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Exploring bainite formation kinetics distinguishing grain-boundary and autocatalytic nucleation in high and low-Si steels

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ABSTRACT

Bainite formation in steels begins with nucleation of bainitic ferrite at austenite grain boundaries (γ/γ interfaces). This leads to creation of bainitic ferrite/austenite interfaces (α/γ interfaces). Bainite formation continues through autocatalysis with nucleation of bainitic ferrite at these newly created α/γ interfaces. The displacive theory of bainite formation suggests that the formation of bainitic ferrite is accompanied by carbon enrichment of surrounding austenite. This carbon enrichment generally leads to carbide precipitation unless such a reaction is thermodynamically or kinetically unfavourable. Each bainitic ferrite nucleation event is governed by an activation energy. Depending upon the interface at which nucleation occurs, a specific activation energy would be related to a specific nucleation mechanism. On the basis of this concept, a model has been developed to understand the kinetics of bainite formation during isothermal treatments. This model is derived under the assumptions of displacive mechanism of bainite formation. The fitting parameters used in this model are physical entities related to nucleation and microstructural dimensions. The model is designed in such a way that the carbon redistribution during bainite formation is accounted for, leading to prediction of transformation kinetics both with and without of carbide precipitation during bainite formation. Furthermore, the model is validated using two different sets of kinetic data published in the literature.

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1. Introduction

Bainite was discovered as an ‘acicular, dark-etching aggregate’ nearly eight decades ago [1,2]. Over the years, the research work done in the field of bainite is immense [3–7]. Bainite consists of sheaves of bainitic ferrite separated by untransformed austenite, martensite or cementite. Each bainitic sheaf is composed of a cluster of sub-units which are connected in three dimensions [8]. Due to the complexity of its formation mechanism, even a qualitative theory to explain the bainite formation still remains a subject of controversy [4,9,10]. One “school of thought” advocates a diffusion-controlled transformation where bainitic growth occurs by a diffusional ‘ledge’ mechanism while the other suggests that the bainite reaction is a displacive and diffusionless transformation [4]. Both “schools” have proposed different models to predict the transformation kinetics based on their own assumptions of bainite formation [11–16].

Bainite formation begins at austenite grain boundaries. This bainite formation, at the initial stages of transformation, leads to an increase in the number density of nucleation sites. Bainite formation continues autocatalytically at these newly created nucleation sites. Santofimia et al. [17] reviewed and evaluated several kinetic models which are based on assumptions of displacive theory of bainite formation. Since displacive theory for bainite formation assumes that the rate of bainite formation is driven by the rate of bainitic ferrite nucleation, most of the models are based on nucleation kinetics. It is evident from the review in Ref. [17] that the overall structure for determining the rate of bainite formation is consistent among various models. Santofimia et al. [17] commented that the models mostly vary only in the manner in which the nucleation rate is calculated. With the help of their review, some of the major shortcomings of the existing models can be identified.

Most of the existing nucleation based models developed using the displacive mechanism of bainite formation use several empirical constants to account for the number density of grain-boundary nucleation sites and the number density of autocatalytic nucleation sites [17]. However, the physical significance of the values obtained for the empirical constants is still unclear [18]. Although some models describe the autocatalytic nucleation using other means,

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such as geometrical conception of the transformation [14] and with the help of prior austenite grain size [19], they still use empirical constants to calculate the grain-boundary nucleation kinetics.

Furthermore, in case of steels that are lean in silicon, these existing models fail to properly calculate of the maximum volume fraction of bainite formed during transformation [17]. According to the displacive theory of bainite transformation, the formation of bainitic ferrite from austenite is accompanied by a subsequent partitioning of carbon into surrounding austenite matrix [8]. This leads to a carbon enrichment of austenite during the course of the transformation. Depending on the chemical composition of the steel, the degree of carbon enrichment of austenite can vary. In lean-silicon steels, the degree of carbon enrichment is negligible due to the precipitation of carbides during bainite formation. Most existing models do not account for this variable degree of enrichment which sometimes results in an underestimation of the maximum volume fraction bainite. Such an underestimation will lead to an improper prediction of the bainite formation kinetics.

In order to tackle the problem of predicting the kinetics of isothermal bainite formation in lean-silicon steels, Van Bohemen and Sietsma [15] developed a kinetic model based on nucleation kinetics. This model was developed using the concepts of displacive theory of bainite and martensite formation. Unlike previous models where several empirical constants were used, Van Bohemen and Sietsma used physical parameters to calculate the number density of grain-boundary nucleation sites. Since this model does not predict the incomplete reaction phenomenon which is exhibited by high silicon steels, Van Bohemen and Hanlon [16] proposed a modified version of the Van Bohemen and Sietsma model [15] for this purpose.

