

KINETICS MODELLING OF ISOTHERMAL BAINITE TRANSFORMATION IN LOW CARBON MULTI-MICROALLOYED STEEL

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Abstract

In this work, the isothermal bainite transformation in a low carbon multi-microalloyed steel was investigated using a dilatometer. Two different transformation behaviors were identified according to the measured dilatometric curves. The variation of transformation mechanism was explored based on the microstructural observation and the change of the Avrami exponent with the holding time and isothermal temperature. For example, the solute drag effect is mainly responsible for the transformation stasis phenomenon at the first stage of the abnormal reaction curve. A physically-based model was proposed to describe the normal bainite formation kinetics curves on the basis of the modification of the Johnson-Mehl-Avrami (JMA) equation. The transformation mode is considered as the site saturation of nucleation and interface controlled growth under the large undercooling condition. Comparison between the calculation curves and the experimental data in this experimental steel gives a very good agreement.

Introduction

The control of transformation of the austenite to ferrite phase during the production of steels is extremely important to tune and optimize the final microstructure and mechanical properties [1-3]. Taking as a typical example the accelerated cooling technology, the properties of the low carbon microalloyed steels can achieve easily a good combination of high strength and toughness as well as good weldability as the accelerated cooling method assists the bainite transformation [4]. Thus, this kind of steels has been widely applied in building, bridge, pipeline and offshore structures. To reveal the potential laws of the microstructural evolution as a function of transformation time and/or temperature, much effort has been spent on the modelling of phase transformations in the last several decades [1-3, 5-8]. However, it is no easy task to extract from such experimental data quantitative information on the operating modes of nucleation, growth and impingement of product phase because of the diversity of phase transformation.

The classical Johnson-Mehl-Avrami (JMA) equation often plays an important role in studies of the solid state phase transformations, which is valid to describe the phase transformation behaviors under most circumstances [3]. For example, there usually exist two methods to model the kinetics of isothermal bainite transformation according to different viewpoints to the bainite transformation mechanism: One is carbon diffusion controlled growth mechanism [7, 9]; the other is martensite-like shear mechanism [8, 10]. In the end, the "expand volume" of phase transformation based on the above bainite transformation mechanisms should be modified using the JMA phenomenological model [7-10]. However, because the JMA model is assumed that the nuclei are dispersed randomly in the total volume and grow isotropically, some of abnormal

phase transformation kinetics is not suitable to be described by the traditional JMA model [3, 5, 6]. For example, under the lower driving force of transformation condition overall transformation kinetics seems not to obey the exponent growth mode when the occurrence of solute drag effect retards the reaction kinetics [2, 11]. Therefore, the solute drag theory was applied to model ferrite formation in multiphase steels, as proposed in some papers [12, 13]. Recently, the modular transformation model has been developed and applied very successfully to a number of strikingly diverse transformations [5, 6].

In this work, the isothermal bainite transformation of a low carbon multi-microalloyed steel was examined by using a dilatometer. Two distinct transformation behaviors were observed in the range of the isothermal bainite formation temperature. However, we mainly focused on the establishment of kinetics model of the normal phase transformation behavior.

Experimental Procedure

The composition of the low carbon multi-microalloyed steel used in this study is 0.053 C-0.22 Si-1.63Mn-1.02(Cr + Cu + Mo)-0.07(Nb + V+ Ti) (wt. %). The dilatometric specimens cut from the hot rolled steel plate were machined into the cylindrical shape with dimensions of $\phi 3 \times 10$ mm. The whole heat treatment process was carried out using a Formastor-FII machine as follows: each specimen was austenitized at the peak temperature of 1300 °C for 5 s to obtain the large austenite grain size. Then it was quenched by a helium jag within the quench time of 0.1 s to the programmed isothermal holding temperature in the range of bainite formation (460~560 °C). After annealing at a holding temperature for 1000 s or more, the transformed specimen was quenched again by a hydrogen jet (about 10 s) down to room temperature. The changes in length of the specimens were recorded in the whole heat treatment process. The microstructures of the dilatometric specimens were observed using an optical microscope after they were prepared by conventional grinding and polishing techniques and etched with 3 % nital etchant.

