Modelling Phase Transformation Kinetics in Fe–Mn Alloys

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A unified model is proposed for the austenite-to-ferrite transformation kinetics in binary Fe–Mn alloys that accounts for solute drag of Mn. To aid the model development, continuous cooling transformation (CCT) tests were conducted for an interstitial-free steel that can be considered as Fe–0.1%Mn alloy. The experimental transformation data are supplemented with literature data for Fe–1%Mn and Fe–2%Mn alloys to establish a CCT database for Fe–Mn alloys. The austenite-to-ferrite transformation kinetics is described from a fundamental perspective by assuming an interface-controlled reaction and including solute drag of Mn. Using the solute drag model of Fazeli and Militzer, intrinsic interface mobility, trans-interface diffusivity of Mn and its binding energy have been determined from the CCT data. The interfacial parameters are critically analyzed and compared with independent measurements of diffusion and grain boundary segregation.

KEY WORDS: Fe–Mn alloy; phase transformation; interface mobility; solute drag.

1. Introduction

Alloying elements are added for a number of reasons to steels, primarily to attain the desired properties of the steel product or alternatively to facilitate processing of the steel. A variety of substitutional alloying elements are used in addition to carbon which occupies interstitial positions. Many of these alloying elements have a profound effect on the austenite-ferrite transformation and this is exploited as a key metallurgical tool to meet property requirements. Sometimes the term transformation hardening is used to address the role of alloying elements on phase transformations. An important alloying element that is employed in most steels is manganese. It plays an important role in steel making by forming MnS to take S out of solution thereby minimizing the otherwise harmful role of S, e.g. in terms of embrittlement. Further, Mn lowers the activity of carbon thereby effectively lowering the carbon concentration such that Mn-rich regions would tend to attract carbon. Mn is an austenite stabilizer and delays austenite decomposition, i.e. it acts as a transformation hardener. Mn also provides solute solution strengthening. Thus, increased Mn levels are used in advanced high strength steels to increase strength levels. To develop and optimize processing routes for these modern steels it is in particular important to precisely describe the effects of Mn on the phase transformation kinetics.

Austenite decomposition has been widely studied for C–Mn steels and Fe-based alloys and a number of phenomenological models have been proposed to account for the role of Mn.1,2) Nevertheless, it is still a challenge to achieve a full understanding of the mechanisms by which Mn and other alloying elements affect the transformation kinetics.3) The basic phenomenological observation is that increasing the Mn level increases the undercooling below the $Ae_1$ temperature to observe austenite-to-ferrite transformation during continuous cooling of practical relevance, e.g. cooling on the run-out table of a hot mill, where phase transformation takes place without long range redistribution of substitutional alloying elements such as Mn. Mn, however, may locally redistribute around the moving austenite-ferrite interface. There is some evidence of a Mn spike at the interface which is assumed to be the key phenomenon to explain the delaying effect of Mn on austenite decomposition.4) Recent dedicated studies by Zurob et al. on phase transformation during decarburization in Fe–C–Mn alloys indicated a particular complex behavior for Mn with a transition from paraequilibrium (substitutional sub-lattice frozen) at higher temperatures to non-partitioning local equilibrium (i.e. development of a Mn spike at the interface) at lower temperatures.5)

A number of phenomenological transformation models have been proposed. Due to the simplicity and numerical convenience, the Johnson-Mehl-Avrami-Kolmogorov (JMAK) approach is most commonly employed in industrial process models. In this approach, the effect of Mn on the phase transformation kinetics was incorporated by fitting the rate parameter to experimental data1,2) without explicitly describing the effect of Mn on the underlying nucleation and growth rates. Predictions of these models are only valid for the limited range of the experimentally investigated alloys and conditions.

Alternatively, a more rigorous mixed-mode model was developed and applied to low carbon steels.6) Here, both long range carbon diffusion and interface reaction (i.e. the rate of lattice change from FCC to BCC structure) are assumed to control the transformation rate. In this approach,
the interface mobility is typically employed as a fit parameter. Frequently, an Arrhenius relationship is assumed for its temperature dependence with an activation energy of the generally accepted value of 140 kJ/mol. Then the pre-exponential mobility term, $M_0$, is the only fit parameter. Using this model, Kop et al. simulated the transformation kinetics in three Fe–C–Mn alloys based on the assumption of tetrakaidecahedron shaped austenite grains and site saturation for nucleation. The $M_0$ was found to depend on the cooling rate in order to describe experimental CCT data. Mecożzi et al. and Militzer et al. have used phase field modelling to simulate the austenite-to-ferrite transformation in Fe–0.1 mass% C–0.49 mass% Mn. Also, their analysis revealed an apparent cooling rate dependence of $M_0$.

