Modelling Upper and Lower Bainite Transformation in Steels

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Bainite is of considerable importance in the design of high strength steels. There are two types of morphologies, upper and lower bainite. In upper bainite, cementite forms between adjacent bainitic ferrite plates. In certain steels, however, the cementite reaction is suppressed so that carbon-enriched austenite remains untransformed between bainitic ferrite plates. In lower bainite, cementite also has the opportunity to precipitate within bainitic ferrite plates. In order to model the development of these microstructures, it is necessary to treat the simultaneous formation of both the ferritic and carbide components of the microstructure. A theory has been developed to do exactly this, enabling the estimation of the phase fractions, the cementite particle size and the transition from upper to lower bainite. The results have been compared against experimental data.

KEY WORDS: simultaneous reaction; upper/lower bainite; bainitic ferrite; cementite; austenite.

1. Introduction

High strength steels are used widely as structural materials. Their microstructures frequently contain bainite. A number of models for the evolution of bainitic microstructures have been developed1–5) but they either neglect the precipitation of cementite, or treat the whole event as a single reaction. It is in principle necessary to permit the simultaneous formation of cementite and bainitic ferrite, albeit at different rates, in order to properly deal with upper and lower bainitic microstructures. The latter is distinguished from the former by the fact that some cementite is found within the bainitic ferrite plates, whereas in upper bainite the carbides only precipitate between the plates.

The purpose of the present work was to develop a full kinetic theory for the bainite reaction and to compare the predicted phase fractions, particle sizes and the nature of the bainite against experimental data.

2. Experimental Procedure

Table 1 shows the chemical composition of steel, which with its high carbon concentration should be amenable to the upper to lower bainite transition. The steel was melted using vacuum induction heating followed by casting. The ingot was re-heated to 1200°C and hot-rolled. The sheets were homogenized at 1250°C for 3 d and cooled rapidly with air, resulting in a martensitic microstructure which proved difficult to machine. The samples were therefore austenitised at 850°C, cooled to 600°C and held for 3000 s in order to generate softer pearlite which could easily be machined into cylindrical specimens of 10 mm length and 3 mm diameter. These samples were heated in a dilatometer up to 1000°C and cooled at 50°C/s with helium gas to the temperatures between 300 and 450°C for isothermal transformation to bainite. The austenite grain size was 60 μm after reaustenitisation at 1000°C. Specimens were etched using 2 vol% nital (nitric acid in methanol) and the microstructures were observed using optical, scanning and transmission electron microscopy. Precipitates were characterized in a transmission electron microscope (TEM) using X-ray energy-dispersive analysis and electron diffraction. Cementite particle sizes were measured directly from TEM micrographs of the carbon replicas. At least 100 particles were analyzed in each case. The carbide shape was found to be in the form of discs whose thickness and diameters were measured.

3. Modelling

Figure 1 illustrates the essence of the model, which consists of four processes:

1) Diffusionless transformation of austenite into supersaturated bainite.
2) The escape of carbon evacuation from bainitic ferrite into austenite.
3) Cementite precipitation in bainitic ferrite.
4) Cementite precipitation in carbon-enriched residual austenite.

These reactions can be handled simultaneously using the framework developed by Bhadeshia et al.6–8) Extended vol-

Table 1. The chemical composition (mass%).

<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.6</td>
<td>1.5</td>
<td>1.5</td>
</tr>
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</table>
ume corrections were made for the major phases (austenite and bainitic ferrite) but were neglected for the cementite which has a small overall fraction. The resulting numerical scheme allowed the matrix composition to be modified at each stage using the mean-field approximation.

### 3.1. Bainitic Ferrite

The first stage in the transformation is the nucleation and growth of laths or plates of ferrite in the form of arrays of sub-units. Consistent with experimental evidence, it is assumed that nucleation occurs with the diffusion of carbon, and that each plate of bainite also generates other nuclei by autocatalysis. The nucleation rate \( I_{ab} \) is then given by:

\[
I_{ab} = (1 + \beta V_{ab}) I_0  
\]

where \( V_{ab} \) is volume fraction of bainitic ferrite and \( \beta \) is autocatalysis constant, indicating that the nucleation sites increase in compliance with sub-unit formation. The initial nucleation rate \( I_0 \) is by classical nucleation theory:

