Prediction Model for the Austenite Grain Size in the Coarse Grained Heat Affected Zone of Fe–C–Mn Steels: Considering the Effect of Initial Grain Size on Isothermal Growth Behavior

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The austenite grain size in the coarse-grained heat affected zone (CGHAZ) was predicted by analyzing isothermal grain growth behavior of Fe–C–Mn steels which were designed to investigate the effect of alloying elements. A procedure was proposed to prevent inappropriate neglect of initial grain size \( D_0 \) and misreading both time exponent and activation energy for isothermal grain growth. It was found that the time exponent was almost constant, irrespectively of temperature and alloying elements, and activation energy increased with the addition of alloying elements. From quantification of the effect of alloying elements on the activation energy, an isothermal grain growth model was presented. Combining with the additivity rule, the austenite grain size in the CGHAZ was predicted.

KEY WORDS: austenite grain growth; heat affected zone; model.

1. Introduction

Austenite grain growth in the weld heat affected zone (HAZ) is important in that grain size in the weld metal is controlled by that in the HAZ because weld metal grains grow epitaxially from HAZ, as well as austenite grain size affects the kinetics of phase transformation during welding cooling cycle. Studies on models for grain growth have been mostly carried out for thermo-mechanical process, such as reheating process prior to hot rolling.1,2) It was, in most cases, general that isothermal grain growth model which was developed by analyzing growth kinetics was extended to a non-isothermal situation, using the rule of additivity. There were a few studies3) which adopted this procedure for HAZ. In this case, the exact derivation of isothermal model is, however, very important.

The kinetics of isothermal grain growth in metals has been intensively studied and it has been empirically known that the behaviors of grain growth obey the following classical relationship,4)

\[
D^{1/n} - D_0^{1/n} = K \cdot t \quad \text{..................................(1)}
\]

where \( n \) and \( K \) are respectively time exponent and rate constant, which are experimentally determined, depending on material and temperature. \( D \) is an average grain diameter and \( D_0 \) a grain size at \( t=0 \) s. However, a simple Beck’s relationship,5) Eq. (2) for grain growth kinetics has been widely utilized because it has advantages that empirical parameters, \( n \) and \( k \) can be easily obtained by plotting \( \ln D \) against \( \ln t \).

\[
D = k \cdot t^n \quad \text{..............................(2)}
\]

This equation has an implicit assumption that \( D_0 \) is negligible compared to \( D \). Thus, the usage of Eq. (2) should be limited only to cases where \( D \gg D_0 \). However, this limitation has seldom been carefully considered. This inappropriate neglect of the \( D_0 \) term can affect the value of time exponent, especially in the temperature ranges where the mobility of grain boundary is low. In addition, the \( K \) value in Eq. (1) gives the information for the activation energy for boundary migration according to the velocity–driving force correlation established by Rath and Hu.4) However, in the case where the Eq. (2) is applied to analyze the isothermal grain growth kinetics, the relationship, \( K = k^{1/n} \), which was used without any considerations, is mathematically not satisfied, as well as the activation energy can be misinterpreted due to misreading of the \( n \) value.

In this paper, a method which considers the effect of \( D_0 \) by using Eq. (1) will be proposed. The empirical model for isothermal austenite grain growth will be developed through analyzing the effect of alloying elements on growth kinetics by using this method. Combining with the additivity rule which is generally applied to a non-isothermal condition, a model for predicting the austenite grain size in the HAZ will be addressed.

2. Concept for Analyzing Isothermal Grain Growth

As previously mentioned, the Beck’s equation, Eq. (2) has generally been utilized to analyze isothermal grain growth kinetics due to the ease for obtaining parameters.
Migration is high and thus, $n$ approximates 0.3. It seems because the mobility for boundary which is determined by conventional Beck’s equation approaches 0.3. The Beck’s equation, Eq. (2) are also included as dotted lines in Fig. 1. As depicted in Fig. 1, it is found that the deviation from linearity appears in short time range. However, in the case of higher $K$ value assuming high temperature, $n$ value which is determined by conventional Beck’s equation approximates 0.3. It seems because the mobility for boundary migration is high and thus, $D_0$ is very small as compared with $D$. Meanwhile at lower temperatures, the deviation from the linear relation is found to be considerable in short times and thus, time exponent, slope of dotted line is much smaller than 0.3. This example means, in other words, that the direct usage of Beck’s equation without consideration of the effect of $D_0$ can lead lower time exponent and further misinterpretation for isothermal grain growth kinetics.