Results

Figure 1 shows the changes in the morphology of isothermal bainite microstructure with isothermal temperature. The main microstructure is characterized by fine lath bainite (or bainitic ferrite) with a small fraction of massive martensite/austenite (M/A) constituents attached to prior austenite grain boundary when the isothermal temperature is 460 °C (Figure 1a). The M/A constituents are a kind of untransformed austenite in the equilibrium state at the isothermal stage because the carbon segregation on these constituents lowers their chemical free energies [14]. Therefore, the amount of the M/A constituent represents the degree of incomplete bainite transformation. With the increase in the isothermal temperature, the main microstructure changes from bainitic ferrite to granular bainite then to widmanstätten ferrite, as shown in figs. 1b and 1c. And the amount of the M/A constituent increases notably measured by point counting method. However, the residual austenite can be only transformed into a mixture of martensite and lath bainite rather than the fully martensite during the helium jag quenching when the temperature is about 530 °C (Figure 1c). It is evident that the bainitic laths always nucleate on the prior austenite grain boundary and their width sizes become larger with increasing isothermal temperature. When the temperature rises to 550 °C, the grain boundary polygonal ferrite forms during the long isothermal time without the formation of bainite morphology (Figure 1d), indicating that the bainite start formation temperature should be slightly below 550 °C.

The relative change in length of the specimens with the holding time and the temperature is given in Figure 2. Obviously, the maximum relative dilatation is as a function of temperature, increasing with decreasing isothermal temperature (Figure 2a). The dilatometric curves can be

classified into two different types according to the change trend of curves at the end stage of the holding time as well as during the second quenching stage (Figure 2b). The first type curves in the temperature range of 460~500 °C show that the isothermal bainite transformation has finished as all curves become the horizontal lines and no second phase transformation behavior can be detected during the second quenching stage. The finish time of isothermal transformation is extended with the decrease in isothermal temperature, as arrowed in Figure 2a. By contrast, the second type curves lie in the temperature range of 510~540 °C. This type curves show that the dilatation still increase with a certain rate at the last isothermal stage, which suggests that the isothermal transformation does not completely terminate. And the second transformation behavior occurring at the second quenching stage highlighted in Figure 2b strongly proves that quite a number of untransformed austenite must exist before the quenching.

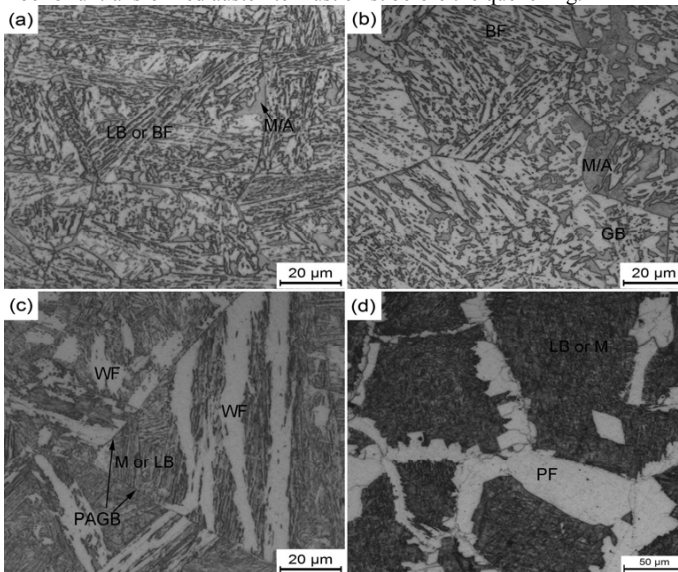


Figure 1. The morphology of isothermal bainite microstructure: (a) 460°C for 1000s; (b) 490°C for 1000s; (c) 530°C for 1000s; (d) 550°C for 7200s (LB: lath bainite, M/A: martensite/austenite constituent, BF: bainitic ferrite, GB: granular bainite, WF: widmanstätten ferrite, PAGB: prior austenite grain boundary, M: martensite, PF: polygonal ferrite)