Clearly, introducing an effective interface mobility in this way does not lead to a satisfactory transformation model when different processing conditions are considered. The apparent deviations of the mobility term from an Arrhenius temperature dependence may be related to potential solute drag by Mn, i.e. Mn tends to segregate to the interface and when the interface moves its migration rate is slowed down by the presence of Mn in solution. Solute drag was first examined by Cahn to describe the solute interaction with moving grain boundaries. Purdy and Bréchet extended Cahn’s model to interphase boundaries moving with quasi-steady velocity. A number of alternative solute drag models have been proposed and applied to phase transformations in steels. Recently, Fazeli and Militzer used their solute drag model to describe the austenite-to-ferrite transformation in a 0.17 mass% C–0.74 mass% Mn steel. In their approach, in addition to $M_0$, the binding energy of the solute to the interface as well as its diffusivity across the interface were further adjustable parameters. A set of interfacial parameters was determined from isothermal transformation studies by taking an activation energy for the trans-interface diffusivity of Mn that is half of its activation energy for bulk diffusion in austenite as well as assuming paraequilibrium conditions and a spherical austenite grain with an outer shell of ferrite. The model developed in this way permitted to predict the continuous cooling transformation kinetics measured in this steel, i.e. without the need to further adjust parameters. However, in order to achieve this, a binding energy had to be introduced that strongly depends on temperature in contradiction to typical observations for binding energies to grain boundaries and other interfaces.

There were also attempts reported to incorporate the effect of Mn on the transformation kinetics directly into a carbon diffusion model by changing the contact conditions at the interface. Militzer et al. introduced a segregation factor which describes the steady-state enrichment of Mn at the moving interface. The increase of the Mn concentration at the interface decreases the interfacial carbon concentration in local equilibrium thereby reducing the calculated transformation rates. Recently, in the study of the ferrite growth kinetics in Fe–C–Mn alloys by Zurob et al., an interface capacity, $X^*_\text{tr}$, which determines the kinetic transition at the moving austenite-ferrite interface, has been introduced to enable the quantitative description of the experimental observations over a range of compositions and temperatures. The value of $X^*_\text{tr}$ depends on carbon content due to the strong C–Mn interaction.

Even though the Fe–C–Mn system is the base system of practical interest for most steels, the significant interaction of C and Mn adds complexity in analyzing the role of Mn on the phase transformation kinetics. Thus, the present paper revisits the austenite-to-ferrite transformation kinetics in Fe–Mn alloys. Krielaart and Van der Zwaag studied continuous cooling transformation in two Fe–Mn alloys containing 1 and 2 mass% Mn, respectively, using cooling rates of up to 20°C/min that are much slower than those employed industrially on a run-out table. They described the transformation kinetics with an interface-controlled transformation model by using an effective interfacial mobility as fit parameter. Militzer investigated the continuous cooling transformation in an interstitial free (IF) steel that can be viewed in a first approximation as a Fe–Mn alloy with a lower Mn content of 0.1 mass%. Experimental data were generated for cooling rates as high as 100°C/s and described by introducing an empirical temperature dependent Mn segregation factor. A drawback of this study was that the high cooling rate tests could not be performed with a constant cooling rate throughout the transformation region. Thus, in the present work, a new set of continuous cooling transformation (CCT) tests was first conducted for the IF steel (Fe–0.1 mass%Mn). Then, a unified transformation model is proposed to explicitly account for the effect of Mn on the austenite-to-ferrite transformation in all three Fe–Mn systems. The model is based on the solute drag approach of Fazeli and Militzer with the aim to establish interfacial parameters that are consistent with independent studies on the interaction of Mn with interfaces in Fe-alloys. The paper concludes with a critical analysis of the interfacial parameters inferred from the transformation data.

### 2. Experimental Investigations and Results

The experimental results for the binary Fe–1%Mn and Fe–2%Mn alloys were taken from the literature. To supplement these data, the austenite decomposition kinetics was investigated in an IF steel with a chemical composition as listed in Table 1. The steel is microalloyed with the strong carbonitride forming element Ti to scavenge carbon and nitrogen from interstitial solid solution. Therefore, the investigated steel can be considered as Fe–0.1%Mn alloy for transformation studies. Using ThermoCalc with the Fe2000 database, the $Ae_3$ temperature was determined to be 905°C.

| C  | Si  | Mn  | P  | S  | Nb  | Ti  | Al  | N  |
|----|-----|-----|----|----|-----|-----|-----|----|
| 0.002 | 0.01 | 0.11 | 0.01 | 0.008 | 0.009 | 0.059 | 0.033 | 0.0041 |