On the other hand, in many studies on the kinetics of grain growth, the activation energy for grain boundary motion was determined through investigating temperature-dependence of $K$. There is a possibility of misinterpretation of activation energy due to above assumption, as well as the possibility of lower time exponent from conventional analysis by using Eq. (2). Thus, to analyze the isothermal grain growth kinetics exactly, it is thought that especially, in the case where $D_0$ is not negligible in comparison with $D$, Eq. (1) should be utilized.

However, there is a problem that the parameters in Eq. (1) are difficult to be directly measured from observed grain growth behavior. In Fig. 1, there is a linear region of logarithmic plot with lower $K$ value and the slope is similar to that with higher $K$ value. Figure 2 compares Beck’s equation (Eq. (2)) with Eq. (1) with $n$ equals 0.3. The Beck’s equation deviates substantially from Eq. (1) when $D$ approaches $D_0$ in a region where the two curves nearly overlap. This region appears to be about $D > 3D_0$, similar to that which was reported to be a “safe region” by Vandermeer et al. It was confirmed that this criterion was satisfied for various $K$ and $n$ values. Therefore, this region provides a criterion for the appropriate use of Beck’s equation. Based on this concept, isothermal grain growth kinetics is analyzed through successive 2 processes. Firstly, the $n$ value in Eq. (1) is determined by using Beck’s equation within a region defined by only $D > 3D_0$. Subsequently, $K$ parameter in Eq. (1) is obtained using least-squares plots of $D^{(1/n)} - D_0^{(1/n)}$ against a time.

3. Experimental

Steels used in this study were melted in a vacuum induction furnace to examine the effect of alloying elements on isothermal grain growth kinetics. The final compositions are given in Table 1. As shown in Table 1, contents of carbon, manganese, nickel and chromium are varied with fixed molybdenum and silicon contents. Ingots were homogenized for 12 h at 1200°C to eliminate segregation and forged into rods with 10 mm diameters and then austenitized at 950°C for 1 h, followed by air cooling. Rods were machined into specimens with 3 mm diameters and 10 mm lengths.

The initial microstructures were investigated by both optical and transmission electron microscopy. Carbon extraction replicas were analyzed using TEM (JEOL2010) with an EDAX system. Figure 3 shows the initial microstructures by optical microscopy. Steels mostly consist of low temperature phases such as bainite and/or martensite with little amount of polygonal ferrite, while steel A with the lowest carbon equivalent has more than 80% polygonal ferrite with about 150 μm in grain diameter.

A dilatometer equipped with gas quenching system was used to perform the heat treatment that required to observe isothermal grain growth behaviors and to compare the prediction results with grain sizes in the simulated HAZ. Figure 4 indicates the experimental thermal cycles. Isothermal test temperature ranged from 900 to 1300°C. Specimens were quenched to room temperature after holding with various intervals at each test temperature to secure prior austenite grain structure. Thermal cycles to simulate...
HAZ microstructure were obtained from a simple model based on heat-flow equation. Quenched specimens were polished by conventional metallographic techniques and then etched in an aqueous mixed solution of picric acid and HCl at 70–80°C to reveal prior austenite grain boundary. Figure 5 is the optical micrographs showing the prior austenite grain boundaries. Average austenite grain sizes were measured by quantitative image analyzer (Image-Pro Plus), ensuring that the area of measurement was at least $7.8 \times 10^6 \mu m^2$.

### 4. Experimental Results and Discussions

#### 4.1. Grain Growth Behavior

Figure 6 shows the austenite grain growth behaviors at several isothermal temperatures. Although there are some scatters, the average grain size increases, as expected, with temperature and time.

#### 4.2. Time Exponents

Figure 7 shows the logarithmic plot for austenite grain growth behavior of steel D representatively. As previously mentioned, a deviation from the linearity in short times especially at low temperatures can be observed. Figure 8 compares $n$ values by conventional Beck’s equation with by

| Steel | C   | Mn  | Ni  | Cr  | Mo  | Si  | Ceq* |
|-------|-----|-----|-----|-----|-----|-----|------|
| A     | 0.081 | 0.487 | 0.007 | -   | 0.400 | 0.263 | 0.26 |
| B     | 0.087 | 0.500 | 2.04  | 1.00 | 0.497 | 0.270 | 0.68 |
| C     | 0.083 | 1.480 | 3.88  | 1.98 | 0.496 | 0.256 | 1.09 |
| D     | 0.192 | 0.515 | 2.03  | 2.00 | 0.488 | 0.389 | 0.91 |
| E     | 0.198 | 1.040 | 4.00  | -   | 0.493 | 0.258 | 0.74 |
| F     | 0.213 | 1.470 | 0.002 | 1.00 | 0.495 | 0.270 | 0.76 |
| G     | 0.305 | 0.540 | 4.00  | 0.99 | 0.489 | 0.276 | 0.96 |
| H     | 0.316 | 1.010 | 0.003 | 2.03 | 0.499 | 0.285 | 0.99 |
| I     | 0.321 | 1.510 | 2.02  | -   | 0.497 | 0.267 | 0.81 |