\[
I_0 = N_0 \frac{RT}{h} \exp\left(-\frac{Q_C^B}{RT}\right) \exp\left(-\frac{\Delta G^{\gamma\to\alpha B}}{RT}\right)  
\]

where \( N_0 \) is the initial site density, \( R \) and \( h \) are gas and Planck constants respectively and \( T \) is temperature in K. \( Q_C^B \) is the activation energy for carbon diffusion in austenite because the nucleation process has been reported to involve the diffusion of carbon. \( \Delta G^{\gamma\to\alpha B} \) is the driving force for bainitic ferrite formation. It is given by:

\[
\Delta G^{\gamma\to\alpha B} = \Delta G^{\gamma\to\alpha - G_N}  
\]

\[
\Delta G^{\gamma\to\alpha - G_N} = \frac{16}{3} \pi \frac{\sigma^2}{\Delta G_C^{\gamma\to\alpha}}  
\]

\[
G_N = 3.637 \times (T-273.18) - 2.540  
\]

where \( \Delta G^{\gamma\to\alpha} \) is the driving force for ferrite nucleation, \( \Delta G^{\gamma\to\alpha - G_N} \) is the chemical free energy change per unit volume for ferrite nucleation under para-equilibrium, \( \sigma \) is the interfacial energy per unit area. \( G_N \) is the critical energy change to form bainitic ferrite nucleus, which is as a function of temperature.

The transformation exhibits an incomplete reaction in which growth is arrested well before the austenite achieves its para-equilibrium composition, consistent with diffusionless growth. It is therefore reasonable to assume that once nucleated, the growth rapidly to a finite volume. The sub-unit size is as a function of temperature, austenite strength and driving force for bainitic ferrite formation under para-equilibrium. The width of sub-unit \( W_{ab} \), of which unit is \( \mu m \), is given by:

\[
W_{ab} = f(T, S\gamma, \Delta G^{\gamma\to\alpha}) = 0.478 + 1.20 \times 10^{-4}T + 1.25 \times 10^{-4}T - \Delta G^{\gamma\to\alpha} - 2.20 \times 10^{-3}S\gamma  
\]

where \( S\gamma \) is the strength of austenite in MPa, which has also been obtained empirically. However, the original equation doesn’t include Si effect on the strength, so it was added:

\[
S\gamma = (1 - 0.26 \times 10^{-2}(T - 298) + 0.47 \times 10^{-5}(T - 298)0.326 \times 10^{-8}(T - 298)^3 \times 15.4 \times (3.6 + 23W_i + 1.3W_S + 0.65W_Mb)  
\]

where \( W_i \) is mass% of \( i \)-th alloying element. It is assumed that the aspect ratio of sub-unit is constant in the temperatures between 300 and 500°C. The value is obtained experimentally to be six so that the length of sub-unit \( L_{ab} \) is given by:

\[
L_{ab} = 6W_{ab}  
\]

### 3.2. Carbon Evacuation From Bainitic Ferrite to Austenite

Since the initial growth event is diffusionless, the excess carbon in the ferrite is expected to partition into the residual austenite or precipitate as cementite. Since carbon diffusion in austenite is slower than in ferrite, the former is assumed to control the partitioning. Figure 2 illustrates the carbon concentration profile at the interface between austenite (\( \gamma \)) and bainitic ferrite (\( \alpha B \)).

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**Fig. 1.** Schematic illustration of upper and lower bainite microstructure development.

**Fig. 2.** Schematic illustration of the carbon concentration profile at austenite (\( \gamma \))/bainitic ferrite (\( \alpha B \)) interface.
3.3. Cementite Precipitation in Bainitic Ferrite

This model simultaneously deals with several reactions such as cementite precipitation both within bainitic ferrite and in austenite, of which nucleation sites depend on the chemical composition and heat treatment conditions. In lower bainite, cementite precipitates within bainitic ferrite. Lower bainite tends to be found at low temperatures where substitutional elements do not diffuse.\(^{16,17}\) It is assumed that cementite precipitates under para-equilibrium. Nucleation is treated by classical nucleation theory.\(^{10,11}\) The nucleation rate \(I_{\theta_{\text{in}}}\) is then, given by:

\[
I_{\theta_{\text{in}}} = N_0 B_{\text{in}} \frac{RT}{h} \exp \left( -\frac{Q_\alpha^a}{RT} \right) \exp \left( -\frac{-\Delta G^{\alpha_{\theta_{\text{in}}},\theta}}{RT} \right) \\
\tag{12}
\]

where \(Q_\alpha^a\) is the activation energy for carbon diffusion in ferrite. \(-\Delta G^{\alpha_{\theta_{\text{in}}},\theta}\) is the driving force for cementite nucleation under para-equilibrium condition.