The specimens were applied and combined with resistive heating to avoid faster cooling than prescribed. The final microstructure was
revealed by Nital etching and consists of 100% polygonal ferrite. The ferrite grain size, $d_{\alpha}$, was quantified by the “Average Equivalent Area Diameter (EQAD) Measurement” according to ASTM E1382-97. The grain size decreases with cooling rate, i.e. 86, 38, 26 and 24 $\mu$m for 1, 10, 55, 100°C/s, respectively. The austenite-to-ferrite transformation kinetics was quantified from the dilation data by applying the lever rule. Figure 1 shows the experimental results indicating that the transformation is shifted to lower temperature as cooling rate is increased. Similar observations had been made for the alloys with higher Mn content. The ferrite grain sizes in these alloys were 97 $\mu$m and 143 $\mu$m for Fe–1%Mn and Fe–2%Mn, respectively, when cooled at 10–20°C/min.7)

### 3. Modelling of Phase Transformation

#### 3.1. Model Formulation

In Fe–Mn alloys, the austenite-ferrite transformation occurs without long range diffusion of Mn for the cooling rates employed in the CCT tests. Then, the reaction is interface controlled. Some further insight into nucleation conditions and growth geometries may be obtained by evaluating the observed transformation kinetics with the JMAK model. The lines in Fig. 1 provide the best fit using the JMAK model and the additivity rule. In the JMAK approach, the fraction transformed, $X$, during continuous cooling can be written such that

$$X = 1 - \exp\left\{-\frac{T}{q}\left[\frac{k(T)^{n}}{d_{\alpha}}\right]^{1/n}\right\} \quad \text{(1)}$$

where $T$ is the temperature, $T_{S}$ is the transformation start temperature, $q$ the constant cooling rate, $n$ is the JMAK exponent and $k$ is a temperature dependent rate parameter. Using a previously developed numerical approach22) $k$ and $n$ were determined for the Fe–0.1%Mn alloy as listed in Table 2. The lines in Fig. 1 provide the best fit indicating good agreement between calculated and measured transformation kinetics confirming the additive nature of the ferrite formation. Further, $n = 1$ may be taken as an indication that ferrite formation occurs under nucleation site saturation conditions with one dimensional growth geometry. The rate parameter $k$ is then given by the product of nuclei density, $N_{n}$, per unit length and the velocity, $v$, of the austenite-ferrite interface

$$\frac{dX}{dt} = N_{n}v(1-X) \quad \text{............... (2)}$$

Here, the nucleation density can be taken as the reciprocal of the ferrite grain size, i.e. $1/d_{\alpha}$.

Krielaart and Van der Zwaag7) had used an effective mobility approach to describe the interface velocity, *i.e.*

$$v = M_{ef} \Delta G_{chem} \quad \text{............... (3)}$$

where $M_{ef}$ is the effective interface mobility and $\Delta G_{chem}$ is the chemical driving pressure. For the Fe–1%Mn and Fe–2%Mn alloys a good fit was obtained with the experimental data using an activation energy of 140 kJ/mol for the effective mobility. For the 0.1%Mn alloy, however, a much lower activation energy of approximately 12 kJ/mol would be required. Thus, a more rigorous approach to include solute drag is proposed as an alternative, *i.e.*

$$v = M(\Delta G_{chem} - \Delta G_{SD}) \quad \text{............... (4)}$$

where $M$ is the intrinsic interface mobility and $\Delta G_{SD}$ is the solute drag pressure. Assuming Mn atoms remain configurationally frozen during the transformation process, $\Delta G_{chem}$ is determined as the Gibbs free energy difference between austenite and ferrite of the nominal alloy composition using ThermoCalc with the Fe2000 database. To calculate the solute drag pressure, the model proposed by Fazeli and Militzer17) is employed. In the solute drag model, the segregated solute profile $c_{s}$ across the interface is determined. It is useful to introduce normalized variables for concentration, distance and velocity, *i.e.* $C = C_{s}/C_{S0}$, $X = x/d$ and $V = vD_{b}$, respectively. Here, $C_{S0}$ is the bulk solute content, $x$ is the position in the interface, $d$ is half of the interface thickness and $D_{b}$ is the trans-interface diffusivity of the solute. An asymmetrical wedge-shaped well is assumed for the interaction potential, $E$, of the solute with the interface. The potential gradient $\partial E/\partial X$ is given by

$$\left\{\frac{\partial E}{\partial X}\right\}_{new} = \Delta E - E_{0} \quad \text{............... (6)}$$

where $E_{0}$ is the solute-interface binding energy and $\Delta E$ is half of the chemical potential difference for the solute in austenite and ferrite. Physically, the binding energy is negative to be consistent with lowering the free energy for attractive solute-interface interaction. For convenience, binding energies are given as positive numbers here to characterize the strength of the solute-interface interaction. Fazeli and Militzer17) introduced effective concentration and potential terms, *i.e.*

$$C_{new} = C \exp\left(\frac{\Delta E}{RT} X\right)$$

Table 2. JMAK parameters for Fe–0.1%Mn.