* Carbon equivalent $C_{eq} = C + \frac{Mn}{6} + \frac{Cr + Mo + V}{5} + \frac{Ni + Cu}{15}$
a newly proposed one bearing in mind that meaningful data should be $D > 3D_0$. The data measured at 1000°C were not included in Fig. 8(b), because average grain size after holding for 600 s did not satisfy the above criteria for applying new method. This seems to be caused by low mobility for boundary migration at the low temperature though the driving force for grain growth may be large.

Measured by the conventional method, time exponents
seem to decrease with temperature in Fig. 8(a). However, when considering the effect of inappropriate neglect of \( D_0 \) on analyzing growth kinetics by applying the new method (Fig. 8(b)), the temperature-dependence of time exponents almost disappears and then the time exponent can be reasonably regarded as a constant, 0.2432±0.019, irrespectively of temperature. There also seems to be little variation with chemical compositions.

Meanwhile, measured time exponents appear to be much lower than the theoretical value (0.5) which was reported in high purity metals at high temperature. There were many studies\(^4,8,9\) to explain the variations of time exponent. In those studies, the lower value of time exponents was attributed to several factors such as pinning force by second phase precipitates, specimen thickness effect, solute drag effect and so on. Figure 9 shows the equilibrium phase diagram and volume fraction calculated by Thermo-Calc. for the given composition. Though there are many types of carbides at equilibrium, only cementites are experimentally observed (Fig. 10). These cementites can be thought to exert the pinning force for grain growth. However, although the superheating of dissolution temperature of precipitates occurs by the fast heating rate in the experiments, the measured low \( n \) value (<0.5) seems not to be attributed to precipitates because the equilibrium dissolution temperatures of carbides, including cementite, are very low. Growth inhibition effect by the specimen thickness is also not a main factor when considering the dimension of specimen. On the other hand, there is a transition region between high velocity limit in which the grain boundary migration is mainly controlled by the diffusion of solvent atoms and low velocity limit in which the migration rate is governed by diffusion of solute atoms, according to impurity-drag theories\(^10,11\). The transition region shows the non-linear velocity vs. dri-
ving force relationship, i.e. $n < 0.5$ and implies that a solute drag effect is involved in the grain boundary migration process.\textsuperscript{12,14} Therefore, observed lower time exponents can be thought to be an indicative of the solute drag effect even if more research is required.

### 4.3. Activation Energy for Austenite Grain Growth

Figure 11(a) shows the steps analyzing the isothermal grain growth kinetics to obtain the $K$ value. From the form of Eq. (1), when plotting $(D^{1/0.2432} - D_0^{1/0.2432})$ vs. $t$ plot for steel $F$, (b) Arrhenius-plot for activation energy.

| Fig. 10. TEM micrograph and EDAX for carbon extracted replicas. |
| --- |
| Fig. 11. Method for determination of activation energy for grain growth. (a) $(D^{1/0.2432} - D_0^{1/0.2432})$ vs. $t$ plot for steel $F$, (b) Arrhenius-plot for activation energy. |
| Fig. 12. Effect of alloying elements on activation energies. |

The measured activation energies, in general, appear to be similar to each other and averaged about $409 \pm 21$ kJ/mol. This value is very large, even compared to the activation energies for substitutional interdiffusion of transition elements in dilute austenite solution, known as \textit{ca.} 260 kJ/mol,\textsuperscript{13} as well as that for lattice self diffusion in pure $\gamma$-iron, 284 kJ/mol.\textsuperscript{13} These large activation energies are, however, not surprising if it is accepted that the observed growth kinetics comes into the transition region in boundary velocity and driving force relationship, because, in the transition region, the activation energies can not meaningfully and simply correspond to that for diffusion of either solvent or solute. Large activation energies were also reported in many studies\textsuperscript{13,14} and associated with the non-linear relationship between driving force and boundary velocity.