As indicated in Fig. 3, the shape of cementite particles is experimentally found to be disc-like. The thickening rate can be assumed to be parabolic, so the thickness \(W_{\theta_{\text{in}}}\) is given by\(^{18}\):

\[
W_{\theta_{\text{in}}} = \frac{2}{D_\alpha^\alpha} \left( \frac{X_{\text{C}} - X_{\text{C}}^a} {X_{\text{C}}^a - X_{\text{C}}^a} \right) t \\
\tag{13}
\]

where \(X_{\text{C}}^a\) is carbon concentration in bainitic ferrite. \(X_{\text{C}}^a\) and \(X_{\text{C}}^\alpha\) are the para-equilibrium carbon concentrations at the bainitic ferrite/cementite interface. The aspect ratio is assumed to be constant value of five from TEM observations. The diameter \(L_{\theta_{\text{in}}}\) is, then, given by:

\[
L_{\theta_{\text{in}}} = 5W_{\theta_{\text{in}}} \\
\tag{14}
\]

It is reported that cementite within bainitic ferrite has orientation relationship with bainitic ferrite.\(^{19,20}\) This is caused by decreasing the interfacial energy, which is dealt as one of fitting parameters in this model.

3.4. Cementite Precipitation in Austenite

In upper bainite, cementite grows by a para-equilibrium mechanism from the carbon-enriched residual austenite.\(^{16,17}\) Cementite precipitation is modelled by the same way as from supersaturated bainitic ferrite as in the previous section.

4. Experimental Results

The micrographs of the specimens heat-treated at 300°C and 450°C for 1 000 s are presented in Fig. 3. At 450°C, the microstructure consists of bainite and retained austenite (γR) and cementite precipitates between adjacent two bainitic ferrite plates, which is a typical upper bainite microstructure. At 300°C, the microstructure also consists of bainite and retained austenite but cementite precipitates within bainitic ferrite plates which is lower bainite microstructure.

Figure 4 shows TEM micrographs of cementite particle analyzed (a), diffraction pattern from the particle (b) and EDX analysis for the particle (c) for the carbon replica specimens held at 450°C for 1 000 s respectively. As indicated in Fig. 4(c), cementite particle contains Si and Mn. From EDX analysis, Si/Fe and Mn/Fe ratio in cementite are calculated by the integration of energy dispersion for each element and listed in Table 2. Si/Fe and Mn/Fe ratios in cementite are essentially equal to those in austenite. This remains the case for the 450°C heat-treatment, justifying the assumption of para-equilibrium precipitation.

5. Comparison with Experiments

5.1. Parameters for Calculations

Thermodynamic parameters, such as driving forces for each nucleation, have been obtained using Thermo-Calc\(^ {21}\). The values of diffusion coefficients and activation energies for carbon in both ferrite and austenite listed in Table 3\(^ {22}\) were used for the calculations.