| $A$ (°C$^{-1}$) | $T_{S}$ (°C) | $B$ | $n$ |
|----|----|----|----|
| $-1.0 \times 10^{-3}$ | 823.3 | 1.9 | 1 |

$\frac{\partial E}{\partial X}_{new} = \frac{\partial E}{\partial X} - \Delta E \quad \text{............... (6)}$
to avoid the artifact of residual solute drag at zero interface velocity. Then the segregation profile can be obtained by solving the following equation

\[
\frac{\partial C_{\text{new}}}{\partial X} + \frac{C_{\text{new}} \partial E}{RT \partial X_{\text{new}}} + V \left( C_{\text{new}} - \exp \left( \frac{\Delta E}{RT} \right) \right) X = 0 \quad \ldots (7)
\]

Here, \( R \) is the gas constant and \( T \) is the temperature. By integrating the contribution from all the segregated solute atoms at the interface, the solute drag pressure is calculated as

\[
\Delta G_{\text{SD}} = -C_{5.0} \left( \frac{\partial E}{\partial X_{\text{new}}} \right) dX \quad \ldots (8)
\]

### 3.2. Model Application

Intrinsic mobility, binding energy and trans-interfacial diffusivity are the parameters in the proposed model that have to be adjusted in a consistent way to describe the transformation kinetics. For both mobility, \( M \), and diffusivity, \( D_b \), an Arrhenius temperature relationship is assumed, i.e.

\[
M = M_0 \exp \left( \frac{-Q}{RT} \right) \quad \ldots (9)
\]

and

\[
\frac{D_b}{\delta} = D_0 \delta \exp \left( \frac{-Q_b}{RT} \right) \quad \ldots (10)
\]

where the diffusivity is expressed as the normalizing factor for the interface velocity. Here, \( M_0 \) and \( D_0 \) are the pre-exponential factors and \( Q \) and \( Q_b \) are the activation energies. The pre-exponential factor for the diffusivity has been chosen as the geometric mean of the values for Mn bulk diffusion in ferrite\(^{23}\) and austenite.\(^{24}\) By taking an interface thickness of 1 nm, \( D_0/\delta \) is calculated to be 1.67 cm/s. The activation energy, \( Q_b \), is employed as an adjustable parameter. For the mobility, the activation energy is pre-set and \( M_0 \) is used as an adjustable parameter. In addition to the well accepted value of 140 kJ/mol for \( Q \), an alternative value of 16 kJ/mol\(^{25}\) was considered which was determined from molecular dynamics simulations. This value is similar to that concluded from the effective mobility fit for the Fe–0.1%Mn alloy. Finally, the binding energy, \( E_0 \), is another fit parameter.

Table 3 shows the two sets of model parameters (\( M_0, Q_b \), and \( E_0 \)) determined from the simulation assuming the two different values for \( Q \). Figures 2 and 3 show the corresponding comparison between the measured and calculated transformation kinetics. In general, reasonable agreement is achieved independently of the pre-selected \( Q \). The binding energy is the only fit parameter that depends on alloy composition. Thus, further criteria have to be considered to evaluate the merit of the fit parameter sets.

### 4. Discussion

The values obtained for the solute drag parameters depend critically on the selected intrinsic mobility. By having a larger activation energy \( Q \), the temperature dependence of the intrinsic mobility becomes steeper. For the applied range of cooling rates, most of the transformation

| Parameter Set | \( Q \) (kJ/mol) | \( M_0 \) (cm mol/Js) | \( Q_b \) (kJ/mol) | \( E_0 \) (kJ/mol) |
|---------------|-----------------|----------------------|------------------|------------------|
| Fe–0.1%Mn     | 16              | \( 6 \times 10^{-4} \) | 206              | 6                |
| Fe–1%Mn       | 140             | 450                  | 216              | 16               |
| Fe–2%Mn       | 140             | 450                  | 216              | 11               |

Fig. 2. Comparison between measured (symbols) and calculated (solid line) ferrite fractions using parameter set I.
takes place at higher temperature (>840°C) in Fe–0.1%Mn, while the transformation happens at lower temperature (<760°C) in Fe–2%Mn. Therefore, compared with the first set of \( M_0 \) and \( Q \) in Table 3, the second set would comparatively increase and decrease the intrinsic mobility for Fe–0.1%Mn and Fe–2%Mn, respectively. As a result, the alloy with the lower Mn content requires a comparatively larger solute drag term as indicated by the larger binding energy. The magnitude of solute drag increases with binding energy as shown in Fig. 4(a).

