtained by extrapolating the continuously measured magnetic susceptibility lines shown in Fig. 3 to the phase transformation temperatures as shown in solid point marks in Fig. 3. That is, those points show the relations between the values of the magnetic susceptibility in the \( \gamma \) phase that were extrapolated to the transformation temperatures and the \( \gamma_0 \)-phase carbon concentrations given in Table 1. According to Fig. 5, the magnetic susceptibilities in the \( \gamma \) phase can be interpolated under given temperature and carbon concentration and are shown by white points. The value of \( c_a \) was calculated by substituting the values of \( c_e \) and the values of \( c_g \) shown by the white points in Fig. 5, which were interpolated by use of the data given by solid points, into Eq. (10). And the value of the \( c_g \) is estimated by extrapolating the magnetic susceptibility of 0.1 mass\% C–Fe to the phase transformation temperature as shown in Fig. 3. The calculated values of \( \gamma \) are \( \gamma_0 \) and \( \gamma \), at the given temperatures are listed in Table 2. Then, the transitional variations of \( f_g \), \( f_{\gamma_0} \) and \( f_{\gamma} \) with time were calculated by substituting the values listed in Table 2 into Eqs. (6), (8) and (9), and shown in Fig. 6, where each point indicates datum obtained in 10 s interval. The start and end points of the phase transformation can be clearly observed. At every transformation temperature, the \( \alpha \) phase began to precipitate in 40 s and gradually reached at the equilibrium state after about 300 s via its rapid variation period between them. The phase transformation ratio \( P \) is defined as \( P=f_{\alpha}/f_{\gamma_0} \), where \( f_{\gamma_0} \) is the value at the equilibrium state of the \( \alpha \) phase. Figure 7 shows the times reaching at the start point \( (P=0.01) \), the middle point \( (P=0.5) \) and the end point \( (P=0.99) \) at the given transformation temperatures 800, 825 and 845°C. The phase transformation started at nearly the same moment regardless of the transformation temperatures, but the times reaching at \( P=0.5 \) and \( P=1.0 \) depended on the phase transformation temperatures. A nose shape, which is ordinary observed in TTT diagrams below an eutectoid temperature was figured out by connecting the times reaching at \( P=0.5 \) or \( P=1.0 \) with the different transformation temperatures.

6. Conclusion
A quantitative evaluation method of phase transformation in solid metals has newly been developed. And the \( \gamma/\alpha \) transformation rate of Fe–C alloy was quantified above the Curie temperature by use of the method. The following results have been obtained.

(1) The magnetic susceptibility of the \( \gamma \) phase increases with increase of carbon concentration.

(2) By distinguishing precipitated \( \gamma \) phase from the original \( \gamma_0 \) phase as different phases, the transitional \( \gamma/\alpha \) phase transformation with time was accurately quantified.

(3) The nose shape which is ordinary observed in TTT diagrams below the eutectoid temperature was detected above the Curie temperature.

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REFERENCES

Table 2. The value of magnetic susceptibilities of \( \chi_{\gamma_0} \), \( \chi_{\gamma} \), \( \chi_{\alpha} \) and \( \chi_e \).

<table>
<thead>
<tr>
<th>Transformation temperature (°C)</th>
<th>Magnetic susceptibility ( m^3/kg \times 10^8 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>890</td>
<td>0.362 0.372 20 14.6</td>
</tr>
<tr>
<td>825</td>
<td>0.361 0.367 16 10.2</td>
</tr>
<tr>
<td>845</td>
<td>0.360 0.364 15 6.78</td>
</tr>
</tbody>
</table>

Fig. 6. Transitional variation of the fraction ratio of the \( \gamma_0 \), \( \gamma \) and \( \alpha \) phases in sample of 0.1 mass\% C at different transformation temperatures.

Fig. 7. The phase transformation time reaching at phase transformation ratios of \( P=0.01 \), \( P=0.5 \) and \( P=0.99 \) under different phase transformation temperatures.