The volume change of the sample is assumed to be isotropic. That is to say, the volume change accompanying the transformation is directly related to the measured length change of the sample. Therefore, the volume fraction transformed $f(t)$ can be calculated by Equation (1) according to the classic lever rule based on the dilatometric curves, as shown in the Figure 3a.

$$\frac{f(t)}{f_{\max}} = \frac{\Delta L(t)}{\Delta L_f} \quad (1)$$

where f_{\max} is the maximum volume fraction at each isothermal temperature, $\Delta L(t)$ and ΔL_f corresponding to the relative dilatation at t s and the quenching time, respectively. The maximum volume fraction transformed can be obtained based on the optical micrographs (Figure 1). It can be seen that the kinetics of the isothermal bainite transformation is very slower at the higher

isothermal temperature (e.g. 540 °C) compared with the lower isothermal temperature (Figure 3b). The kinetics of the second type curves also exhibit two-stage transformation behavior: the transformation rate is relatively fast at first stage, followed by a second, linearly steady stage. While the kinetics of the first type curves show the typical “S” shape, which is called the normal phase transformation kinetics curve.

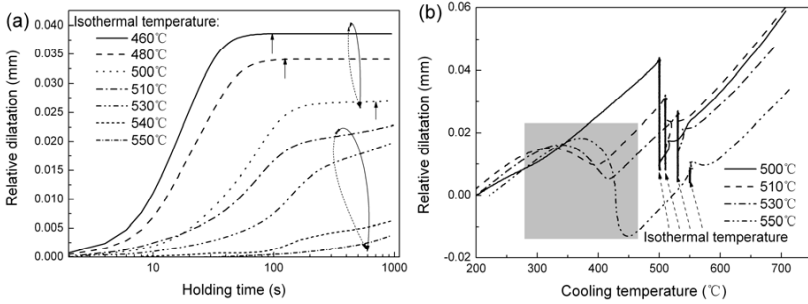


Figure 2. (a) The variation of relative dilatation as a function of holding time, the transformation finish times are indicated with arrows and two different types of curves were circled separately, (b) the variation of relative dilatation with the cooling temperature and isothermal temperature signified with arrows

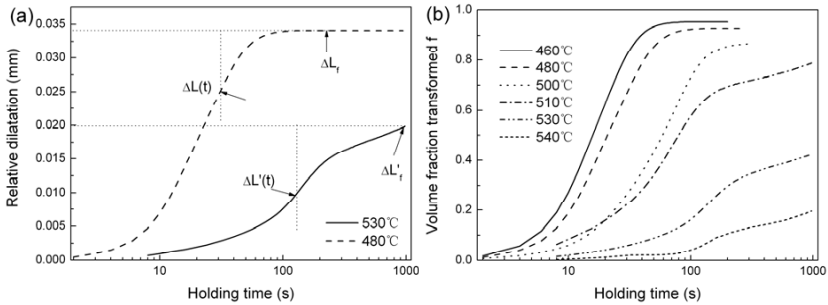


Figure 3. Kinetics of isothermal bainite transformation (a) the lever rule method, (b) actual volume fraction transformed

Discussion

The conventional JMA equation is generally used to model phase transformation kinetics under isothermal conditions as a purely phenomenological description. Therefore, this formal theory is largely independent of the particular models used in detailed descriptions of the mechanism of transformation [3]. The general expression of the JMA equation is shown in Equation (2).

$$f = 1 - \exp[-k(t - t_0)^n] \quad (2)$$

where f is the volume fraction of the transformation product, t is the reaction time, t_0 is the incubation time, k is the rate constant, and n is Avrami exponent indicative of the transformation mechanism. Because the incubation time of bainite transformation in the low carbon steel is very short (Fig. 3b), it is assumed to be zero in this study. If the Avrami exponent can be regarded as a

constant at the whole isothermal transformation stage, the above JMA equation can be changed into Equation (3) by taking the Equation (2) twice logarithm.