However, both Van Bohemen and Sietsma model [15] and Van Bohemen and Hanlon model [16] do not account for the condition that diffusionless growth of bainite can occur only when the transformation temperature is below a certain thermodynamic limit as proposed by Ref. [20]. According to the displacive approach of the bainite transformation, it has been suggested that the displacive formation of bainite can proceed if and only if the following conditions,

$$\Delta G_m < G_N; \quad \text{where} \quad \Delta G_m = G_m^\alpha - G_m^\gamma \quad (1)$$

$$\Delta G^{\gamma \rightarrow \alpha} < -G_{SB}; \quad \text{where} \quad \Delta G^{\gamma \rightarrow \alpha} = G^\alpha - G^\gamma \quad (2)$$

are satisfied [20]. $\Delta G^{\gamma \rightarrow \alpha}$ represents the free energy change during bainite formation. G^α and G^γ give the ferrite free energy and austenite free energy respectively, when both the composition of ferrite and of austenite is equal to the composition of interest. ΔG_m is the maximum driving force for nucleation. It is the greatest possible reduction in free energy that can be achieved during formation of a ferrite nucleus such that the composition of surrounding austenite matrix remains unaffected. It is calculated using parallel tangent construction. G_m^α and G_m^γ give the ferrite free energy and austenite free energy when this condition of maximum free energy reduction is achieved. G_N is the universal nucleation function [3]. G_{SB} is the stored energy of bainite which is usually considered to be 400 J mol^{-1} [20]. Eq. (1) indicates that a bainite nucleus can develop only at temperatures where ΔG_m is more negative than G_N . The temperature at which $\Delta G_m = G_N$ is called the T_h temperature. Furthermore, a diffusionless growth of bainite can occur only if Eq. (2) is satisfied. The maximum temperature below which diffusionless growth of bainite can occur ($\Delta G^{\gamma \rightarrow \alpha} = -G_{SB}$) is called the T'_0 temperature [8]. Therefore, according to Eq. (1) and Eq. (2), bainite formation can occur only when the isothermal transformation temperature is below both T_h and T'_0 temperature.

Both T_h and T'_0 temperatures decrease with increasing carbon

enrichment of austenite during the course of bainite formation. Generally, it is observed that the T'_0 temperature decreases at a much faster rate with increasing carbon enrichment compared to the T_h temperature. When, during the transformation, either T_h or T'_0 temperature becomes equal to the transformation temperature, the bainite reaction will terminate and an incomplete reaction phenomenon will be exhibited. The nucleation rate at this point is equal to 0. Van Bohemen and Sietsma model and Van Bohemen and Hanlon model only consider the dependence of the nucleation rate on the T_h temperature and not on the T'_0 temperature. Its effects would not be significant in the prediction of bainite kinetics in lean silicon steels due to negligible effective carbon enrichment of austenite during transformation. However without such a dependence, in case of high silicon steels, the nucleation rate at the end of the transformation may not always reach 0. This implies that the model predicts further bainite formation which is physically unrealistic.

In this work, a unified model to predict the kinetics of isothermal bainite formation regardless of the degree of carbon enrichment of austenite is proposed. In an attempt to better treat the autocatalytic nucleation, a physically based approach considering the difference in the activation energy for grain-boundary nucleation and for autocatalytic nucleation is proposed here. The model is derived under the assumptions given by the displacive theory of bainite formation and draws inspiration from previously proposed models [15,21]. The model fitting parameters are used in such a way that its physical significance can be interpreted.

2. The model

2.1. Nucleation rate

Bainitic ferrite sub-units may nucleate either at austenite grain boundaries (γ/γ interface) or at the interphase boundary of a previously nucleated sub-unit (α/γ interface). The latter is interpreted as autocatalytic bainite nucleation [8]. The total nucleation rate during bainite formation from a fully austenitic phase, dN/dt , can be given as

$$\frac{dN}{dt} = \left(\frac{dN}{dt}\right)_G + \left(\frac{dN}{dt}\right)_A \quad (3)$$

where $(dN/dt)_G$ is the nucleation rate per unit volume due to nucleation at austenite grain boundaries and $(dN/dt)_A$ is the nucleation rate per unit volume due to autocatalytic nucleation.

It is generally accepted that bainite nucleation is a thermally activated process [3]. According to displacive theory of bainite formation, two types of atomic processes may require thermal activation [3,20,22,23]. Firstly, the mechanism of bainite nucleation involves dissociation of certain dislocation defects which are already present in the austenite phase. Secondly, in order to create the necessary driving force for nucleation, carbon must partition from the bainitic nucleus into the surrounding austenite matrix. Both these processes require thermal activation. The nucleation rate is usually expressed as an exponential function of the temperature [17]. Using this approach, the nucleation rate due to grain-boundary nucleation can be written as

$$\left(\frac{dN}{dt}\right)_G = \frac{kT}{h} N_{tG} \exp\left(-\frac{Q_G^*}{kT}\right) \quad (4)$$

where k is the Boltzmann's constant, h is the Planck's constant, N_{tG} is the number density of potential grain-boundary nucleation sites at given time t , Q_G^* is the activation energy for grain-boundary nucleation and T is the isothermal transformation temperature.

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