Further, the solute drag pressure is a function of interface velocity that is normalized by \( D_0/\delta \). Solute drag is maximized for a normalized velocity of approximately 1.8. Thus, the value of the trans-interfacial activation energy \( Q_b \) determines the temperature range where solute drag is significant, as shown in Fig. 4(b). Here, the solute drag pressure gradually increases with decreasing temperature to reach its maximum at the end of transformation for an activation energy of 190 kJ/mol, whereas for 216 kJ/mol the maximum of solute drag pressure is attained at approximately 850°C, i.e. at much earlier transformation stages.

The values obtained in the present analysis for \( Q_b \) are similar in both data sets. They are smaller than the activation energies for bulk diffusion in austenite and ferrite, respectively. In detail, they amount to 88% and 92%, respectively, of the value for paramagnetic ferrite. This can be considered as very reasonable for trans-interface diffusion. Using \( Q_b \) rather than \( D_0/\delta \) as adjustable parameter for the trans-interface diffusivity has also produced binding energies that are independent of temperature. In contrast, strongly temperature dependent binding energies were reported for the Fe–C–Mn system by Fazeli and Militzer\(^{17} \) who assumed \( Q_b \) and fitted \( D_0/\delta \). Pre-scribing \( Q_b \) may thus have led to the unrealistic temperature dependence of the binding energies in this previous analysis.

The magnitude of the calculated binding energies in the present study is comparable with experimental measurements of Mn segregation on austenite grain boundaries (8 ± 3 kJ/mol\(^{18} \)) and ferrite grain boundaries (5.5 kJ/mol).\(^{19} \) The trend of the binding energy with Mn content, however, is
very different in both model sets. In set I the alloy with the intermediate Mn level has the highest binding energy whereas in set II the binding energy increases with decreasing Mn alloying. The latter relationship between binding energy and bulk Mn content appears to be much more realistic and can be rationalized as follows. In a first approximation, the binding energy may be considered to be the same when a Mn atom segregates to any site in the interface. However, in detail there may be preferred sites in the interface with a higher binding energy.26) When Mn atoms segregate to the interface, they first will occupy favorable sites. Increasing the Mn concentration in the interface, only less favorable sites remain available. Their lower binding energy leads to a decrease of the apparent average binding energy. In order to compare the segregation levels in the interface for the three Fe–Mn alloys, the maximum equilibrium Mn segregation content is calculated, i.e.

$$C_{\text{max}} = C_{i,0} \exp \left( \frac{E_0 - \Delta E}{RT} \right)$$ \hspace{1cm} (11)

which is the concentration at position $x = 0$ for the interfacial concentration profile in the limit of zero interface velocity, i.e. for a stationary interface. Using the binding energies of parameter set II in Table 3, the temperature dependence of the maximum Mn segregation level is shown for the three alloys in Fig. 5. The maximum interfacial Mn content increases with the bulk Mn content even though the apparent average binding energy increases with decreasing Mn content. This behavior is consistent with the proposed assumption of different interfacial sites.

5. Conclusion

A detailed analysis of solute drag of Mn has been conducted by revisiting the austenite-to-ferrite transformation kinetics during continuous cooling of Fe–Mn alloys. A consistent description of the effect of Mn on delaying the phase transformation is obtained with an activation energy of 216 kJ/mol for Mn diffusion across the interface and binding energies of Mn to the interface that decrease with increasing Mn content from 16 to 6 kJ/mol. The magnitude of the obtained values for the activation energy and binding energies is in the range expected from independent studies on diffusion and Mn grain boundary segregation. The activation energy amounts to 92% of that for Mn bulk diffusion in paramagnetic BCC-iron indicating fast diffusion in the interface region. The effect of Mn concentration on the binding energies can be rationalized with the assumption of having preferred interfacial sites with higher binding energies such that the apparent average binding energy decreases as more and more sites are occupied. To further evaluate these phenomenological findings it would be useful to conduct atomistic simulations of the Mn interaction with the austenite-ferrite interface in iron. For example, first principle density functional theory simulations can be used to determine binding and activation energies at different interface sites.

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