$$\ln[-\ln(1-f)] = n \ln(t) + \ln k \quad (3)$$

It is clear that the n value is equal to the slope of the linear portion of the curves in Figure 4. For the normal transformation curves, the n value is almost a constant at each isothermal temperature, and the value decreases slightly with the increase in the isothermal temperature. Thus, the transformation kinetics is supposed to be described reasonably by the JMA equation under this situation. As regards the abnormal transformation curves (510 to 540 °C), the n value presents several linear change stages obviously with the holding time. It is interesting found that the n value decreases to the same value (about 0.35) at the last stage for each abnormal curve. In combination with microstructural observation that the ferrite laths become wider with the increasing in holding time. It can be concluded that the transformation may be controlled by carbon diffusion at the last isothermal stage. In addition, the n value becomes zero at the second stage for 540 °C isothermal temperature, indicating the occurrence of transformation stasis. This phenomenon may be mainly attributed to the solute drag effect inhibiting the interface movement at the relatively lower chemical driving force condition as reported in various literatures [2, 11, 12]. As a result, the traditional JMA equation can not describe this bainite transformation.

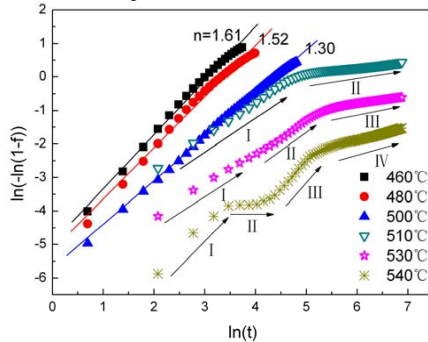


Figure 4. The relationship between $\ln[-\ln(1-f)]$ and $\ln(t)$ indicating the change of the Avrami exponent with the isothermal temperature and holding time

As mentioned above, the JMA model is a kind of overall phenomenological description for transformation kinetics. To provide a physically based overall kinetics model of the bainite transformation, we need a description of both the nucleation and growth kinetics of the new product. Here, the modular phase transformation model mainly proposed by Liu et al. [5] was employed to analyze the normal bainite transformation kinetics.

For isothermal transformation, different nucleation modes can prevail in the transformation process [3, 5, 6], e.g. continuous nucleation mode, nucleation site saturation and the mixed nucleation mechanism mode. According to the present experimental results, the large degree of supercooling can provides the saturation of the nucleation sites at the very early in the transformation and the number of the pre-existing nuclei increases with increasing the undercooling degree. Thus, the nucleation can be supposed to be site saturation mode. That is, all nuclei are present at the beginning of transformation and the further nucleation rate is zero. This implies for the nucleation rate:

$$\dot{N}(T) = N^* \delta(t-0) \quad (4)$$

where N^* is the number of pre-existing nuclei per unit volume, T is the absolute temperature and $\delta(t-0)$ denotes the Dirac function.

Each nucleus of bainitic lath grows in the length direction with the holding time. Their growth rate can be controlled by interface movement and/or carbon diffusion rate. For the case of interface-controlled growth with large undercooling, the interface velocity is given by [5]

$$v(T(t)) = v_0 \exp\left(-\frac{Q_G}{RT(t)}\right) \quad (5)$$

where v_0 is the pre-exponential factor for growth, Q_G is the activation energy for the transfer of atoms through the interface and R is the gas constant.

Under the carbon diffusion-controlled growth condition, the growth rate of bainitic lath is determined by the carbon diffusivity in the untransformed austenite. Based on the assumption of local equilibrium of carbon atom at the moving interface, Hillert [15] proposed an empirical equation to calculate the diffusion controlled growth rate as follows:

$$v(T(t)) = 2.5 \times 10^6 \frac{DT}{C^0} (C^{\gamma\alpha} - C^0)^2 \quad (6)$$

where D is the carbon diffusivity in austenite, C^0 is the bulk carbon concentration and $C^{\gamma\alpha}$ is the carbon concentration at the boundary in austenite.