On the other hand, as can be seen in Fig. 12, activation
energies tend to increases with the concentration of alloying elements. To quantitatively express the effect of alloying elements, measured activation energies were analyzed by multiple-regression and then following equation could be obtained.

\[ Q = \frac{352.185.31}{100} + 21.827.26X_C + 19950.94X_{Mn} 
  + 7185.49X_{Cr} + 7378.06X_{Ni} \]

where \( X_i \) is the concentration of element, \( i \) in weight percent. From Eq. (4), it is found that activation energy is largely affected by carbon and is followed by the order of Mn, Ni and Cr. In the dissolved state, alloying elements has been known to inhibit the grain growth by exerting elastic attraction of the atoms toward grain boundary. This interaction of solute-boundary was reported to be affected by the difference of atom size and segregation into boundary which is inversely related to solid solubility. These parameters are summarized in Table 2. The relative extent of coefficients in Eq. (4) appears to be reasonable.

### 5. Isothermal and HAZ Grain Growth Model

From analyzing the isothermal grain growth kinetics in Fe–C–Mn steels, the following equation for isothermal grain growth can be derived.

\[ D^{0.243} - D_0^{0.243} = 1.72 \times 10^{-21} \exp\left\{ -\left[352.185.31 + 21.827.26X_C + 19950.94X_{Mn} 
  + 7185.49X_{Cr} + 7378.06X_{Ni}\right]/RT\right\} \]

where \( T \) is the absolute temperature in unit K.

**Figure 13** compares the austenite grain size during isothermal holding between calculation and measurement. Solid line indicates the equality of data, showing the reasonable agreement between calculations and experimental measurements.

Utilizing Eq. (5) for the isothermal grain growth kinetics and an additivity rule, a model for predicting the austenite grain size during a continuous welding thermal cycle can be established. The additivity rule, which has been widely used in modeling for phase transformation kinetics, regards the welding thermal cycle as a series of isothermal steps. Therefore, the summation of grain size increments calculated by isothermal duration at each temperature step leads to final grain size.

**Figure 14** shows the calculated austenite grain size for steel B and F at a constant heating rate, 100°C/s.

| Table 2. Physical constants for alloying elements. |
|---------------------------------|------|------|------|------|
| Difference of atomic radius from Fe (nm)\(^{11}\) | C   | Mn   | Cr   | Ni   |
| 0.1164 | 0.0106 | 0.0008 | 0.0005 |
| Max. solubility in γ-Fe (wt%)\(^{11}\) | 1.7  | 100  | 100  | 11.2 |

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temperatures increases with initial grain size after austenitization, \( D_0 \), the difference between grain sizes at temperatures higher than 1100°C, generally coarse grained HAZ, is negligible. In this study, the initial austenite grain size is, therefore, fixed as 0.1 \( \mu m \) at 900°C. Of course, to predict the grain size at lower temperatures exactly, for example, Fine-grained (FG) HAZ and even Intercritically-reheated (IC) HAZ, the austenite starting grain size after transformation to austenite should be predicted. This requires the information about the theories of nucleation and growth for phase transformation.

**Figure 15** shows the prediction results at various peak temperature in the HAZ for SA508-cl.3 (Fe–0.2C–0.26Si–1.34Mn–0.15Cr–0.86Ni–0.49Mo) at the heat input of 40 kJ/cm. At lower temperatures the predicted size is smaller than measured size. This seems to be because the initial grain size is fixed as 0.1 \( \mu m \), as mentioned in previous paragraph. However, austenite grain size in the CGHAZ region has a good agreement with measurements.

6. Conclusions

Through analyzing the isothermal grain growth kinetics in Fe–C–Mn–Cr–Ni–Mo–Si, the model for predicting the austenite grain size in the CGHAZ was developed. When analyzing isothermal grain growth, it was found that inappropriate neglect of \( D_0 \) could cause time exponent to be lower and activation energy could be misread by using conventional method for analysis. To prevent this misinterpretation of rate constant as well as time exponent, the new method was proposed and utilized to analyze grain growth kinetics.

(1) Time exponent determined by new method is almost constant regardless of isothermal temperature and concentration of alloying elements. The lower value of time exponent, 0.2432 than 0.5 is thought to be caused by a solute drag effect.

(2) Activation energy for grain growth is found to increase with the addition of alloying elements by the order of C, Mn, Ni and Cr.

(3) The isothermal austenite grain growth model and HAZ model were developed and evaluated. In HAZ model, prediction results at high temperatures have a good agreement with measurement, while calculation at low temperatures appears to be poor. It is thought that this poor accuracy at low temperatures is attributed to the arbitrarily fixed value of \( D_0 \) after austenitization and can be improved by take into account the phase transformation into austenite.

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