There remain two unknown parameters, the initial nucleation site density \(N_0\) and the interfacial energy \(\sigma\) for each phase. \(N_0\) may be deeply related with grain size and dislocation density, which may work as nucleation site. \(\sigma\) should depend on coherency. However, there may be difficulties to obtain physically reliable values of these parameters. For simplicity, they were treated as fitting parameters with constant values during whole event in this model. Figure 5 shows calculated changes in volume fractions of bainitic ferrite (αB) and cementite (θ) with site density of bainitic ferrite \(N_0^a\) and interfacial energy between αB and austenite (γ)\(\sigma\) for kinetics at 450°C, where upper bainite is expected (Fig. 3). As \(N_0^a\) decreases or \(\sigma\) increases, the overall rate of reaction is reduced. Figures 6 and 7 show calculated changes in volume fractions of αB and θ and in
cementite particle sizes in $\gamma$ with site density of cementite $N_0^{\gamma}$ and interfacial energy between $\theta$ and $\gamma$ $\sigma^{\theta\gamma}$ for kinetics at 450°C respectively. Smaller values of $N_0^{\gamma}$ or bigger values of $\sigma^{\theta\gamma}$ retard the formations of $\alpha B$ and $\theta$, especially after the stasis, and lead to bigger mean sizes of cementite in $\gamma$. Figures 8 and 9 show calculated changes in volume fractions of $\alpha B$ and $\theta$ and in cementite particle sizes with site density of cementite $N_0^{\alpha B}$ and interfacial energy between $\theta$ and $\alpha B$ $\sigma^{\theta\alpha B}$ for kinetics at 300°C, where lower bainite is expected (Fig. 3), respectively. The trends of kinetics changes with $N_0^{\alpha B}$ and $\sigma^{\theta\alpha B}$ at 300°C are similar to those at 450°C while cementite mainly forms in $\alpha B$.

A set of $N_0$ and $\sigma$ listed in Table 4 has been chosen and calculations were carried out, comparing with experiments.

## 5.2. Volume Fraction Changes and Transition Between Upper and Lower Bainite

Volume fraction changes for each phase during isothermal heat treatment at 450, 400 and 300°C are presented in Figs. 10, 11 and 12 respectively.

At 450 and 400°C (Figs. 10 and 11), cementite doesn’t precipitate in bainitic ferrite, but mainly in austenite, which is typical of upper bainite. This is consistent with the TEM observation shown in Fig. 3(b). Calculated volume fraction changes of bainitic ferrite, indicated by solid line, are in good agreement with experiments using dilatometry. Because cementite forms slower at 400°C than 450°C, transformation stasis, which is typical phenomena of bainite transformation, is observed more clearly at 400°C than 450°C.
As shown in Fig. 12, it takes longer for cementite to precipitate during transformation at 300°C, which results in a lower bainite microstructure, consistent with Fig. 3(d). Calculated volume fraction change of bainitic ferrite is also in good agreement with experiments. The model seems to give the correct trends for the simultaneous reactions, including transition between upper and lower bainite.

5.3. Cementite Particle Size

As shown in Fig. 12, it takes longer for cementite to precipitate during transformation at 300°C, which results in a lower bainite microstructure, consistent with Fig. 3(d). Calculated volume fraction change of bainitic ferrite is also in good agreement with experiments.

The model seems to give the correct trends for the simultaneous reactions, including transition between upper and lower bainite.

Fig. 5. Effects of initial nucleation site density of bainitic ferrite \( N_{0B}^{B} \) (a) and interfacial energy between bainitic ferrite (\( \alpha B \)) and austenite (\( \gamma \)) \( \sigma_{B}^{B/\gamma} \) (b) on volume fraction changes of \( \alpha B \) and cementite (\( \theta \)) in austenite as a function of holding time at 450°C.

Fig. 6. Effects of initial nucleation site density of cementite \( N_{0q}^{q} / H_{11002} \) (a) and interfacial energy between cementite (\( q \)) and austenite (\( \gamma \)) \( \sigma_{q}^{q/\gamma} \) (b) on volume fraction changes of \( \alpha B \) and \( \theta \) in austenite as a function of holding time at 450°C.
tation.

Figure 14 shows the diameter change of cementite particle in bainitic ferrite with holding time at 300°C. In the early stage of isothermal holding, both volume fraction of bainitic ferrite and number of cementite particles are so small that it may be difficult to detect certain number of cementite particles experimentally. There are therefore only experiments during prolonged holding stage, where the calculated size of cementite in ferrite is in reasonable agreement with experiments.

6. Conclusions

A complete bainite kinetics theory which is consistent with the mechanism of transformation has been established. The model can deal simultaneously with several reactions listed below.

1) Bainitic ferrite formation in austenite.
2) The escape of carbon evacuation from bainitic ferrite to austenite.

3) Cementite precipitation in bainitic ferrite.

4) Cementite precipitation in austenite.

The calculated volume fraction changes of bainitic ferrite and cementite particle size are in good agreement with the experiments. Furthermore, development of both upper and lower bainite microstructures can be simulated using the model.

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