According to the morphology of bainite lath in two dimensions (Figure 1), its spatial structure can be considered as a cylindrical shape. The width of bainitic lath w_B is found to be determined mainly by the isothermal temperature and the average carbon content of parent austenite [9], as shown in an empirical equation of Equation (7).

$$w_B (\mu m) = [4.2 \times 10^{-4}(T - 273) - 8.7 \times 10^{-2}] / C_\gamma^{0.36} \quad (7)$$

Therefore, the volume of a growing bainitic lath can be calculated at the time t after it nucleated at time τ :

$$Y(T) = 0.25\pi w_B^2 \int_{\tau}^t v(T(t)) d\tau \quad (8)$$

Assuming that the transformation occurs in an infinite large volume, the nucleation and growth of the new phase is not affected by the existing product phase. In other words, the impingement of product is neglected. According to the above nucleation mode (site saturation) and growth mode (e.g. interface controlled growth), the extended volume of the bainite formation is given by:

$$V_e = \int_0^t V\dot{N}(T)Y(T(\tau))d\tau = 0.25\pi V\dot{N}(T)w_B^2 v(T)t^n \quad (9)$$

where n is the above Avrami exponent. However, the impingement between the product phases is absolutely present in the actual transformation process. Therefore, the extended volume should be modified by the JMA equation. The actual volume fraction transformed is given as:

$$f = 1 - \exp(-\pi\lambda w_B^2 \dot{N} v t^n) \quad (10)$$

where λ is a parameter which has to be adjusted from the experimental isothermal kinetics. Because the JMA model is assumed that the transformation is random nucleation, constant nucleation rate, and linear growth, the calculated volume fraction may have some deviation from the measured volume fraction. Figure 5 shows the predicted isothermal kinetics according to Equation (10). The predicted kinetics is in a good agreement with the experimental results. It is worth noting that, in order to simplify the parameters, the nucleation rate \dot{N} can be firstly estimated by austenite grain size and the growth rate of bainitic laths with the large undercooling can be considered as the interface controlled growth mechanism. The effective activation energy, which equals to the growth activation energy in Equation (5) under the site saturation of nucleation condition, can be estimated in terms of the Kissinger-like method [6]. Figure 5 shows that the calculation describes the experimental data with sufficient accuracy almost in the whole

isothermal process. Because the degree of incomplete bainite transformation increases with increasing isothermal temperature, the apparent discrepancy occurs at the last stage of transformation for 500 °C isothermal temperature. However, this model is, admittedly, too simple to simulate the abnormal transformation kinetics, e.g. the kinetics curve of 540 °C isothermal temperature.

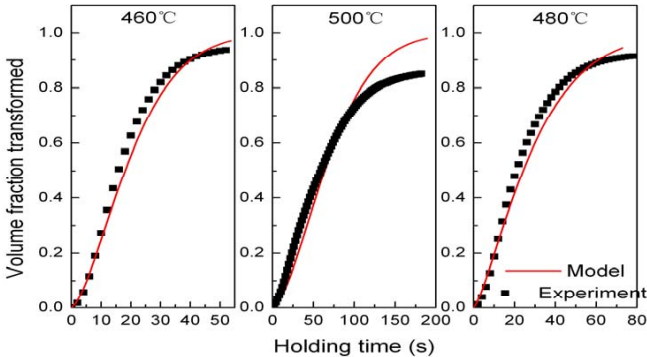


Figure 5. Comparison of experimental and calculated kinetics for isothermal bainite formation in the normal transformation temperature range

Conclusions

In the present study, the isothermal bainite transformation of a low carbon multi-microalloyed steel was carried out using a dilatometer. The main microstructure changed from lath bainite to widmanstätten ferrite with the increase in the isothermal temperature. The dilatation curve showed that two distinct kinetic curves can be found in the range of bainite formation temperature. One is that the curves show a one-stage transformation feature with the fast transformation rate; the other is that the curves present a two-stage transformation behavior. According to the general JMA mode, the normal transformation kinetic curves have the same Avrami exponent in the whole isothermal process. In contrast, the Avrami exponent changes with the holding time for the two-stage kinetic curves and the Avrami exponent abruptly lowers to zero at the beginning stage for this abnormal transformation kinetics curve. A physically-based model is proposed to describe the normal transformation kinetic behavior on the basis of the analysis of the nucleation and growth mode as well as the modification of the JMA model. The calculation curves are in a good agreement with the experimental curves.

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